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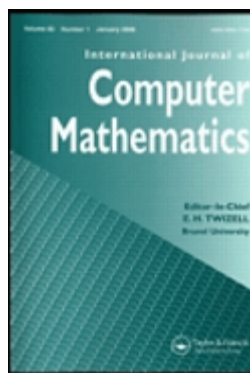
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Mathematical study of the selective removal of atmospheric aerosols by coagulation, condensation and gravitational settling

Review Article

Mathematical study of the selective removal of different classes of atmospheric aerosols by coagulation, condensation and gravitational settling and health impact (SI – CMMSE 2006)

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Abstract

The aim of this paper is to study the scavenging efficiencies of aerosol particles after some given dynamical mechanisms of removal known as coagulation, condensation and gravitational settling as a function of time. It also analyses the health impact of aerosol before and after the above dynamical mechanisms by comparing the respirable dust fractions. The well-known equations of scavenging are applied to eight different classes of atmospheric aerosols: marine background (MB), clean continental background (CCB), average background (AB), background and aged urban plume (BAUP), background and local sources (BLS), urban average (UA), urban and freeway (UF) and central power plant (CCP). From this work it is inferred that respirable dust is scavenged with relative difficulty by coagulation, condensation and gravitational settling. The deposition of particles in the lungs relies on the same basic mechanisms that cause collection in a filter, but the relative importance of each mechanism is quite different. While filtration occurs in a fixed system at a steady flow rate, respiratory deposition occurs in a system of changing geometry, with a flow that changes with time and cycles in direction. This added complexity means that predicting deposition from the basic theory is much more difficult, and we must rely to a greater extent on experimental data and empirically derived equations. Therefore, an understanding of how and where particles deposit in our lungs is necessary to evaluate properly the health hazard of aerosols. As compared with the initial volume of respirable aerosol, roughly 10% remains after 18 hours of coagulation, condensation and gravitational settling. Besides, gravitational settling is the main removal mechanism of respirable aerosol compared to condensation and coagulation.

Keywords: Computational fluid dynamics; Aerosol; Precipitation scavenging; Numerical methods; Respirable dust; Health impact

AMS Subject Classification: 76N99; 82B21; 74S30; 34K28

1 Introduction

Particle matter (aerosol) processes are emerging as a new frontier in environmental studies. Aerosols [11] negatively affect human health, reduce visibility, and modify warming through scattering and absorption of solar radiation.

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To study accurately the effects of aerosols it is necessary to resolve aerosol number and mass distributions as a function of size. Treatment of aerosol processes leads to an order of magnitude increase in the overall computational time of an air quality model. This is mainly due to repeatedly solving the aerosol chemistry (or chemical equilibria) for different particle sizes. Therefore there is a clear need for rigorous, reliable and efficient computational techniques for aerosol simulations. In particular, there is a need for methods that accurately solve aerosol dynamics using a small number of size bins (discretization points), such that the time for aerosol chemistry calculations is manageable.

Simulation of aerosols plays a significant role in air pollution modelling. For a correct representation of particles in the atmosphere one needs to accurately solve for the size distribution of particle populations. In this paper we propose a discretization method for the aerosol dynamic equation based on piecewise polynomial discretizations of the particle size distribution. This paper presents a discretization technique known as 'time splitting' in order to solve the general dynamic equation (GDE) of aerosol. The growth term is solved using the flux-based modified method of characteristics, and coagulation term by the J -space transform and a cubic spline method. Numerical tests reveal that accurate solutions can be obtained with a very small number of size bins. We have implemented an efficient software package to solve the proposed algorithms.

Atmospheric aerosols consist of particles ranging in size from a few tens of angstroms to several hundreds micrometers [1, 9, 11, 14]. Particles less than 2.5 μm in diameter are generally referred to as 'fine' and those greater than 2.5 μm diameter as 'coarse'. The fine and coarse particle modes, in general, originate separately, are transformed separately, are removed from the atmosphere by different mechanisms, require different techniques for their removal from sources, have different chemical composition, have different optical properties, and differ significantly in their deposition patterns in the respiratory tract [8]. Therefore, the distinction between fine and coarse particles is a fundamental one in any discussion of the physics, chemistry, measurement, or 'health effects of aerosols'. Fine particles can often be divided roughly in two modes: the nuclei mode and the accumulation mode. The nuclei mode, extending from about 0.005 to 0.1 μm diameter, accounts for the preponderance of particles by number. Because of their small size, these particles rarely account for more than a few percent of the total mass of airborne particles. Particles in the nuclei mode are formed from condensation of hot vapours during combustion processes and from the nucleation of atmospheric species to form fresh particles. They are lost principally by coagulation with larger particles. The accumulation mode, extending from 0.1 to about 2.5 μm , usually accounts for most of the aerosol surface area and a substantial part of the aerosol mass [3]. The source of particles in the accumulation mode is the coagulation of particles in the nuclei mode and from condensation of vapours onto existing particles, causing them to grow into this size range. The accumulation mode is so named because particle removal mechanisms are least efficient in this regime, causing particles to accumulate there. The coarse mode, from $>2.5 \mu\text{m}$ diameter, is formed by mechanical processes and usually consists of man-made and natural dust particles. Coarse particles have sufficiently large sedimentation velocities that they settle out of the atmosphere in a reasonably short time. Because removal mechanisms that are efficient at the small and large particle extremes of the size spectrum are inefficient in the accumulation range, particles in the accumulation mode tend to have considerably longer atmospheric residence times than those in either the nuclei or coarse mode. Dry deposition is, broadly speaking, the transport of gaseous and particulate species from the atmosphere onto surfaces in the absence of precipitation [6]. The motion of the particle, however, arises in our case because of the action of an external force on the particle called 'gravity'. The drag force arises as soon as there is a difference between the velocity of the particle and that of the fluid. The basis of the description of the behaviour of a particle in a fluid is its equation of motion.

The hazard caused by inhaled particles depends on their chemical composition and on the site at which they deposit within the respiratory system. Thus, an understanding of how and where particles deposit in our lungs is necessary to evaluate properly the health hazards of aerosols. Such an understanding is also central to the effective administration of pharmaceutical aerosols by inhalation [8]. Humans have evolved effective defence mechanisms against aerosol hazards, and we consider here the first line of defence: mechanisms that restrict access of particles the sensitive regions of the lungs. In this work we focus on the scavenging efficiencies of aerosol particles after some given mechanisms of removal known as coagulation, condensation and gravitational settling as a function of time and we also analyse the health impact of aerosol before and after the above dynamical mechanisms by comparing the respirable dust fractions [7, 14] for eight different classes of typical atmospheric aerosols.

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2 Physical-mathematical model

The equation describing the behaviour of aerosols is a non-linear, integro-differential equation of considerable complexity [4, 12, 20]. If all the deposition, condensation, and coagulation mechanisms are included, it is clear that the only feasible method of solution is a numerical one. Indeed, in practical calculations this is the procedure employed and many computer codes exist which solve the dynamic equation using finite difference or finite element methods [19]. A major drawback of numerical methods is the vast amount of computer time necessary to obtain a useful survey of all the relevant parameters. This causes difficulty in gaining physical insight into the problem. The Continuous General Dynamic Equation (Continuous GDE) can be written for the three mechanisms (coagulation, condensation and gravitational settling) according to the following expression [9, 16-17]:

$$\begin{aligned} \frac{\partial N_v}{\partial t} = & \frac{1}{2} \int_{v_0}^{v-v_0} K(v-\tilde{v}, \tilde{v}) N_v(v-\tilde{v}, t) N_v(\tilde{v}, t) d\tilde{v} - N_v(v, t) \int_{v_0}^{\infty} K(v, \tilde{v}) N_v(\tilde{v}, t) d\tilde{v} \\ & - \frac{\partial(I_v N_v)}{\partial v} - \frac{v_g}{h} \cdot N_v \end{aligned} \quad (1)$$

where $N_v(v, t)$ ($\mu\text{m}^3 \text{cm}^{-3}$) is the continuous particle size distribution density function, v_0 is the volume of the smallest stable particle and $K(v-\tilde{v}, \tilde{v})$ is the continuous coagulation coefficient between particles of volume $v-\tilde{v}$ and \tilde{v} . The first two integrals on the right-hand side represent the coagulation, the following term represents the condensation and the last term is due to gravitational settling. In order to close this initial boundary-value problem (IBVP), we have naturally to impose to the previous equation (GDE) the following initial and boundary conditions [17]:

$$IC \quad N_v(v, 0) = N_0(v) \quad (2)$$

$$BC \quad N_v(v, t) = 0 \quad \forall v < v_0 \quad (3)$$

where *IC* means initial condition and *BC* means boundary condition. Of this form, the final GDE is written as [9, 15-16]:

$$\begin{aligned} \frac{\partial N_v}{\partial t} = & \frac{1}{2} \int_0^v K(v-\tilde{v}, \tilde{v}) N_v(v-\tilde{v}, t) N_v(\tilde{v}, t) d\tilde{v} - N_v(v, t) \int_0^{\infty} K(v, \tilde{v}) N_v(\tilde{v}, t) d\tilde{v} \\ & - \frac{\partial(I_v N_v)}{\partial v} - \frac{v_g}{h} \cdot N_v \end{aligned} \quad (4)$$

2.1 Coagulation

A population of aerosol particles evolves in time as the consequence of colliding with each other in order to adhere. This process is termed 'coagulation'. It is known that two kinds of coagulation exist, depending on the collision mechanism [19]: (a) one due to the Brownian motion of aerosol particles or 'Brownian coagulation' and (b) another due to turbulent flow or 'coagulation in turbulence'.

Coagulation modifies the particle size distribution (PSD) since it reduces the overall number of particles and it increases their mean size (diameter), which facilitates the removal of particles by other mechanisms as gravitational settling. Either kind of coagulation reduces the particle number but the total particle volume remains constant.

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2.2 Nucleation

The formation and growth of an aerosol by condensation require a surface on which the vapour can condense initially. This surface can be, for example, a small cluster of vapour molecules, an ion or a solid salt particle termed *condensation nucleus*. Homogeneous nucleation is the nucleation of vapour on embryos consisting of vapour molecules in the absence of foreign substances, whereas heterogeneous nucleation [3] is the nucleation on a foreign substance or surface, such as an ion or a solid particle.

2.3 Deposition

Deposition is the removal of aerosol from the atmosphere to the Earth's surface so that the aerosol PSD suffers modifications in the particle number and in the overall mass of these. Particle deposition due to dynamic processes is studied, dispensing with deposition caused by chemical processes. In 'gravitational settling', the large particles ($D_p > 20 \mu\text{m}$) settle on the Earth's surface due to the force of gravity acting on these. If a box model is applied to study aerosol evolution by gravity, calling h the height of the box (this will be the mixing layer height in the atmosphere), the dynamic equation of the gravitational settling is [6]:

$$\frac{dN_v(v,t)}{dt} = -\frac{v_g}{h} \cdot N_v(v,t) \quad (5)$$

3 Numerical method

The exact solutions serve as benchmarks, and together with the approximate solutions, are useful in parametric studies as well as for providing insight into the nature of aerosol evolution. Their utility for practical use is limited for several reasons:

Exact solutions are feasible only for special, and not necessarily realistic, representations of the rate processes and for certain initial and boundary conditions.

Aerosol evolution, especially if there are abrupt changes such as source reinforcement, may not follow a prescribed form of distribution. Often bimodal, trimodal, or more complicated distributions may evolve, and these can not be dealt with in the context of log-normal, gamma, or other forms.

In many applications, those in nuclear industry in particular, particle evolution is strongly coupled to the environment. Thus, the dynamical equation must be solved simultaneously with equations that describe the thermal hydraulic and chemical conditions of the gaseous/vaporous environment of the particles.

For practical applications, we must resort to numerical methods. From a numerical viewpoint, at least in principle, the dynamical equation is too difficult to solve. Since methods for solving partial differential equations (PDEs) have been studied extensively and many algorithms and computer programs are available, the preferred approach here has been to reduce the general dynamical equation (GDE) to a system of ordinary differential equations by employing some approximation to the integral of the coagulation terms [12].

The integral can be evaluated (approximated) in various ways. The approximations are likely to be most successful if they preserve the basic properties of coagulation. One such property is the conservation of mass or volume in coagulation. The various quadratures must either be accurate enough to preserve this property or one must preserve it forcibly by evaluating the integrals in a special manner or by applying corrective steps to the calculations.

The term of coagulation is difficult to integrate since the habitual methods (finite difference, finite elements, etc.) add numerical dispersion (spurious oscillations appear in the solution) and numerical diffusion (the

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spectra are smoothed). This term represents a non-linear integro-differential equation. Since the integrals that appear vary slowly when the particle size distribution is modified, it is possible to apply a classical method of integration for systems of ordinary differential equations [12]. In the case of stiff systems of equations it is necessary to solve the integrals numerically, so we propose to make it through a Gaussian quadrature via interpolation with cubic splines [10]. The other term of the GDE is integrated as the coagulation term. In this work we have used a method of Runge-Kutta-Fehlberg (RKF) [12]. The conjoint application of the RKF method and the denominated flux-based modified method of characteristics [13] to solve the condensation equation has been carried out by means of time splitting. This method is based on a supposition of independence of the evolutions of the different processes during a Δt time (being this is small enough), applying consecutively the suitable methods to each physical process during the time Δt .

3.1 The J -space transform and cubic spline method

This method has been used by several authors [18-19]. In particular, to describe particle size variations over several decades in size, we introduce the transformations:

$$v(J) = v_0 \cdot a^{b(J-1)} \quad (6)$$

and

$$Y(J, t) = N(v, t) v(J) b \ln(a) \quad (7)$$

where v_0 is the smallest aerosol size corresponding to $J = 1$, and a and b are some arbitrary constants. J is an integer and varies from 1 to a fairly large number that would allow consideration of all sizes of interest. With this transformation we have:

$$\begin{aligned} \frac{\partial}{\partial t} Y(J, t) = & \int_1^{J_d} dJ' \frac{v(J)}{v(J_c)} Y(J_c, t) K(J_c, t) Y(J', t) - \int_1^{\infty} dJ' Y(J, t) K(J, J') Y(J', t) \\ & - \frac{\partial}{\partial J} \left[\frac{I(v(J))}{v(J) b \ln(a)} Y(J, t) \right] + \tilde{S}(v(J), t) \end{aligned} \quad (8)$$

where:

$$v(J_c) = v(J) - v(J') \quad (9)$$

$$J_c = J + \frac{1}{b \ln(a)} \ln(1 - a^{(J'-J)b}) \quad (10)$$

and

$$J_d = J - \frac{\ln(2)}{b \ln(a)} \quad (11)$$

Equation (8) is converted to a system of ordinary differential equations by use of quadrature formulas for the integrals and cubic splines for the derivative on J [10] and we write:

$$\frac{\partial}{\partial t} Y(J, t) = f_J(Y(1, t), Y(2, t), Y(3, t), \dots, Y(m, t), \tilde{S}(J, t)) \quad (12)$$

together with the associated initial conditions. Here, $J = 1, 2, 3, \dots, m$, where m is chosen sufficiently large so that all particles of interest are covered. The method gives rise to fairly accurate results, but the computational costs can be quite high because of the repeated cubic spline fits. Note that we have taken $a = 2$ and $b = 1/2$.

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3.2 Time splitting

The integration of the aerosol evolution equation (GDE) can not be carried out by applying a unique method. The basic idea of operator splitting, which is also called *time splitting* or *the method of fractional steps* [12], is the following one. Suppose we have an initial value equation of the form:

$$\frac{\partial u}{\partial t} = Lu \quad (13)$$

where L is some operator. While L is not necessarily linear, suppose that it can at least be written as a linear sum of m pieces, which act additively on u ,

$$Lu = L_1 u + L_2 u + \dots + L_m u \quad (14)$$

Finally, suppose that for each of the pieces, we already know a differencing scheme for updating the variable u from the time step n to time step $n + 1$, valid if that piece of the operator were the only one on the right-hand side. We will write these updates symbolically as:

$$\begin{aligned} u^{n+1} &= U_1(u^n, \Delta t) \\ u^{n+1} &= U_2(u^n, \Delta t) \\ &\dots \\ u^{n+1} &= U_m(u^n, \Delta t) \end{aligned} \quad (15)$$

Now, one form of operator splitting would be to get from n to $n + 1$ by the following sequence of updates:

$$\begin{aligned} u^{n+(1/m)} &= U_1(u^n, \Delta t) \\ u^{n+(2/m)} &= U_2(u^{n+(1/m)}, \Delta t) \\ &\dots \\ u^{n+1} &= U_m(u^{n+(m-1)/m}, \Delta t) \end{aligned} \quad (16)$$

For example, a combined advective-diffusion equation, such as:

$$\frac{\partial u}{\partial t} = -v \frac{\partial u}{\partial x} + D \frac{\partial^2 u}{\partial x^2} \quad (17)$$

use an explicit scheme for the advective term combined with Crank-Nicholson or other implicit scheme for the diffusion term [9].

Other form of operator splitting with a slightly different twist would be to reinterpret the previous equations to have a different meaning. Let U_1 now denote an updating method that includes algebraically all the pieces of the total operator L , but which is desirably stable only for the L_1 piece. Likewise U_2, \dots, U_m . Then a method of getting from u^n to u^{n+1} is:

$$\begin{aligned} u^{n+(1/m)} &= U_1(u^n, \Delta t / m) \\ u^{n+(2/m)} &= U_2(u^{n+(1/m)}, \Delta t / m) \end{aligned} \quad (18)$$

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$$u^{n+1} = U_m \left(u^{n+(m-1)/m}, \Delta t / m \right)$$

The time step for each fractional step in equations (18) is now only $1/m$ of full time step, because each partial operation acts with all the terms of the original operator [17].

3.3 Numerical method to solve the condensation equation

In order to solve the condensation equation we will be based on the denominated flux-based modified method of characteristics [5, 13]. Now we introduce the notion of first-order partial differential equations and an important technique for solving initial-value problems: *method of characteristics*. The problem we will solve is the initial-value problem:

$$PDE \quad a(x,t) \frac{\partial u}{\partial x} + b(x,t) \frac{\partial u}{\partial t} + c(x,t)u = 0 \quad -\infty < x < +\infty, \quad 0 < t < +\infty \quad (19)$$

$$IC \quad u(x,0) = \phi(x) \quad -\infty < x < +\infty \quad (20)$$

where *PDE* means partial differential equation and *IC* means initial condition. Note that we are solved problems with variable coefficients. It turns out that if we change coordinates from (x, t) to appropriate new coordinates (s, τ) (characteristic coordinates), then our differential equation becomes an ordinary differential equation. Hence, we solve the ordinary differential equation (ODE) to find $u(s, \tau)$, and then the last step is to plug in the values of s and τ in terms of x and t to get $u(x, t)$. If we recall when the diffusion equations is solved:

$$\frac{\partial u}{\partial t} = \alpha^2 \frac{\partial^2 u}{\partial x^2} - v \frac{\partial u}{\partial x} \quad -\infty < x < +\infty, \quad 0 < t < +\infty \quad (21)$$

the constant α^2 stood for the amount of diffusion, while v stood for the velocity of the medium. Hence, if $\alpha = 0$ (no diffusion), it is clear (since we only have convection) that the solution travel along the x -axis with velocity v . In other words, if the initial solution is $u(x, 0) = \phi(x)$, then the solution to:

$$\frac{\partial u}{\partial t} = -v \frac{\partial u}{\partial x} \quad -\infty < x < +\infty, \quad 0 < t < +\infty \quad (22)$$

would be $u(x, t) = \phi(x - vt)$. So we can think of the first-order equation:

$$a(x,t) \frac{\partial u}{\partial x} + b(x,t) \frac{\partial u}{\partial t} = 0 \quad (23)$$

as the concentration along a stream where the velocity is given by:

$$v = -a(x,t)/b(x,t) \quad (24)$$

Of course, if $a(x, t)$ and $b(x, t)$ are constants, then we have travelling wave solutions. On the other hand, if $a(x, t)$ and $b(x, t)$ change in x and t , then the stream velocity varies as we go along the stream and with the time. If we get back to our basic problem (19), the solution to this equation is based on a physical fact, namely, that an initial disturbance at some point x propagates along a line in the tx -plane called a characteristic. This phenomenon contrasts with many other equations (such as the heat equation) where an initial disturbance at a point affects the solution everywhere else later in time. With this in mind, the idea is to introduce two new

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coordinates s and τ (to replace x and t) that have the properties: (a) s will change along the characteristic curves, and (b) τ will change along the initial curve (most likely the line $t = 0$).

First, we consider the new coordinate s . By choosing s having the above property, the partial differential equation (PDE):

$$a(x,t) \frac{\partial u}{\partial x} + b(x,t) \frac{\partial u}{\partial t} + c(x,t)u = 0 \quad (25)$$

is transformed into the ordinary differential equation (ODE):

$$\frac{du}{ds} + c(x,t)u = 0 \quad (26)$$

Of course, the question is how to find these characteristics. The answer is simple. We pick the characteristic curves $\{[x(s), t(s)]: 0 < s < +\infty\}$ so that:

$$\frac{dx}{ds} = a(x,t) \quad (27)$$

$$\frac{dt}{ds} = b(x,t) \quad (28)$$

By doing this, it is clear that:

$$\frac{du}{ds} = \frac{\partial u}{\partial x} \frac{dx}{ds} + \frac{\partial u}{\partial t} \frac{dt}{ds} = a(x,t) \frac{\partial u}{\partial x} + b(x,t) \frac{\partial u}{\partial t} \quad (29)$$

In other words, along the curves $\{[x(s), t(s)]: 0 < s < +\infty\}$, the partial differential equation (PDE) becomes an ordinary differential equation (ODE). A detailed analysis of this method as well as its range of application can be found in Reference [5].

4 Analysis of results

The selective scavenging efficiencies have been determined for coagulation, condensation and gravitational settling of atmospheric aerosols with several typical size distributions labelled as [16]: (1) marine background (MB), (2) clean continental background (CCB), (3) average background (AB), (4) background and aged urban plume (BAUP), (5) background and local sources (BLS), (6) urban average (UA), (7) urban and freeway (UF) and (8) central power plant (CPP). These names refer to the atmospheric environments which characterize them. The size range is divided into three well-defined subintervals referred to the literature as: (a) Aitken nuclei, (b) accumulation mode, and (c) coarse mode. The mean geometrical diameter, the geometrical Standard deviation and the overall volume for these PSD's can be consulted in Table 1 [16]. The 'scavenging efficiency' is defined by [7, 14]:

$$\eta(D_p, t) = \frac{n(D_p, 0) - n(D_p, t)}{n(D_p, 0)} = 1 - \frac{n(D_p, t)}{n(D_p, 0)} \quad (30)$$

where $n(D_p, 0)$ is the initial PSD of aerosol particle concentration (particle $\mu\text{m}^{-1} \text{cm}^{-3}$) and $n(D_p, t)$ is the PSD of aerosol particle concentration at the end of time t . A temperature of 293 K and an atmospheric pressure of 1 atm are assumed in all calculations. The results for other close temperatures and pressures do not differ much from those obtained with the values taken. In this work, the evolution of volume particle size distribution has been calculated at different time intervals (6, 12 and 18 hours), for the eight above-mentioned environments, due to gravitational settling as the main dynamical mechanism of aerosol removal in atmosphere.

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Table 1. Size distributions (log-normal) of different classes of atmospheric aerosols.

5 Health impact

The effective PSD was determined considering retention by size in the human pulmonary region (alveolar region and tracheo-bronchial tree) for mouth breathing [2]. This can be approximately established as a function of inhaled particle diameter. A simplification of retention by size, suitable for our computations, is shown in Fig. 1 [8, 14].

With mass respirable sampling, an attempt is made to separate the aerosol into two fractions representing the mass that would be deposited in the alveolar region and the mass that would not be deposited in this region. For that purpose, it is necessary to define the size distribution of particles deposited in the alveolar region. This material is defined as *respirable dust*. There are several definitions of respirable dust available. In 1952, the British Medical Research Council (BMRC) defined the respirable fraction (RF) in terms of the particle aerodynamic diameter, D_{pi} (μm), and it is calculated from the expression [14]:

$$RF_{i-BMRC} = 1 - \frac{D_{pi}^2}{50} \quad (31)$$

In 1968, the American Conference of Government Industrial Hygienists (ACGIH) defined the respirable dust fraction [14]:

$$RF_{i-BMRC} = 10^{0.325 - 0.185 D_{pi}} \quad (32)$$

In 1982, the United States Environment Protection Agency (USEPA) [9] defined the respirable dust fraction as a function of the particle aerodynamic diameter by means of a semi-empirical curve which is also shown in Fig. 1.

Fig. 1. Filters: Settled fraction of monodisperse aerosol in the pulmonary region as a function of particle diameter (from data in References [8] and U.S. EPA [9]).

The form of the first two filters (BMRC and ACGIH) indicates that for the given aerosol the BMRC definition will indicate slightly more respirable mass than the ACGIH definition [14]. Moreover, because retention under a $2 \mu\text{m}$ particle diameter is not well defined in the first two filters, the respirable fraction measured according to the BMRC and ACGIH criteria would be smaller than results obtained with the USEPA filter.

The effect of retention filters differs greatly. The BMRC respirable dust and the USEPA respirable dust have been represented in Figs. 2 and 3, at the initial time and after 6, 12 and 18 hours of gravitational settling in an urban environment (UA), respectively.

Fig. 2. Evolution of respirable dust in an urban environment (UA) as a function of elapsed time by coagulation, condensation and gravitational settling for BMRC filter.

Fig. 3. Evolution of respirable dust in an urban environment (UA) as a function of elapsed time by coagulation, condensation and gravitational settling for USEPA filter.

With BMRC filter, the accumulation mode of the urban environment remains roughly unaltered with regard to its mean geometrical diameter and its mass, and the coarse mode decreases in height because of the main effect to the gravity force (see Fig. 2). However, with USEPA filter, we can observe that gravitational settling affects to the coarse mode mainly while the accumulation mode remains unchanged. In any case, its effect is of an essential importance (see Fig. 3).

The numerical results obtained regarding overall efficiencies of removal can be seen in Table 2. The first interesting result is: only 25.6% of initial urban aerosol volume (UA) is settled in the pulmonary region

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without mechanisms of removal (scavenging mechanisms), that is to say, without coagulation, condensation and gravitational settling (with USEPA filter). After 18 hours of gravitational settling, this percentage of respirable dust decreases to 14.3%. This is an important result from a health perspective. Therefore, gravitational settling is the main mechanism yielding lower retention efficiencies in the lungs over time.

Table 2. Percentages of respirable dust versus overall initial volume and versus respirable initial volume, during 6, 12 and 18 hours of coagulation, condensation and gravitational settling.

6 Conclusions

A simple linear fit of data appearing in Table 2 for the USEPA filter indicates that gravitational settling keep on the straight line of equation $p(\%) = -3.7t + 28.6$, where $p(\%)$ is the percentage of respirable dust and t is the time in hours (6, 12 and 18 hours in this case). The correlation coefficient (measuring the goodness of fit) is $r^2 = 0.99$ so that the fit can be considered as very good. Hence, the main mechanism to remove respirable dust is gravitational settling since in all eight atmospheric environments studied, the presence of gravitational settling decreases greatly the initial respirable dust volume with respect to that after 18 hours of gravitational settling (see Table 2).

A computational procedure has been developed based on the general-purpose code developed for us for modelling and simulating the deposition by coagulation, condensation and gravitational settling of aerosol particles for eight different environments and in order to calculate the percentage of respirable dust versus respirable initial volume as a function of elapsed time. The findings of this study suggest that it may be possible to devise a practical procedure for establishing a removal's model by using a combined experimental/computational approach.

Once deposited, particles are retained in the lung for varying times, depending on their physicochemical properties, their location within the lung, and the type of clearance mechanism involved. It is known that insoluble particles deposited in alveolar region are cleared very slowly, over a period of months or years. Dissolved particles pass through the thin alveolar membrane into the bloodstream. Solid particles may dissolve slowly or be engulfed by alveolar macrophages (phagocytic cells) and dissolved or transported to lymph nodes or mucociliary escalator. Fibrogenic dusts, such as silica, interfere with this clearance mechanism and cause gradual scarring or fibrosis of the alveolar region. This study has tried to determine the respirable dust comparing to the initial respirable dust with the respirable dust after 6, 12, and 18 hours of coagulation, condensation and gravitational settling for eight characteristic atmospheric environments, given place to the results exposed above.

Aerosol removal by coagulation, condensation and gravitational settling is more efficient in urban environments (UA) as in other environments characterized as clean areas (CCB), due to the large relative volume of coarse mode in the urban case with respect to the other modes (accumulation mode and Aitken mode). In an urban environment (UA) for longer times, many aerosol particles of coarse mode are scavenged, but the Aitken nuclei and accumulation mode are scavenged with a great difficulty by coagulation, condensation and gravitational settling.

The equation describing the behaviour of aerosols is a non-linear, integro-differential equation of considerable complexity [4, 12]. If all of deposition, condensation, rainout, coagulation mechanisms, etc., are included, it is clear that the only feasible method of solution is a numerical one. Indeed, in practical calculations this is the procedure and many computer codes exist which solve the dynamic equation using finite difference or finite element methods.

Our program can provide useful results when one species is dominant, the particles are nearly spherical, the aerosol is homogeneous, temporal variations are not large, and the coupling with the surroundings is weak. Use of the available programs for analysis or design of experiments or natural/industrial processes must be undertaken with considerable care. It is important to select a method and rate process models appropriate to the

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problem as the computed results are dependent on these. A major drawback of numerical methods is the vast amount of computer time necessary to obtain a useful of all the relevant parameters. This causes difficulty in gaining physical insight into the problem. For this reason a variety of approximate techniques have been developed which enable analytic solutions to be able. Such techniques use the complementary methods of kernel approximation and equation approximation or a combination of both.

The numerical simulation of fluid mechanics problems has become a routine part of engineering practice as well as a focus for fundamental and applied research. Though there are still various topical areas where our physical understanding and ineffective numerical algorithms limit the investigation, a large number of complex phenomena can now be confidently studied via numerical simulation. Therefore, the model developed in this work is certainly incomplete, and it can be used as one part of a more extensive one that takes into account other phenomena (i.e. rainout, washout, phoretic phenomena, etc.) in order to forecast the real evolution of aerosols PSD's [3, 6]. A big number of computer programs have been written that attempt to solve various versions of the dynamical equations by using some of the methods described above [9, 19]. Improvements in computer hardware and system software (e.g., powerful workstations and window environments) have contributed significantly to streamlining the numerical process. Thus, these programs provide results with a good numerical accuracy for prescribed forms of the rate processes and for single component aerosols and so to integrate the overall mathematical model of the atmosphere.

Acknowledgements

The authors express deep gratitude to Department of Mathematics and Department of Construction at Oviedo University for their computational support and useful assistance. Helpful comments and discussion are gratefully acknowledged. This work has been partially supported by the basic research grant CN-04-209.

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Figure Caption

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Mathematical study of the selective removal of atmospheric aerosols by coagulation, condensation and gravitational settling

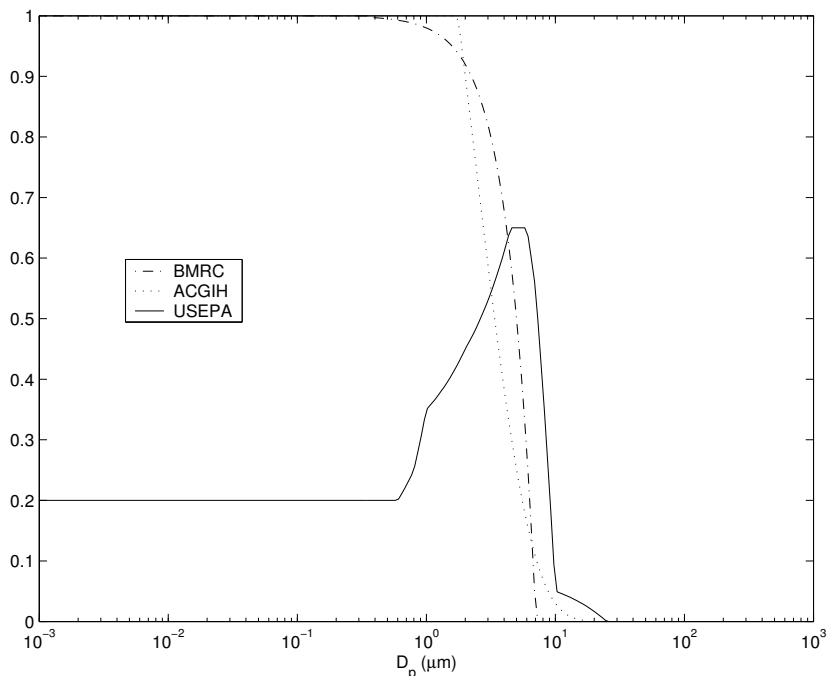


Fig. 1. Filters: Settled fraction of monodisperse aerosol in the pulmonary region as a function of particle diameter (from data in References [8] and U.S. EPA [9]).

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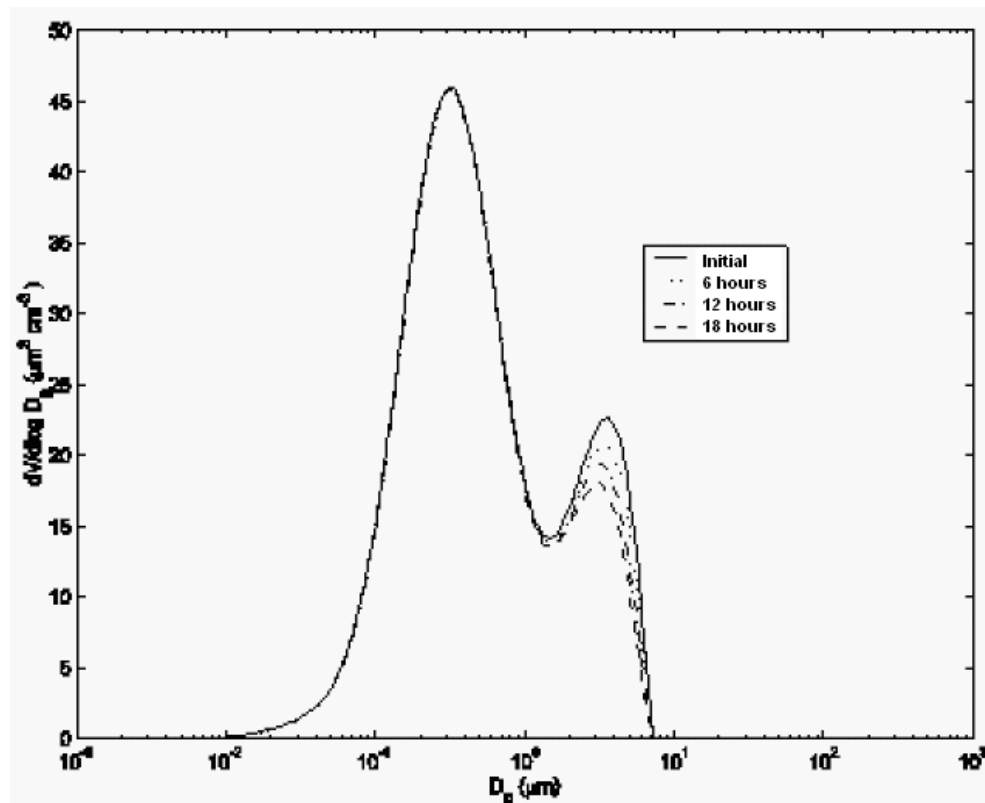


Fig. 2. Evolution of respirable dust in an urban environment (UA) as a function of elapsed time by coagulation, condensation and gravitational settling for BMRC filter.

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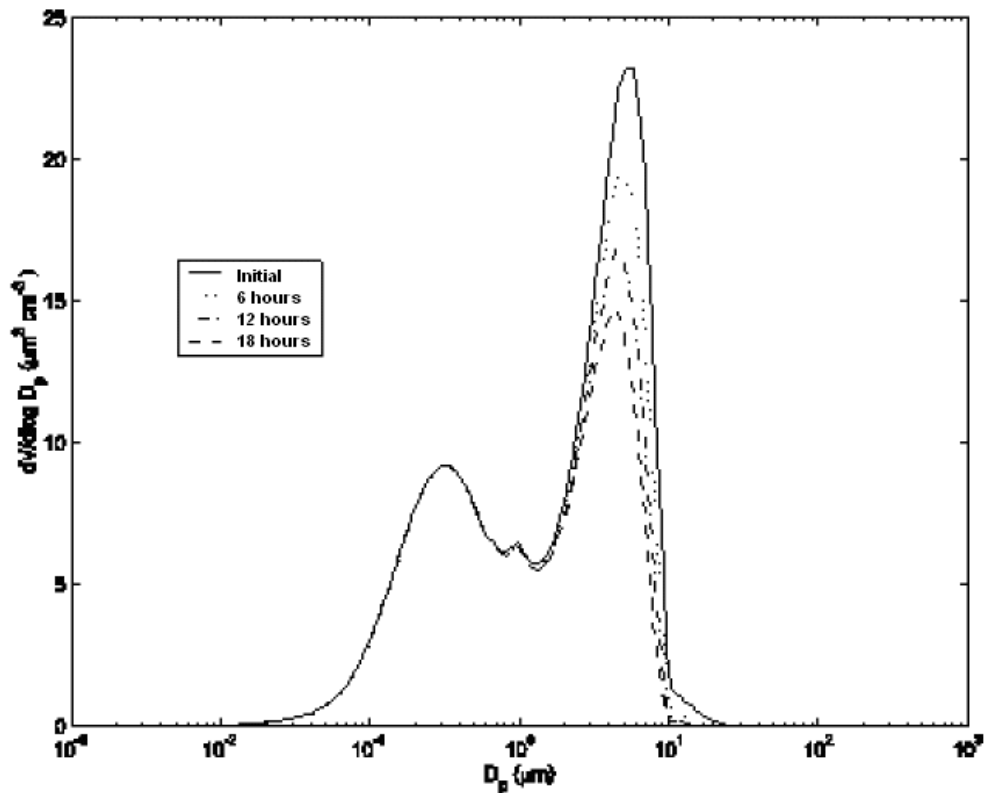


Fig. 3. Evolution of respirable dust in an urban environment (UA) as a function of elapsed time by coagulation, condensation and gravitational settling for USEPA filter.

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Table Caption

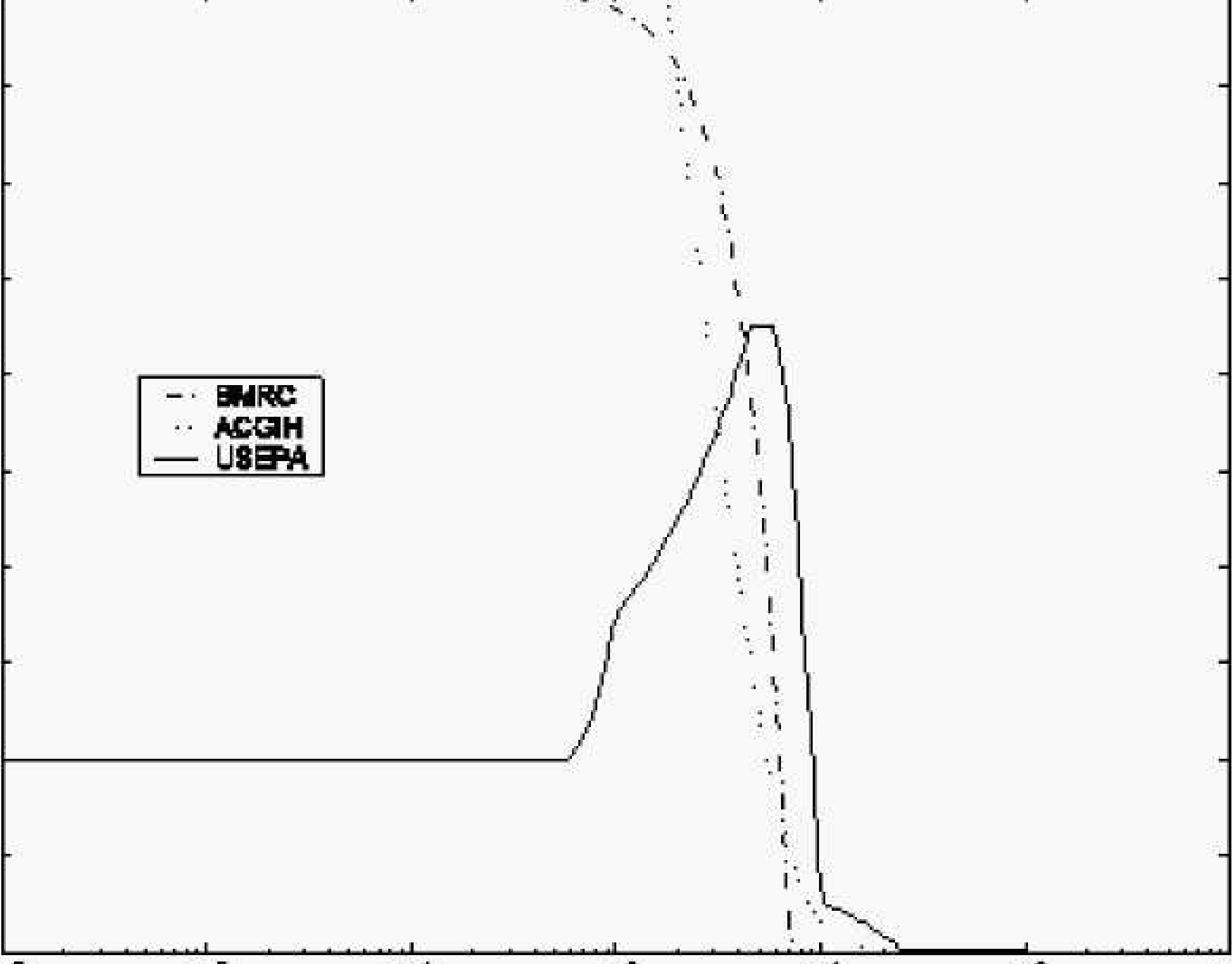
Table 1. Size distributions (Log-Normal) of different classes of atmospheric aerosol.

	Nuclei Mode			Accumulation Mode			Coarse Mode		
	D_{pg} (μm)	σ_g	V ($\mu\text{m}^3 \text{cm}^{-3}$)	D_{pg} (μm)	σ_g	V ($\mu\text{m}^3 \text{cm}^{-3}$)	D_{pg} (μm)	σ_g	V ($\mu\text{m}^3 \text{cm}^{-3}$)
Environment	0.019	1.6	0.0005	0.3	2.0	0.10	12.0	2.7	12.0
Marine background	0.03	1.6	0.006	0.35	2.1	1.5	6.2	2.2	5.0
Clean continental background	0.034	1.7	0.037	0.32	2.0	4.45	6.04	2.16	25.9
Average background	0.028	1.6	0.029	0.36	1.84	44.0	4.51	2.12	27.4
Background and aged urban plume	0.021	1.7	0.62	0.25	2.11	3.02	5.6	2.09	39.1
Background and local sources	0.038	1.8	0.63	0.32	2.16	38.4	5.7	2.21	30.8
Urban average	0.032	1.74	9.2	0.25	1.98	37.5	6.0	2.13	42.7
Urban and freeway	0.015	1.5	0.1	0.18	1.96	12.0	5.5	2.5	24.0
Central Power Plant									

Table 2. Percentages of respirable dust versus overall initial volume and versus respirable initial volume, during 6, 12 and 18 hours of coagulation, condensation and gravitational settling.

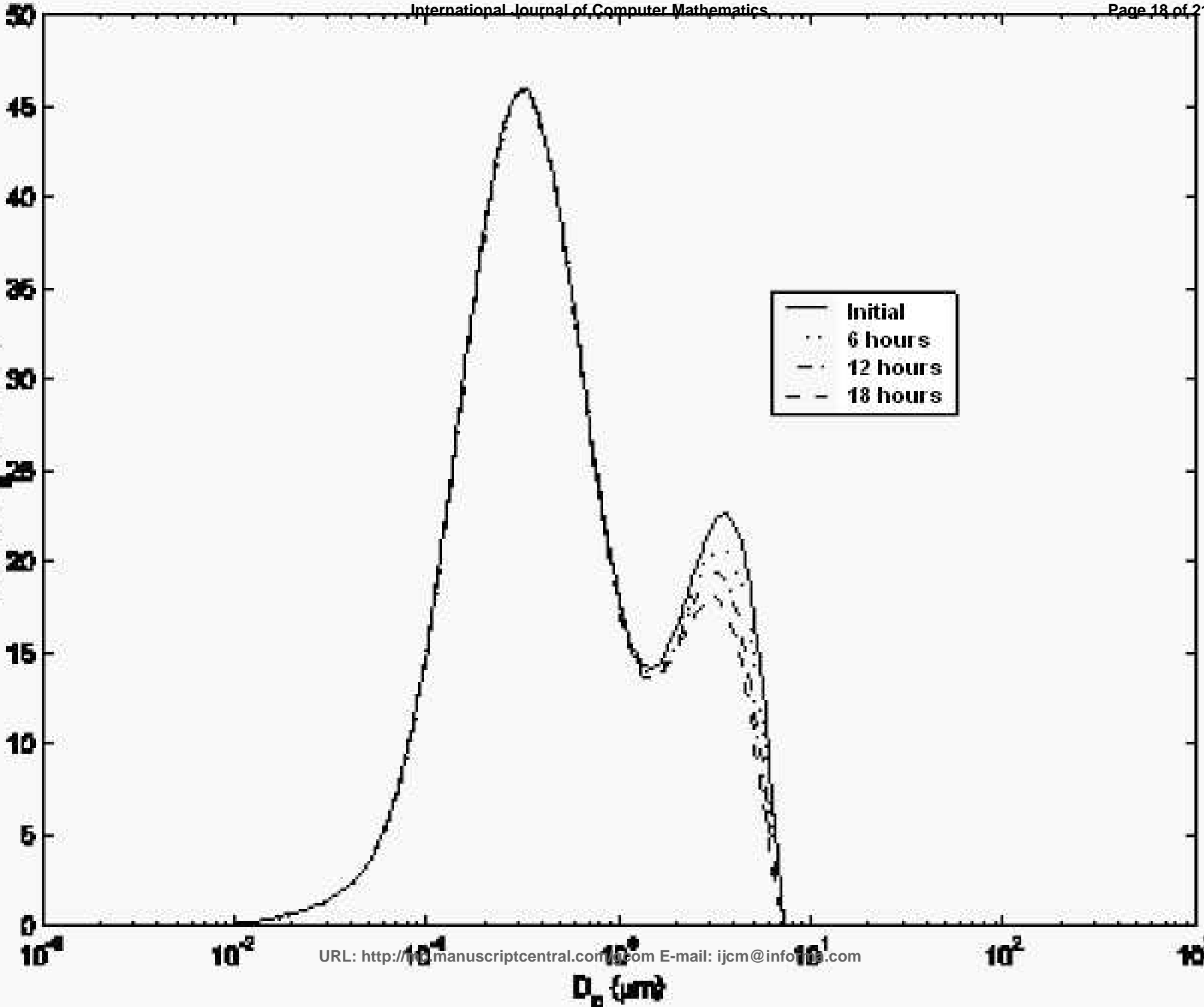
<i>Environment/Filter</i>	Initial	6 hours	12 hours	18 hours
MB/BMRC	4.8/100	4.3/25.8	3.8/23.0	3.4/20.6
MB/ACGIH	4.0/100	3.3/24.2	2.9/20.9	2.5/18.4
MB/USEPA	9.7/100	7.0/20.9	5.4/16.3	4.4/13.1
CCB/BMRC	17.6/100	16.0/106.1	14.5/96.5	13.3/88.2
CCB/ACGIH	14.0/100	12.4/103.4	11.1/92.9	10.1/84.7
CCB/USEPA	25.6/100	19.9/91.2	16.1/73.9	13.4/61.4
AB/BMRC	16.5/100	14.9/19.6	13.4/17.7	12.2/16.1
AB/ACGIH	12.9/100	11.3/19.0	10.1/17.0	9.1/15.3
AB/USEPA	24.8/100	19.3/16.9	15.6/13.7	12.9/11.4
BAUP/BMRC	33.8/100	31.5/22.9	29.6/21.5	27.9/20.3
BAUP/ACGIH	28.3/100	26.3/22.9	24.8/21.5	23.5/20.4
BAUP/USEPA	32.4/100	26.7/20.2	22.6/17.1	19.5/14.8
BLS/BMRC	18.7/100	16.8/14.0	15.1/12.6	13.7/11.5
BLS/ACGIH	14.3/100	12.5/13.7	11.1/12.1	10.0/11.0
BLS/USEPA	27.7/100	21.8/12.3	17.7/10.0	14.7/8.3
UA/BMRC	22.6/100	21.0/16.6	19.5/15.5	18.3/14.5
UA/ACGIH	19.0/100	17.4/16.4	16.1/15.3	15.2/14.4
UA/USEPA	25.6/100	20.4/14.3	16.9/11.9	14.3/10.1
UF/BMRC	18.0/100	16.4/8.4	15.0/7.7	13.7/7.1
UF/ACGIH	14.4/100	12.8/8.2	11.6/7.5	10.7/6.8
UF/USEPA	24.9/100	19.5/7.2	15.8/5.9	13.2/4.9
CPP/BMRC	17.6/100	16.1/22.0	14.7/20.2	13.6/18.6
CPP/ACGIH	14.1/100	12.7/21.6	11.5/19.6	10.6/18.1
CPP/USEPA	23.0/100	18.1/19.0	14.9/15.6	12.5/13.1

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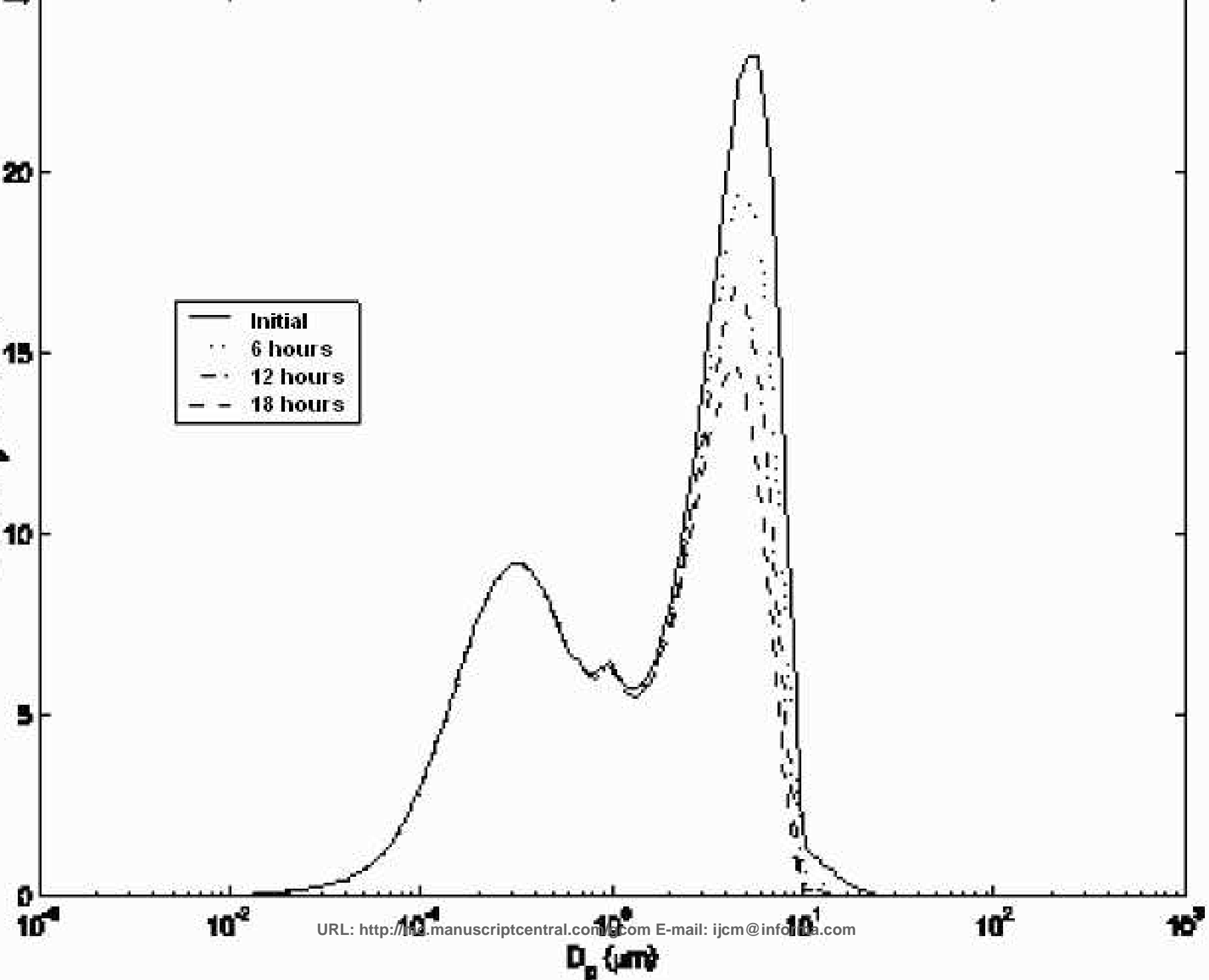


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Clean continental background	0.03	1.6	0.006	0.35	2.1	1.5	6.2	2.2	5.0
Average background	0.034	1.7	0.037	0.32	2.0	4.45	6.04	2.16	25.9
Background and aged urban plume	0.028	1.6	0.029	0.36	1.84	44.0	4.51	2.12	27.4
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Central Power Plant	0.015	1.5	0.1	0.18	1.96	12.0	5.5	2.5	24.0

Table 2. Percentages of respirable dust versus overall initial volume and versus respirable initial volume, during 6, 12 and 18 hours of coagulation, condensation and gravitational settling.

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MB/USEPA	9.7/100	7.0/20.9	5.4/16.3	4.4/13.1
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AB/BMRC	16.5/100	14.9/19.6	13.4/17.7	12.2/16.1
AB/ACGIH	12.9/100	11.3/19.0	10.1/17.0	9.1/15.3
AB/USEPA	24.8/100	19.3/16.9	15.6/13.7	12.9/11.4
BAUP/BMRC	33.8/100	31.5/22.9	29.6/21.5	27.9/20.3
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BAUP/USEPA	32.4/100	26.7/20.2	22.6/17.1	19.5/14.8
BLS/BMRC	18.7/100	16.8/14.0	15.1/12.6	13.7/11.5
BLS/ACGIH	14.3/100	12.5/13.7	11.1/12.1	10.0/11.0
BLS/USEPA	27.7/100	21.8/12.3	17.7/10.0	14.7/8.3
UA/BMRC	22.6/100	21.0/16.6	19.5/15.5	18.3/14.5
UA/ACGIH	19.0/100	17.4/16.4	16.1/15.3	15.2/14.4
UA/USEPA	25.6/100	20.4/14.3	16.9/11.9	14.3/10.1
UF/BMRC	18.0/100	16.4/8.4	15.0/7.7	13.7/7.1
UF/ACGIH	14.4/100	12.8/8.2	11.6/7.5	10.7/6.8
UF/USEPA	24.9/100	19.5/7.2	15.8/5.9	13.2/4.9
CPP/BMRC	17.6/100	16.1/22.0	14.7/20.2	13.6/18.6
CPP/ACGIH	14.1/100	12.7/21.6	11.5/19.6	10.6/18.1
CPP/USEPA	23.0/100	18.1/19.0	14.9/15.6	12.5/13.1