Computational Aspects of Data Assimilation for Aerosol Dynamics*

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Abstract. In this paper we discuss the algorithmic tools needed for data assimilation for aerosol dynamics. Continuous and discrete adjoints of the aerosol dynamic equation are considered, as well as sensitivity coefficients with respect to the coagulation kernel, the growth rate, and emission and deposition coefficients. Numerical experiments performed in the twin experiment framework for a single component model problem show that initial distributions and the dynamic parameters can be recovered from time series of observations of particle size distributions.

1 Introduction

As our fundamental understanding of atmospheric particles and their transformations advances, novel computational tools are needed to integrate observational data and models together to provide the best, physically consistent estimate of the evolving state of the atmosphere. Such an analysis state better defines the spatial and temporal fields of key gas and particle phase chemical components in relation to their sources and sinks. Assimilation of chemical information is only now beginning in air quality/chemistry arenas [1,7,8,10,11], but offers the same motivations as those realized in the field of meteorology. Assimilation techniques can be utilized to produce three-dimensional, time varying optimal representations of the particle distributions in the atmosphere, that are consistent with the observed physical and chemical states.

Forward modeling of aerosols predicts the evolution of particle size distributions given the known parameters of the evolution (coagulation kernel, growth rate, emission rates and deposition velocities) as well as the initial size distribution. Numerous numerical methods have been proposed in the literature for

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solving particle dynamics, e.g. [4,6] etc. The *inverse modeling* problem consists of recovering the initial or emitted size distribution and the parameters of evolution given information about the time evolution of the system, for example periodic measurements of the number, surface, mass, or volume density.

In this paper we discuss the computational tools needed for inverse modeling of aerosol dynamics, including continuous and discrete adjoints of the dynamic equation are derived, as well as sensitivity coefficients with respect to the coagulation kernel, the growth rate, and emission and deposition coefficients. A detailed derivation is given in [12,13]. At this time we do not treat chemical and thermodynamic transformations, and consider only space independent (box) models. The methods discussed here are a first step toward the goal of performing data assimilation for comprehensive three-dimensional particle, chemistry, and transport models.

2 Forward Modeling of Particle Dynamics

2.1 Continuous Formulation

We consider the particle dynamic equation in number density formulation, with particle volume the independent variable. 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 $n(v,t)dv$. This describes completely a population of single-component particles. The aerosol population undergoes physical and chemical transformations (Seinfeld and Pandis, [14]) which change the number density in time:

$$\begin{aligned}\frac{\partial n(v,t)}{\partial t} &= -\frac{\partial}{\partial v}\left[I(v,t)\, n(v,t)\right] \\ &\quad +\frac{1}{2}\int_0^v \beta_{v-v',v'}\; n(v',t)\; n(v-v',t)\, dv' \\ &\quad -n(v,t)\int_0^\infty \beta_{v,v'}\; n(v',t)dv' + S(v,t) - L(v,t)\, n(v,t)\;, \\ n(v,t=t^o) &= n^o(v)\;, \quad n(v=0,t) = 0\;.\end{aligned} \tag{1}$$

In this equation $I(v,t) = dv/dt$ is the rate of particle growth (e.g., due to condensation, evaporation, deposition and sublimation), $\beta_{v,v'}$ is the coagulation kernel, $S(v,t)$ is any source of particles of volume v (e.g., nucleation and emissions) and $L(v,t)$ is the first-order rate of removal of particles of volume v (e.g., by deposition).

2.2 Discrete Formulation

Here we use a discrete formulation of (1) based on the piecewise polynomial approach as described by Sandu [9] and (for simplicity of the presentation) on the Forward Euler time stepping scheme. The finite dimensional approximation of

the number distribution $n(v,t)$ is taken in the space spanned by the set $\{\phi_i\}_{1\le i\le s}$ of piecewise polynomial basis functions. The equation (1) is discretized as

$$n^{k+1} = n^k + \Delta t \left\{ G(t^k) n^k + F(n^k) + A^{-1}\, B\left(n^k, n^k\right) + S(t^k) - L(t^k) n^k \right\}. \quad (2)$$

The growth terms are discretized using a Discontinuous Galerkin approach where $F(n^k)$ is the Godunov flux difference and

$$G_{i,j}(t) = \int_0^\infty I(v,t)\phi_i(v)\phi_j^{'}(v)\, dv\ .$$

The coagulation terms are discretized using a collocation approach [9] with collocation points $\{c_j\}_{1\le i\le s}$, such that

$$A_{i,j} = \phi_j(c_i)\ , \quad \{B(n,n)\}_i = n^T\ B^i\ n\ ,$$
$$B^j_{i,k} = \frac{1}{2}\int_0^{c_j} \beta_{v,c_j-v}\phi_i(c_j)\phi_k(c_j-v)\, dv - \int_0^\infty \beta_{v,c_j-v}\phi_i(v)\phi_k(c_j-v)\, dv\ .$$

3 The Inverse Modeling Problem

Variational methods (e.g., 4D-Var) provide an optimal control approach to the data assimilation problem. The optimal analysis state is obtained through a minimization process to provide the best fit to the background estimate and to all observational data available in the assimilation window.

To be specific assume that observations $y^1 \ldots y^N$ are available at the discrete times $t^1 \ldots t^N$. The observed quantities are functions of the model state, $y(t) = h\big(n(v,t)\big)$. Observations can be particle number or mass densities, optical properties etc. The initial value of the solution n^o and/or the problem parameters p (i.e. I, β, S and L) are not known accurately, and data assimilation uses the information from observations to better estimate them.

A cost functional that measures the mismatch between model predictions and observations is

$$\mathcal{J}(n^o,p) = \frac{1}{2}\ (n^o - n^o_B)^T\ R_0^{-1}\ (n^o - n^o_B) + \frac{1}{2}\ (p-p_B)^T\ B^{-1}\ (p-p_B) \quad (3)$$
$$+\frac{1}{2}\sum_{k=1}^{N} \left(y^k - h\big(n^k\big)\right)^T\ R_k^{-1}\ \left(y^k - h\big(n^k\big)\right)\ .$$

The first two terms penalize the departure of the initial solution and parameters from the apriori estimates (n^o_B and p_B), and are called background terms. The matrices R_0 and B are error covariances associated with background terms and R_k are error covariances associated with observations. The parameters n^o and p, which initially have values n^o_B and p_B, are adjusted such that the value of the functional is minimized. The minimization procedure needs the value of $\mathcal{J}$ and of its gradients with respect to the model parameters $\nabla_{n^o}\mathcal{J}$ and $\nabla_p\mathcal{J}$. For given values of (n^o,p) the functional value is obtained by a forward integration of the forward model, while its derivatives can be efficiently obtained through adjoint modeling as explained below.

3.1 Continuous Adjoints of the Dynamic Equation

In the continuous formulation one derives the adjoint of the exact dynamic equation (1), to obtain the derivative of the exact solution. The *adjoint equation* of the tangent linear model of (1) for the cost functional (3) is defined on each time interval (t^{k-1}, t^k) between consecutive observations for $k = N, N-1, \ldots, 1$ by

$$\begin{aligned}
\frac{\partial \lambda(v,t)}{\partial t} &= -\frac{\partial \lambda(v,t)}{\partial v} I(v,t) - \int_v^\infty \lambda(v',t)\beta_{v,v'-v} n(v'-v,t)\, dv' \qquad (4)\\
&+\lambda(v,t)\int_0^\infty \beta_{v,v'} n(v',t)\, dv' + \int_0^\infty \lambda(v',t) n(v',t)\beta_{v',v}\, dv'\\
&+\lambda(v,t) L(v,t)\ , \qquad t^{k-1}+\epsilon \le t \le t^k - \epsilon \quad (\epsilon \to 0)\\
\lambda\left(v, t^k - \epsilon\right) &= \lambda\left(v, t^k + \epsilon\right) + R_k^{-1}\ \left(y^k - h\left(n^k\right)\right)\ ,\\
\lambda\left(v, t^N + \epsilon\right) &= 0\ , \quad \lambda(v, t^o) = \lambda(v, t^o + \epsilon) + R_0^{-1}\ \left(n^o - n_B^o\right)\ ,\\
\lambda(v=0,t) &= 0\ , \quad \lambda(v=\infty, t) = 0\ .
\end{aligned}$$

A complete derivation of the adjoint equations and the sensitivity relations is given in [12]. The gradients of the cost functional (3) with respect to forward model parameters are [12]:

$$\begin{aligned}
\nabla_{n^o}\mathcal{J} &= \lambda(v,t^o)\ , \quad \nabla_{I(v,t)}\mathcal{J} = \frac{\partial \lambda(v,t)}{\partial v} n(v,t)\ ,\\
\nabla_{\beta_{v,v'}}\mathcal{J} &= \int_{t_0}^T \left(-\lambda(v,t) + \frac{1}{2}\lambda(v+v',t)\right) n(v,t) n(v',t)\, dt\\
\nabla_{S(v,t)}\mathcal{J} &= \lambda(v,t)\ , \quad \nabla_{L(v,t)}\mathcal{J} = -\lambda(v,t) n(v,t)\ .
\end{aligned}$$

All the above derivatives of the cost functional can be obtained by one integration of the forward model (1) during which the state $n(v,t)$ is saved, followed by a single backwards in time integration of the adjoint model (4). When solving the data assimilation problem each optimization iteration requires one evaluation of the cost functional and one evaluation of its gradient, i.e. one forward and one backward integration.

3.2 Discrete Adjoints of the Dynamic Equation

In the discrete approach the numerical discretization (2) of the the particle dynamic equation is considered to be the forward model. This is a pragmatic view, as only the numerical model is in fact available for analysis. The adjoint of the discrete model (2) is formulated and solved. The approach amounts to computing the derivatives of the numerical solution, rather than approximating the derivatives of the exact solution. Clearly, the formulation of the discrete adjoint equation depends not only on the dynamic equation, but also on the numerical method used to solve it.

Direct Approach. Taking the adjoint of the discrete equation (2) leads to a method to propagate the adjoint variables backwards in time [12]. The adjoint equation of (2) reads

$$\lambda^{k-1} = \lambda^k + \Delta t \ \left\{G^T(t^{k-1}) + F_n^T(n^{k-1}) + \left[(B+B^T)\times n^{k-1}\right] A^{-T} - L(t^{k-1})\right\} \cdot \lambda^k \quad (6)$$
$$+ R_k^{-1} \ \left(y^k - h\big(n^k\big)\right) \ , \qquad k = N, N-1 \ldots 1$$
$$\lambda^N = 0 \ , \qquad \lambda^o \longleftarrow \lambda^o + R_0^{-1} \ (n^o - n_B^o) \ . \quad (7)$$

The adjoint variable at t^o gives the gradient of the cost functional (3) with respect to the initial distribution, $\lambda^0 = \nabla_{n^o} J$. Note that this is the derivative of the numerical solution as used in the definition of (3), as opposed to the continuous adjoint formulation (5a), where λ^0 defines the derivative of the continuous solution.

In practice the continuous forward model (1) is solved numerically, and so is the continuous adjoint equation (4). Therefore the continuous adjoint approach is in practice a hybrid approach. The operations of numerical discretization and adjoint do not commute in general, and consequently the numerical solution of (4) is different from the discrete adjoint (6). For data assimilation problems one needs the derivative of the numerical solution, i.e. the discrete adjoints are in principle preferred. For sensitivity studies using the adjoint method one wants to approximate the sensitivities of the continuous model, and the continuous adjoint seems more appropriate.

Automatic Differentiation. Given a program that implements the forward model, automatic differentiation builds a new, augmented program, that computes the analytical derivatives (accurate up to machine precision) along with the original program [3]. Different modes of automatic differentiation can produce the tangent linear model or the discrete adjoint model. In this paper we use the automatic differentiation tool TAMC [15] of Giering and Kaminski to generate the discrete adjoint derivatives.

Automatic differentiation only requires (an implementation of) the original forward model. It constructs gradients for complicated forward models, and even models which contain legacy code. Next, automatic differentiation produces derivatives of the numerical solution which are appropriate for optimization purposes.

4 Numerical Results

The Test Problem. For the numerical experiments we consider the test problem from [2]. With N_t the total initial number of particles, V_m the mean initial volume, the constant coagulation kernel $\beta_{v,w} = \beta_0$, and the linear growth rate $I(v) = \sigma_o\, v$, the problem admits the analytical solution

$$n^A(v,t) = \frac{4\,N_\mathrm{t}}{V_\mathrm{m}(N_\mathrm{t}\beta_o t + 2)^2} \cdot \exp\left(\frac{-2v\exp\left(\sigma_o t\right)}{V_\mathrm{m}(N_\mathrm{t}\beta_o t + 2)} - \sigma_o t\right) \ .$$

We solve the dynamic equation for $\beta_o = 2.166 \times 10^{-6}$ cm^{3}h^{-1}particles^{-1}, $\sigma_o = 0.02$ h^{-1}, $N_{\rm t} = 10^4$ particles, $V_{\rm m} = 0.03$ μm^3. The values are chosen such that coagulation and growth have effects of comparable magnitude. The size range is truncated to $V_{min} = 10^{-3}\,\mu m^3$, $V_{max} = 1\mu m^3$. A piecewise linear discretization with 8 log-uniform bins is employed. The time interval is $[t_0 = 0, T = 48]$ hours, and the time step $\Delta t = 6$ minutes. The actual implementation is done in the (equivalent) volume concentration density formulation.

The experiments are carried out in the twin experiment framework. A run with the reference values for initial conditions, coagulation kernel, and growth rate is used to generate hourly pseudo-observations $\{y_1 \dots y_M\}$ of the number density, or a function of the number density.

The model is re-run with perturbed values of the initial conditions, coagulation kernel, and growth rate. The cost functional (3) is defined without background terms, and with observation covariance matrices equal to identity $R_k = I$. The lack of background terms is justified by our apriori knowledge of the fact that the initial guesses are wrong, while the observations are correct. The minimization algorithm is LBFGS [5], and the adjoint gradients are generated by TAMC. During optimization we prescribe the following bounds for the growth and coagulation parameters: $0.0 \le \sigma \le 0.5$ and $0.0 \le \beta \le 10^{-4}$.

As a measure of accuracy in the optimization process we consider the RMS difference between the reference and the optimized solutions:

$$RMS = \sqrt{\frac{1}{\text{ndof}+2} \sum_{j=1}^{\text{ndof}} \left(\frac{n^o_{\rm ref}[j] - n^o_{\rm opt}[j]}{n^o_{\rm ref}[j]}\right)^2 + \left(\frac{\sigma_{\rm ref} - \sigma_{\rm opt}}{\sigma_{\rm ref}}\right)^2 + \left(\frac{\beta_{\rm ref} - \beta_{\rm opt}}{\beta_{\rm ref}}\right)^2}$$

In the data assimilation experiments the initial guesses are obtained by significant perturbations of the reference values, with $\beta_p = 5\,\beta_o$ and $\sigma_p = 25\,\sigma_o$.

Sensitivity Analysis. Adjoint calculations are a useful tool for sensitivity analysis. A widely used measure of the relative change in the functional due to relative changes in the input parameters is given by the logarithmic sensitivities

$$s = \partial \log J\left(t^F\right) \;/\; \partial \log V\left(t^o\right) \;.$$

Figure 1 shows the logarithmic sensitivity of the mean volume densities in bins 4 and 8 at the end of the 48 hours simulation interval with respect to the initial mean volume densities (left) and slopes (right). For both cases relative changes in the bin slopes have a considerably smaller influence on the final result than relative changes in the mean densities.

Data Assimilation. We first consider a complete set of hourly observations, i.e. all parameters of the solution (mean concentrations and slopes in each bin) are observed once every simulation hour. The test problem consists of the evolution under both coagulation and growth.

In Figure 2 the results for recovering simultaneously the initial distribution, β and σ are shown. This experiment is challenging for the inversion procedure since perturbations now affect not only the initial distribution, but the dynamic

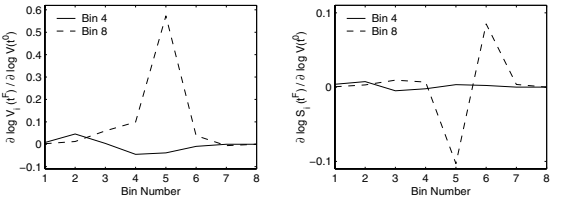

Fig. 1. The logarithmic sensitivities of the mean volume densities in bins 4 and 8 w.r.t. the initial distribution bin averages (left) and bin slopes (right).

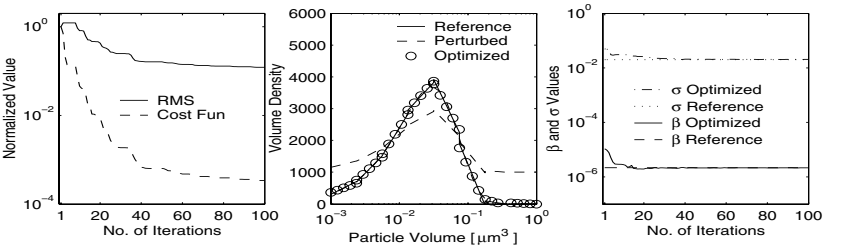

Fig. 2. Results for recovering V^o, β and σ for coagulation and growth from a complete set of hourly observations.

equation itself (through β and σ). The left Figure 2 panel shows the decrease of the cost function and RMS error with the number of iterations. The central panel shows the exact, reference, perturbed, and optimized distributions at the initial time. The optimized distribution is visually identical to the reference one. The right panel shows the reference and optimized β and σ, which are recovered accurately. Recovering only the initial distribution or only the parameters σ, β is easier and in both cases the minimization procedure converges in a small number of iterations.

Additional data assimilation experiments were carried out and the following conclusions were drawn: (1) The recovery procedure works even if observations are provided in only some of the bins. However less frequent observations in all bins are to be preferred to frequent but partial observations; (2) Observations of total surface, number, or volume density lead to a good recovery of densities only in the large size bins; (3) For the growth equation the optimization converges faster with the continuous adjoints, as compared to discrete adjoints; (4) The optimization performs similarly with discrete adjoints obtained by the direct and the automatic implementation.

5 Conclusions and Future Work

In this paper we have discussed the algorithmic tools needed for inverse aerosol modeling. Continuous and discrete adjoints of the integro-differential particle dynamic equation have been derived, together with formulas for sensitivity coefficients with respect to the coagulation kernel, the growth rate, and emission and

deposition coefficients. Numerical tests were carried out using a single component particle dynamics model problem. From hourly measurements of the particle size one can recover the initial distribution as well as the parameters of the model.

The overall conclusion is that variational data assimilation is a feasible approach for particle dynamics. Discrete adjoints can be obtained easily through automatic differentiation, while continuous adjoints are a useful alternative.

A testing of the computational tools developed in this paper for a realistic growth problem of three-species aerosols is presented in [13]. Future work will extend the inverse modeling of aerosols to include chemical and thermodynamic processes. The techniques developed will be used ultimately to perform data assimilation in full three-dimensional models.

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