Modeling aerosol dynamics : a stochastic algorithm *

Edouard Debry[1], Benjamin Jourdain[2], and Bruno Sportisse[1]

[1] Centre d'Enseignement et de Recherche sur l'Eau, la Ville et l'Environnement, Ecole Nationale des Ponts et Chaussées (ENPC-CEREVE-Air team), rue Blaise Pascal, 77455 Champs sur Marne, France.
E-mail:debry@cereve.enpc.fr , sportiss@cereve.enpc.fr

[2] Centre d'enseignement et de recherche en Mathématiques Informatique et Calcul scientifique, Ecole Nationale des Ponts et Chaussées, rue Blaise Pascal, 77455 Champs sur Marne, France. E-mail:jourdain@cermics.enpc.fr

Abstract. We present in this article a stochastic algorithm based mainly on [3] and [2] applied to the integration of the General Dynamics Equation (GDE) for aerosols . This algorithm is validated by comparison with an analytical solution of a coagulation-condensation-evaporation model .

Introduction

Aerosol dynamics is a key issue in Atmospheric Modeling . Many aerosol properties such as optical properties or health impact depend on the aerosol size . Thus it appears important to modelize the size distribution of aerosols and the physical processes by which aerosols are affected [8] : coagulation, condensation-evaporation and nucleation .
Splitting is usually advocated in order to focus on each process separately .
The behavior of a size distribution undergoing coagulation is modelized by the Smoluchowski equation and reference solutions are only known for particular forms of coagulation kernels in academic cases [8] . The usually advocated numerical methods are the so-called "size binning" algorithms . We refer for instance to Stratton [9], Jacobson [5] and Diaz [4] for more details . A major drawback of such approaches is the lack of convergence results as pointed out by Levin in [10] . The search for a reference method is then necessary .
The article is organized as follows . In first section we quickly recall the GDE for atmospheric aerosols and we present the mass flow formulation . We present our algorithm in second section . Some numerical tests are summarized in third section with a coagulation-condensation-evaporation model for which an analytical solution is available [7] .

* APMS conference, April 2001, Paris, France

1 General dynamics equation and mass flow equation

1.1 General dynamics equation

Let us first recall the general dynamic equation in its continuous version :

$$\begin{aligned}&\forall\, x \geq x_0\ ,\quad c(x,0)=c_0(x)\\ \frac{\partial c}{\partial t}(x,t) &= \frac{1}{2}\int_{x_0}^{x-x_0} K(y,x-y)c(y,t)c(x-y,t)\,dy - c(x,t)\int_{x_0}^{+\infty} K(x,y)c(y,t)\,dy\\ &\quad - \frac{\partial (I(x,t)c(x,t))}{\partial x} + J_0(t)\delta_{(x_0,x)}\end{aligned} \tag{1}$$

where $c(x,t)\,dx$ represents the aerosol concentration whose volume range between x and $x+dx$ at time t, x_0 is the smallest volume from which an aerosol becomes stable, i.e. the volume of nucleation, $K(x,y)$ is the coagulation kernel between an aerosol of volume x and an aerosol of volume y, $I(x,t)$ is the condensation-evaporation kernel, positive in case of condensation and negative in case of evaporation and $J_0(t)$ is the nucleation rate, i.e. a number of aerosols per air volume per seconds .
In our case, for atmospheric aerosols, the coagulation is mostly due to the Brownian activity and the condensation-evaporation is a diffusion process between aerosol and gas phases, therefore their respective kernels are in the continuous regime[1] :

$$K(x,y) = K\frac{(x^{\frac{1}{3}}+y^{\frac{1}{3}})^2}{(xy)^{\frac{1}{3}}}\ ,\quad I(x,t)=c(t)x^{\frac{1}{3}} \tag{2}$$

where K is called the Brownian constant and $c(t)$ is related to the equilibrium processes between gas and aerosol phases .
The most natural idea to simulate coagulation with stochastic processes is to introduce numerical particles representing physical aerosol particles and to let them coagulate . This kind of algorithm has been developped by Lushnikov [2] . The drawback of such algorithms is that the number of numerical particles decreases with time whereas they are valid in the limit of the total number of particles going to infinity . Numerical particles have then to be added on the fly, which is not necessary easy to do in a rigorous way .
An alternative approach is to introduce numerical particles representing a fixed volume of aerosol so that their number remains constant . Such algorithms were developped by Babovsky [1], Wagner and Eibeck [3] for the Smoluchowski equation (i.e. only coagulation) in order to avoid the problem raised above, and were called “mass flow algorithm” since they are derived from the equation for the coagulation of the mass distribution .

[1] That is to say when $K_n \ll 1$, K_n Knudsen number

1.2 Mass flow equation

Let $\rho(x,t)$ be the volume distribution[2] of aerosols, i.e. $\rho(x,t)\,dx$ is the total volume of aerosols whose volume ranges between x and $x+dx$ per air volume . The volume distribution is related to the aerosol number distribution by $\rho(x,t) = c(x,t)x$. As we assume the specific mass of aerosols to be constant, the mass distribution and volume distribution are merely proportional . The total aerosol volume per air volume ρ_t at time t is given by :

$$\rho_t = \int_{x_0}^{+\infty} \rho(x,t)\,dx \tag{3}$$

which depends on time due to the condensation-evaporation and nucleation processes . ρ_0 is then the initial total aerosol volume . By multiplying (1) by $\frac{x}{\rho_0}$ we get the mass flow equation :

$$\begin{aligned}
&\forall\ x \geq x_0\ ,\quad \tilde{\rho}(x,0) = \tilde{\rho}_0(x) \\
&\frac{\partial \tilde{\rho}}{\partial t}(x,t) = \underbrace{\int_{x_0}^{x-x_0} \rho_0 \frac{K(y,x-y)}{y}\tilde{\rho}(y,t)\tilde{\rho}(x-y,t)\,dy - \tilde{\rho}(x,t)\int_{x_0}^{+\infty} \rho_0 \frac{K(x,y)}{y}\tilde{\rho}(y,t)\,dy}_{\text{stochastic jump due to coagulation}} \\
&\qquad + \underbrace{\frac{I(x,t)}{x}\tilde{\rho}(x,t) + \frac{J_0(t)x_0}{\rho_0}\delta_{(x_0,x)}}_{\text{non conservative terms}} - \underbrace{\frac{\partial(I(x,t)\tilde{\rho}(x,t))}{\partial x}}_{\text{conservative term}}
\end{aligned} \tag{4}$$

where $\tilde{\rho}(x,t) = \frac{\rho(x,t)}{\rho_0}$. The kernels to be taken into account are now $\frac{K(x,y)}{y}$ for coagulation, $\frac{I(x,t)}{x}$ for condensation and $\frac{J_0(t)x_0}{\rho_0}$ for nucleation . This equation provides us the right way to develop a Mass flow algorithm for coupled coagulation, condensation and nucleation .

2 Mass flow algorithm

2.1 General scope

The mass flow algorithm consists in spreading the initial total aerosol volume ρ_0 per air volume over numerical particles indexed by i, which represent each one a fixed volume $\frac{\rho_0}{P_0}$ where P_0 is then the initial number of numerical particles chosen by the modelist . At time t_k if y_i^k denotes the size of the i^{th} numerical particle then this one stands for $\frac{\rho_0}{y_i^k P_0}$ aerosols of volume y_i^k . As ρ_t may vary with time the number of numerical particles may also vary . Let P_k be the number of numerical particles at time t_k . Thus the total volume

[2] As we assume the specific mass of aerosols to be constant, i.e. time and volume independent, there is equivalence between volume and mass distribution .

and concentration of aerosols per air volume at time t_k are respectively given by :

$$V_k = \sum_{i=1}^{P_k} y_i^k \frac{\rho_0}{y_i^k P_0} = \rho_0 \frac{P_k}{P_0} \ , \quad C_k = \sum_{i=1}^{P_k} \frac{\rho_0}{y_i^k P_0} = \frac{\rho_0}{P_0} \sum_{i=1}^{P_k} \frac{1}{y_i^k} \tag{5}$$

Thus we see that in case of pure coagulation the number of numerical particles has to be conserved in our algorithm on the contrary to the various algorithms of Lushnikov [2] . In case of pure condensation-evaporation C_k should be conserved, which means in term of probability that the average of C_k has to be C_0 .
The algorithm is then the following one for one time period $[0, T]$:

1. **Monte Carlo loop** (labelled by $1 \leq mc \leq MC$) .
   The Monte Carlo process is valid in the limit of the number of experiments going to infinity .
   - *Initialization* . (2.1.1)
     Calculation of the initial state of the system $(y_i^0, \ i = 1, \ldots, P_0)$.
   - *Time loop :* $[0, T]$
     - *Choice of a time step* τ_k . (2.1.2)
       τ_k is randomly calculated according to the state of the system at time t_k : (y_i^k , $i = 1, \ldots, P_k$) .
     - *Calculation of the state of the system at time* $t_{k+1} = t_k + \tau_k$ *:*
       (y_i^{k+1} , $i = 1, \ldots, P_{k+1}$) (2.1.3)
       * Integration of both stochastic jump and non-conservative terms of (4) .
       * Integration of the conservative term of (4) .
     - *Rebuilding of the size distribution :* $c^{mc}(x, t_k^e)$ for some times t_k^e chosen within $[0, T]$. (2.1.4)
2. **Average of concentrations.**
   At the end of the Monte Carlo loop, we calculate the average concentration over the MC experiments :

$$c(x, t_k^e) = \frac{1}{MC} \sum_{mc=1}^{MC} c^{mc}(x, t_k^e) \tag{6}$$

We now give some details for each step .

2.1.1 Initialization This step consists in assigning to each numerical particle i an initial size y_i^0 according to the initial volume distribution $x \mapsto \rho_0(x)$. This step can be performed :

- either by generating the y_i^0 independently according to the probability density :

$$x \mapsto \tilde{\rho_0}(x) = \frac{\rho_0(x)}{\rho_0} \tag{7}$$

- or by using a deterministic initialization based on the associated cumulative distribution function :

$$y_i^0 = \inf(\; x \; / \; \int_0^x \tilde{\rho_0}(y)\, dy \geq \frac{2i-1}{2P_0} \;) \qquad (8)$$

2.1.2 Calculation of a time step The state of the system is constant over each interval $[t_k, t_{k+1}[$ (definition of stochastic jump process) .
In this stochastic algorithm the time step τ_k is randomly generated according to the exponential law (density $p(s) = \lambda_k \exp(-\lambda_k s) 1_{s \geq 0}$) with parameter λ_k related to the various physical processes and the state of the system at time t_k :

$$\lambda_k = \underbrace{\frac{J_0(t_k)x_0}{\rho_0}}_{\text{nucleation}} + \underbrace{\sum_{i=1}^{P_k} \frac{|I(t_k, y_i^k)|}{y_i^k}}_{\text{condensation}} + \underbrace{\frac{\rho_0}{P_0} \sum_{i,j=1}^{P_k} \frac{K(y_i^k, y_j^k)}{y_j^k}}_{\text{coagulation}} \qquad (9)$$

Let us precise the dimensions of each quantity . As the dimension of the aerosol distribution $c(x,t)$ is [3] $V_{sol}^{-1} V_{air}^{-1}$ one can check with (4) that the dimensions of $K(y_i^k, y_j^k)$, $I(t_k, y_i^k)$ and $J_0(t_k)$ are respectively $V_{air} s^{-1}$, $V_{sol} s^{-1}$ and $V_{air}^{-1} s^{-1}$. λ_k is thus expressed in sec^{-1} . Let us recall that the mean value of such a law is $\frac{1}{\lambda_k}$ while its variance is $\frac{1}{\lambda_k^2}$. The average time step of integration is thus $\frac{1}{\lambda_k}$.
The numerical cost of the computation of λ_k is in $O(P_k^2)$ because of the coagulation term . It can be reduced in case the coagulation kernel is factorizable or by using a factorizable upper bound (see A) .

2.1.3 Calculation of the state at time t_{k+1} This step consists in computing the new values y_i^{k+1} of each numerical particles . Equation (4) makes appear a stochastic jump, conservative and non conservative terms which are successively integrated in the algorithm .

1. **Integration of the sochastic jump and non conservative terms :**
   - *Choice of a physical process .*
     One physical process is randomly chosen at each time step among coagulation, condensation and nucleation according to their respective weights in the expression of λ_k (9) .
   - *Updating of the chosen physical process .*
     – If nucleation is chosen, then a new numerical particle is created with the value x_0, i.e. $P_{k+1} = P_k + 1$ and $y_{P_{k+1}}^k = x_0$.

[3] where V_{sol} stands for an aerosol volume (μm^3) and V_{air} stands for an air volume (m^3) .

- If condensation is chosen, then a numerical particle is randomly chosen according to the probability law :

$$\sharp_i = \frac{\frac{|I(t_k, y_i^k)|}{y_i^k}}{\sum_{j=1}^{P_k} \frac{|I(t_k, y_j^k)|}{y_j^k}} \tag{10}$$

  Let i be the numerical particle chosen,
  * if $I(t_k, y_i^k) > 0$ a new numerical particle is created with the value y_i^k, i.e. $P_{k+1} = P_k + 1$ and $y_{P_{k+1}}^k = y_i^k$.
  * if $I(t_k, y_i^k) < 0$ the i^{th} numerical particle is removed, i.e. $P_{k+1} = P_k - 1$.
- If coagulation is chosen, then one pair (i, j) of numerical particles is randomly chosen with the probability law :

$$\sharp_{i,j} = \frac{\frac{K(y_i^k, y_j^k)}{y_j^k}}{\sum_{m,l=1}^{P_k} \frac{K(y_m^k, y_l^k)}{y_l^k}} \tag{11}$$

  This probability law can be simplified for some particular forms of the coagulation kernel (see A) . Then we update coagulation on the chosen pair by :

$$y_i^k + y_j^k \longrightarrow y_i^k \tag{12}$$

$$y_j^k \longrightarrow y_j^k \tag{13}$$

  Let us notice that the coagulation between two numerical particles no longer represents the physical coagulation between aerosols. Indeed (12) means roughly that we have replaced $\frac{1}{y_i^k}$ aerosols of volume y_i^k by $\frac{1}{y_i^k + y_j^k}$ aerosols of volume $y_i^k + y_j^k$ which are the result of the coagulation between aerosols of the i^{th} and j^{th} numerical particles . As coagulation conserves volume, the number of numerical particles remains unchanged, $P_{k+1} = P_k$.

2. **Integration of the conservative term :** This step consists in integrating the following system on the time step τ_k :

$$\begin{aligned} i = 1, \ldots, P_{k+1} \ , \quad & y_i(t=0) = y_i^k \\ & \dot{y}_i = I(t, y_i(t)) \\ & \text{and } y_i^{k+1} = y_i(t = \tau_k) \end{aligned} \tag{14}$$

   When there is evaporation we have to check that all numerical particles are beyond the stable aerosol volume, i.e. the nucleation volume : for $i = 1, \ldots, P_{k+1}$, if $y_i^{k+1} < x_0$ the i^{th} numerical particle is removed . In the general case we have to use a numerical solver for this ODE . In our case we have an analytical solution since $I(t, x) = c(t) x^{\frac{1}{3}}$.

2.1.4 Calculation of concentrations Let t_k^e , $k = 1, \ldots, \mathrm{k}_{max}$ be an array of times distributed within the period of the experiment $[0, T]$. This step is the opposite of initialization, we look for calculating $c^{mc}(x, t_k^e)$ according to the values of the numerical particles at time t_k^e . The state of the system is known only at the randomly generated instants t_k . The state of the system at time t_k^e is approximated by the one at t_I defined by $I = inf(\ i \ /\ t_k^e < t_{i+1}\ )$, then the concentration and volume distribution are respectively given by :

$$c^{mc}(x, t_k^e) = \frac{\rho_0}{P_0} \sum_{j=1}^{P_I} \frac{1}{y_j^I} \delta_{(y_j^I, x)}\ ,\quad v^{mc}(x, t_k^e) = \frac{\rho_0}{P_0} \sum_{j=1}^{P_I} \delta_{(y_j^I, x)} \tag{15}$$

To approximate the continuous distribution from this linear combination of Dirac masses, we divide the size spectrum into bins $B_j = [x_j^-, x_j^+[$ [4] , and we actually calculate

$$C_j^{th}(t_k^e) = \int_{x_j^-}^{x_j^+} c(x, t_I)\, dx\ ,\quad V_j^{th}(t_k^e) = \int_{x_j^-}^{x_j^+} x c(x, t_I)\, dx \tag{16}$$

Then $C_j(t_k^e)$ and $V_j(t_k^e)$ are numerically given by :

$$C_j^{num}(t_k^e) = \frac{\rho_0}{P_0} \sum_{i=1}^{P_I} \frac{1}{y_i^I} \delta_{(y_i^I \in B_j)}\ ,\quad V_j^{num}(t_k^e) = \frac{\rho_0}{P_0} \sum_{i=1}^{P_I} \delta_{(y_i^I \in B_j)} \tag{17}$$

3 Some numerical tests

3.1 Numerical set up

In order to test this algorithm we have chosen kernels and initial conditions for which there exits an analytical solution .
The aerosol population is characterized at the initial time by :

$$x \geq 0\ ,\quad c(t = 0, x) = \frac{c_0}{\alpha} \exp(-\frac{x}{\alpha}) \tag{18}$$

where α is a typical aerosol volume, and c_0 is the initial total concentration of aerosols . We assume the aerosol population to undergo only constant coagulation and linear condensation :

$$K(x, y) = K\ ,\quad I(t, x) = cx\ ,\ c > 0 \tag{19}$$

According to these assumptions the analytical solution is then [7] [6] :

$$c(x, t) = \frac{(M_0(t))^2}{M_1(t)} \exp(-x \frac{M_0(t)}{M_1(t)}) \tag{20}$$

[4] x_j^- and x_j^+ are the lower and upper boundary of bin B_j

where $M_0(t)$ and $M_1(t)$ are the first two moments of the size distribution, i.e. respectively the total aerosol number and the total aerosol volume :

$$M_0(t) = \frac{c_0}{1 + \frac{t}{\tau_c}} \ , \quad M_1(t) = c_0 \alpha \exp(\frac{t}{\tau_d}) \tag{21}$$

Where$\tau_c = \frac{2}{K c_0}$ and $\tau_d = \frac{1}{c}$ are respectively the coagulation and condensation characteristic times .
We check this algorithm in the case when the contribution of both coagulation and condensation remain of the same order, i.e. when their characteristic times are comparable . The constant coagulation kernel is chosen equal to the Brownian constant :

$$K = \frac{2 k_b T}{3\mu} = 1,606.10^{-16} \ m^3 s^{-1} \tag{22}$$

with $k_b = 1,381.10^{-23} \ JK^{-1}$ (Boltzmann's constant), $T = 300 \ K$ (temperature), $\mu = 1,72.10^{-4} \ gcm^{-1}s^{-1}$ (dynamic viscosity of air) . With $c_0 = 10^{12}$ aerosols$/m^3$ the characterisitic time of coagulation is then :

$$\tau_c = \simeq 12500 s \tag{23}$$

We choose $c = 0.0001 \ s^{-1}$ so that $\tau_d = 10000s$ although this may not be a physical time for condensation .

3.2 Calculation of errors

Let $c_{num}(x,t)$ be the solution given by the algorithm at any time t . The two errors of interest are the error on the number distribution and the error on the volume distribution :

$$err_n = \frac{1}{T} \int_0^T \int_0^{+\infty} |c_{num}(x,t) - c_{th}(x,t)| \, dx \, dt \tag{24}$$

$$err_v = \frac{1}{T} \int_0^T \int_0^{+\infty} |v_{num}(x,t) - v_{th}(x,t)| \, dx \, dt \tag{25}$$

In section 2.1.4 we have seen that $c_{num}(x,t)$ cannot be directly obtained for any x value . Then we approximate (24) by

$$err_n = \frac{1}{T} \sum_{k=0}^{kt} \Delta t_k \sum_{i=1}^{+\infty} |(C_i^{\text{num}} - C_i^{th})(t_k^e)| \tag{26}$$

$$err_v = \frac{1}{T} \sum_{k=0}^{kt} \Delta t_k \sum_{i=1}^{+\infty} |(V_i^{\text{num}} - V_i^{th})(t_k^e)| \ , \quad \Delta t_k = t_{k+1}^e - t_k^e \tag{27}$$

3.3 Some results

Figure 1 shows the evolution of the volume distribution in the case of constant coagulation and linear condensation with 100 Monte Carlo experiments and 1000 initial numerical particles over the time period $T = 10000s$. The relative error at $T = 10000s$ is 2.25% .

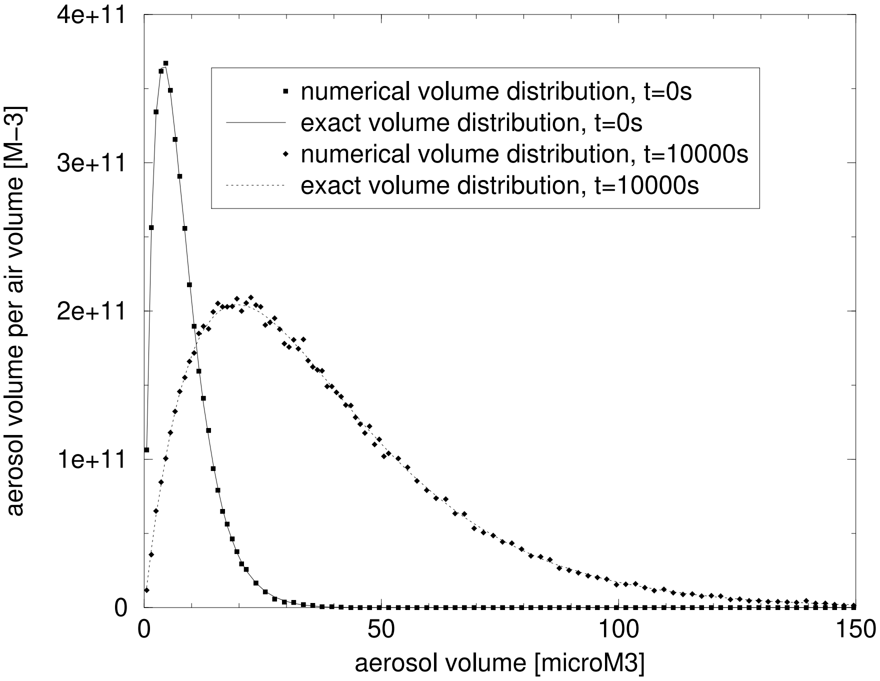

Fig. 1. Evolution of the volume distribution in the case of constant coagulation and linear condensation, MC=100 .

Figures 2 and 3 show respectively the behavior of the relative errors

$$\frac{err_n}{\int_0^\infty c(T,x)\,dx} \;, \quad \frac{err_v}{\int_0^\infty v(T,x)\,dx} \tag{28}$$

for various number of Monte Carlo experiments MC and various numbers of numerical particle P, in the case of pure coagulation . We see roughly that for a given accuracy the product $MC \times P$ remains constant, furthermore as the CPU time is proportional to $MC \times P^2$ a good strategy seems to take a small number of particles .
Figures 4 and 5 show that the errors decrease as $\frac{1}{\sqrt{P}}$ with the total number of particles P : this seems to point out the existence of a central limit theorem associated with the convergence as $P \to \infty$.

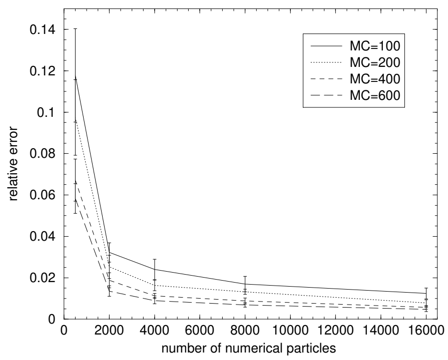

Fig. 2. Relative error on the total number of aerosols per air volume, for constant cocoagulation .

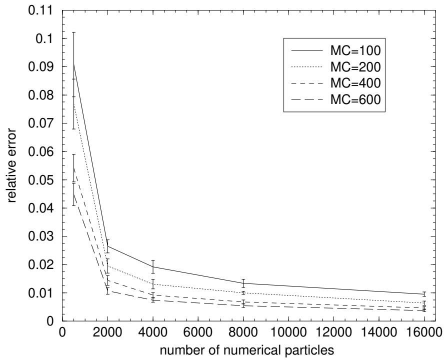

Fig. 3. Relative error on the total volume of aerosol per air volume, for constant coagulation .

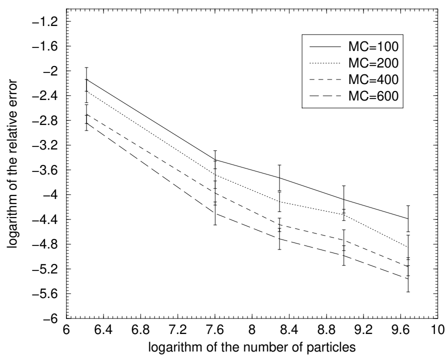

Fig. 4. Logarithm of the relative error on the total number of aerosols per air volume, for constant coagulation .

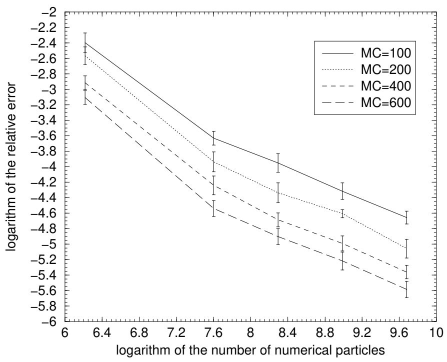

Fig. 5. Logarithm of the relative error on the total volume of aerosols per air volume, for constant coagulation .

Conclusion

We have used a stochastic model for simulating the GDE for atmospheric aerosols . This algorithm provides a reference for testing numerical algorithms devoted to the integration of the GDE . By using appropriate numerical strategies (autocycling), a small number of particles (1000) may ensure a good accuracy .

Acknowledgements We thank our colleagues Gilles Bergametti (LISA), Bernard Aumont (LISA), and Luc Musson-Genon (EDF-CEREVE) for fruitful discussions . This work is a part of a common project devoted to aerosol modeling .

A Factorization of the coagulation term

If the coagulation kernel can be factorized in $K(y_i, y_j) = f(y_i)f(y_j)$, then the probability law (11) can be factorized in two independent probability laws, one for i and one for j :

$$\frac{f(y_i)}{\sum_m f(y_m)} \;,\; \frac{\frac{f(y_j)}{y_j}}{\sum_l \frac{f(y_l)}{y_l}} \tag{29}$$

which reduces to $O(P_k)$ the numerical cost of (11) as well as the cost of the computation of λ_k .
Unfortunately the various coagulation kernels for aerosols [8] cannot be factorized, in this case one may find a function h so that : $K(y_i, y_j) \leq h(y_i)h(y_j)$, i and j are then chosen (29) in which f is replaced by h . To take into account the gap[5] between $K(y_i, y_j)$ and $h(y_i)h(y_j)$, we generate a random number u uniformly over $[0, 1]$ and we update coagulation if only $u \leq \frac{K(y_i,y_j)}{h(y_i)h(y_j)}$.

References

1. Hans Babovsky. On a monte carlo scheme for smoluchowski's coagulation equation. *Monte Carlo Methods and Applications*, 5(1):1–18, 1999.
2. E.R. Domilovskii, A.A. Lushnikov, and V.N. Piskunov. Monte carlo simulation of coagulation processes. *Dockl. Akad. Nauk SSSR, Ser. Phys. Chem.*, 240(1), 1978.
3. A. Eibeck and W. Wagner. Stochastic particle approximations for smoluchowski's coagulation equation. Technical report, Weierstrass-Institut for Applied Analysis and Stochastics, 2000. Preprint No. 585.
4. Fernndez-Daz, Muiz, Braa, Garca, and Nieto. A modified semi-implicit method to obtain the evolution of an aerosol by coagulation. *Atmospheric Environment*, 34:4301–4314, March 2000.
5. M.Z. Jacobson, R.P. Turco, E.J. Jensen, and O.B. Toon. Modeling coagulation among particles of different composition and size. *Atmospheric Environment*, 28(7):1327–1338, 1994.
6. T.W. Peterson, F. Gelbard, and J.H. Seinfeld. dynamics of source-reinforced, coagulating, and condensing aerosols. *Journal of collod and Interface Science*, 63(3):426–445, March 1978.
7. T.E. Ramabhadran, T.W. Peterson, and J.H. Seinfeld. Dynamics of aerosol coagulation and condensation. *AIChe Journal*, 22(5):840–851, September 1976.
8. J.H. Seinfeld and S.N. Pandis. *Atmospheric chemistry and Physics*. Wiley-interscience, 1998.
9. Stratton, Gans, and Williams. Coagulation algorithms with size binning. *Computational Physics*, 112:364–369, 1994.
10. Tzivion, Reisin, and Levin. A numerical solution of the kinetic collection equation using high spectral grid resolution : A proposed reference. *Journal of Computational Physics*, 148:527–544, 1999.

[5] This gap means that with the probability law (29) we may coagulate more than the real coagulation kernel does .