

THE EFFECT OF REDISTRIBUTION ON AEROSOL REMOVAL RATES

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Abstract—A simplified model is constructed for an aerosol interacting with a vapour–gas mixture in a well-mixed cavity. An accurate form is deduced for the initial equilibration timescale between the aerosol and the surrounding mixture, and the corresponding supersaturation is obtained. Analytic estimates are also given for the timescales of gravitational settling, Ostwald ripening from the Kelvin effect, and redistribution of water mass in a hygroscopic salt aerosol.

Calculations are performed for a water aerosol at 20°C and 100°C, and the effect on the gravitational fallout is investigated of redistribution due to an aerosol solute content and of radiative redistribution. The solute effect maintains a disperse aerosol size distribution and enhances settling to a moderate but significant extent. For radiative cooling alone, the redistribution hardly affects the settling at 100°C, although it also maintains a wide size distribution. However, when coupled with heating to maintain a constant temperature, the radiative redistribution is shown to be capable of causing a rapid reduction in the mass of the airborne aerosol.

NOMENCLATURE

A	constant in growth rate equation
A_b	area of base of containment
b	coefficient of solute term in \dot{R}
B_1, B_2, B_3	constants defined by equations (49) and (52)
c	vapour mass concentration
\bar{c}_p	mean specific heat at constant pressure
C	salt volumetric concentration
C_n	condensation number
D	vapour–gas diffusivity
$f(R)$	function of R
g	gravitational acceleration
$G(R)$	dimensionless radiative growth function
G_2	coefficient of R specifying $G(R)$ for large drops
h	cavity height
i	van't Hoff factor
k	thermal conductivity
\bar{K}	constant specifying supersaturation differences
L	latent heat of vaporization
Le	Lewis number, $Le = k/(\rho_p D)$
m_s	mass of solute in droplet
\dot{m}_v	total local aerosol condensation rate
$n(R)$	aerosol number density
N	total aerosol number density
p	total pressure
q	external heat transfer rate into cavity
Q_R	total radiative heat flux from droplet
R	droplet radius
\dot{R}	droplet growth rate
R_g	parameter of log-normal distribution
R_G	intermediate radius
R_{GC}	gas constant
$\overline{R^n}$	n th moment of the aerosol size distribution
R_s	radius of solute mass
R_γ	Kelvin radius ($2\mu_v/(\rho_l R_{GC} T)$)
S	vapour saturation
S_A	sticking probability
S_∞	asymptotic saturation in time
t	time
t_E	equilibrium timescale
$t_{1/2}$	half-life for gravitational sedimentation

t_{OR}	Ostwald ripening timescale
t_{sol}	solute redistributive timescale
T	temperature
u	difference between vapour concentration and its equilibrium value
$v(R, t)$	growth velocity ($= \dot{R}$)
V	volume
$y(R_0, t)$	partial derivative of R with respect to R_0
α	coefficient in exponent (see equation (10))
β	coefficient giving temperature derivative of $p_{ve}(T)$
η	gas viscosity
γ	surface tension for Kelvin radius
γ_N	numerical factor in Ostwald ripening solution
$\lambda(R)$	gravitational settling loss rate
λ_0	constant specifying R dependence of $\lambda(R)$
μ	(subscripted) molecular weight
ρ	total density
σ_g	width parameter in log-normal distribution
ΔT	temperature difference specifying radiative cooling

Subscripts

e	pertaining to equilibrium
g	pertaining to gas
i	pertaining to initial value for solute effect
l	pertaining to liquid
s	pertaining to solute
v	pertaining to vapour
0	pertaining to initial value
∞	pertaining to asymptotic value

Superscripts

'	derivative with respect to T
—	average over the size distribution

1. INTRODUCTION

Water clouds are common in nature and aerosols with large water contents could be formed in the containment of a pressurized water reactor (PWR) following a possible accident, as shown in some large-scale experiments (Schöck *et al.*, 1988). Water droplets can grow or evaporate not only as a result of supersaturation in the surrounding vapour, but also through their size (Kelvin effect), salt content (solute effect) and their interaction with radiation (see e.g. Manton, 1983). Even when a water cloud is in overall equilibrium with the medium, all these effects can cause the redistribution of water mass between cloud droplets (Barrett and Clement, 1988). We have examined in some detail the redistribution due to the interaction with radiation (Barrett and Clement, 1990). Here, we extend the examination to the other mechanisms, particularly the solute effect, as well as calculating the effect of the redistribution on the gravitational settling rate of the aerosol.

In section 2 we describe theoretically vapour–aerosol interactions for an idealized well-mixed cavity which can exchange heat with its surroundings, but whose only mechanism for loss of vapour–aerosol mass is gravitational settling. The basic idea is to calculate how an aerosol size distribution evolves with time, and the necessary data for actual calculations are given for a water aerosol at 20°C and 100°C. The main theoretical development is to derive precisely the timescales for fast transients which bring the aerosol–vapour–gas system into approximate equilibrium and the resulting equilibrium supersaturations. Slower redistributive processes then take place and we show how the consequent changes in supersaturation are given by mass conservation and moments of the size distribution. A method is given for calculating these moments using the original size distribution.

In section 3 we give some results for each of the redistributive mechanisms with an emphasis on the timescales involved. A simple half-life is given for gravitational settling and explicit timescales are obtained for Ostwald ripening (Kelvin effect) and for the solute effect. Redistribution will take place with a hygroscopic aerosol when circumstances such as a sudden injection of steam allow pure growth of droplets first with a saturation above its

equilibrium value (Clement, 1988b). In such a case we show that the final equilibrium aerosol size distribution preserves a log-normal form of the initial distribution.

In the radiative case considered in section 3.4 we obtain results for pure radiative cooling, when the temperature of the cavity falls, and when radiative cooling is combined with internal heating to keep the temperature constant, the main case considered previously (Barrett and Clement, 1990). The amount of aerosol removal by settling is found to depend strongly on redistribution only in the second case. In the solute case with removal discussed in section 3.5, effects of redistribution are found to occur only when the removal timescale is comparable to the redistribution timescale.

Finally, in section 4 we draw some conclusions from this work, especially regarding calculations for water aerosols in PWR containments.

2. VAPOUR-AEROSOL INTERACTIONS

The simplified problem that we wish to examine is a cavity volume V with base area A_b and height h , containing a uniform mixture of a gas, a condensible vapour at a concentration $c = \rho_v/\rho$, and an aerosol with a size distribution $n(R, t)$, where R is the radius. The only aerosol removal mechanism that we include is gravitational settling.*

First, we write down basic equations for the system, some of which are obtained from previous work on droplet growth (Barrett and Clement, 1988) and on aerosol formation (Clement, 1985, 1988), although, because of the simplifications made in the problem, they can easily be obtained *ab initio*. In section 2.3 we derive an equation for the supersaturation which clearly shows the need to divide the problem into two steps, one of equilibration with a short timescale and then one of mass redistribution. It is the latter problem that we are mainly interested in here, but, to avoid fast transients, it is necessary to choose carefully the initial supersaturation. Some results on the evolution of the aerosol size distribution are obtained in section 2.4.

2.1. Basic equations

The vapour losses mass to the aerosol at a mass density growth rate, \dot{m}_v , so that its concentration, c , obeys the exact equation (c not necessarily small):

$$\frac{dc}{dt} = -(1-c)\frac{\dot{m}_v}{\rho}. \quad (1)$$

Latent heat, L , is released on vapour condensation so that the cavity temperature, T , satisfies

$$\rho \bar{c}_p \frac{dT}{dt} = L\dot{m}_v + q, \quad (2)$$

where \bar{c}_p is the mean specific heat of the contents and q is an additional source or sink term to be chosen later. Any effects of pressure changes resulting from vapour removal or temperature change can be included by an appropriate choice of q , but, for most of the redistributive cases we consider, pressure changes will not occur or will be negligible.

The aerosol size distribution satisfies

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial R}(n\dot{R}) = -\lambda(R)n, \quad (3)$$

where, for gravitational settling from a well-mixed volume V onto a surface A_b with $h = V/A_b$, the loss rate is given by

$$\lambda(R) = \frac{2\rho_c g R^2}{9\eta h} \equiv \lambda_0 R^2. \quad (4)$$

* No vapour loss is allowed, other than to the aerosol, but in some cases we do allow for heat sources or sinks in the system. This is necessary to give a realistic treatment of radiative redistribution and removal.

A form for \dot{R} which includes the Kelvin effect, the interaction with radiation, and the effect of dissolved solute to the lowest order is (Barrett and Clement, 1988)

$$\dot{R} = \frac{A}{R + R_{\min}} \left[S - 1 - \frac{R_y}{R} + \frac{bR_s^3}{R^3} + G(R) \right], \quad (5)$$

where S is the saturation given by

$$S = \frac{p_v}{p_{ve}(T)} = 1 + \frac{\mu_v}{\mu_v - (\mu_v - \mu_g)c} \frac{u}{c_e(T)}. \quad (6)$$

Here, S is also given in terms of the equilibrium and actual vapour concentrations, $c_e(T)$ and c and the molecular weights, μ .

A factor R_{\min} has been included in the denominator in equation (5) as a reminder that the $1/R$ dependence of \dot{R} does not extend to very small R (Barrett and Clement, 1988). With its inclusion the following expression for A , valid in the continuum region for growth, must be amended. Values of R_{\min} are quoted in the next section, but we are mainly interested in aerosols with $R \geq 1 \mu\text{m}$, so that for the sake of simplicity, R_{\min} will be omitted from subsequent expressions. From our previous work

$$A = \frac{Dp\rho_{ve}(T)}{\rho_l(p - p_{ve}(T))} \frac{\text{Cn}(1 - c_e)}{\text{Cn}(1 - c_e) + 1} = \frac{kT^2}{\rho_l\beta L} \frac{1}{1 + \text{Cn}(1 - c_e)}, \quad (7)$$

where $\beta = \mu_v L / R_{GC}$ the exponent in the Clausius–Clapeyron relation for $p_{ve}(T)$, ρ_{ve} is the vapour density corresponding to p_{ve} , and the condensation number (Clement, 1985) is

$$\text{Cn} = k / (LD\rho c'_e(T)). \quad (8)$$

The solute term bR_s^3/R^3 arises from the expansion of the vapour pressure change from Raoult's law modified by the van't Hoff factor, i (see, for example, Manton (1983)), so that $b = i\mu_l\rho_s/\mu_s\rho_l$.

R_s is given by the mass of solute in the droplet,

$$m_s = \frac{4}{3}\pi\rho_s R_s^3. \quad (9)$$

For high concentrations of solute, the full water activity needs to be known to specify the vapour pressure (see, for example, Clough *et al.*, 1988; Jokiniemi, 1990).

The final term, $G(R)$, in equation (5) refers to radiation from the aerosol whose redistributive effects we have recently considered (Barrett and Clement, 1990),

$$G(R) = \beta Q_R / (4\pi k T^2 R) = G_2 R (1 - \exp(-\alpha R)), \quad (10)$$

where Q_R is the net radiative heat flux from the droplet, and the final form is that found appropriate for water droplets (Roach, 1976; Barrett and Clement, 1990). The parameters G_2 and α are specified in the next section.

The only approximation regarding c and S we have made in equation (7) and make in the denominator in equation (6) is to put $c = c_e$ and $S = 1$ in the expressions $(\mu_v - \mu_g)c$ and $p - Sp_{ve}$, respectively. This is an extremely accurate approximation in problems involving rearrangement, and does not prejudice the application of the theory to situations when the vapour concentration is not small ($c < 1$).

Finally, in terms of the size distribution, the mass density growth rate of the aerosol is

$$\dot{m}_v = 4\pi\rho_l \int R^2 \dot{R} n(R, t) dR. \quad (11)$$

2.2. Data for water

We have performed calculations for water at 20°C and 100°C, most of the data used being given in Table 1. The values quoted were calculated with the value of 1 for the sticking

Table 1. Parameters for water

Quantity		$T=293\text{ K}$	$T=373\text{ K}$
A	$(\mu\text{m})^2\text{ s}^{-1}$	120	411
R_{\min}	μm	0.20	0.097
R_y	μm	0.00108	0.00071
b^*	CsI	0.54	0.54
	CsOH	0.93	0.93
G_2		1.79×10^{-4}	2.28×10^{-4}
λ_0	$(\mu\text{m})^{-2}\text{ s}^{-1}$	1.2×10^{-4}	1.2×10^{-4}
		h(m)	h(m)
$4\pi\rho_i/(3\rho c_e)$		241, 900	6715
Le		0.841	0.768
Cn		0.38	0.033
α	μm^{-1}	0.3	0.3
B_1		-0.049	-0.0064
B_2		0.951	0.994
B_3	$^{\circ}\text{C}$	3.80×10^6	1.05×10^5

* Values calculated using, for CsI, $i=1.72$, $\rho_s=4510\text{ kg m}^{-3}$, $\mu_s=259.8$; for CsOH, $i=2.1$, $\rho_s=3675\text{ kg m}^{-3}$, $\mu_s=149.9$ (Jokiniemi, 1990).

probability, S_A , of water molecules. At 20°C a small value of S_A would increase R_{\min} (Barrett and Clement, 1988). For radiation across a small temperature difference, ΔT , to an absorbing surface, we have (Barrett and Clement, 1990):

$$G_2 = 1.2 \times 10^{-3} \left(\frac{T}{280} \right)^2 \frac{4\Delta T}{T}. \quad (12)$$

The values quoted in Table 1 correspond to $\Delta T=10^{\circ}\text{C}$.

2.3. Supersaturation and timescales

We first define various averages of the aerosol size distribution by

$$N(t) = \int n(R, t) dR, \quad (13)$$

$$\overline{Q_R(t)} = \int Q_R n(R, t) dR / N, \quad (14)$$

with similar expressions for \bar{R} , \bar{R}^2 and \bar{R}^{-2} .

We now derive an equation for $u=c-c_e(T)$. From equations (1) and (2) we obtain

$$\frac{du}{dt} = -\frac{\dot{m}_v [\text{Cn}(1-c_e-u) + \text{Le}]}{\rho} - \frac{1}{\rho L \text{Cn}} \frac{\text{Le}}{\text{Cn}} q, \quad (15)$$

The next step is to substitute in equation (11) for \dot{m}_v the expression (5) for \dot{R} and use equation (7) for A . To obtain the result in terms of u we substitute from equation (6) for $S-1$. Equation (15) for u then becomes

$$\frac{du}{dt} = -4\pi D N \bar{R} \frac{\text{Cn}(1-c_e+u) + \text{Le}}{\text{Cn}(1-c_e) + 1} \left\{ u + c_e \frac{\mu_v - (\mu_v - \mu_g)c_e}{\mu_v} \left(-\frac{R_y}{\bar{R}} + \frac{bR_s^3 \bar{R}^{-2}}{\bar{R}} + \overline{RG(R)} \right) \right\} - \frac{1}{\rho L \text{Cn}} q. \quad (16)$$

The u in the factor $\text{Cn}(1-c_e+u)$ may practically always be neglected so that this equation is a first order linear differential equation for u with an exponential equilibration

timescale given by

$$t_E = \frac{1}{4\pi DN\bar{R}} \frac{Cn(1-c_e)+1}{Cn(1-c_e)+Le} \quad (17)$$

$$\approx \frac{1}{4\pi N\bar{R}k/(\rho\bar{c}_p)}, \quad Cn \ll 1.$$

The first form is a more accurate result than a previously deduced timescale (Clement, 1988) where the second radio did not appear. This previous result applies for $Cn \gg 1$, whereas the final form is for $Cn \ll 1$, the case for water vapour in air at high temperatures. As might be expected in this case where the transmission of latent heat controls the condensation process, it is the heat transfer rate, $k/\rho\bar{c}_p$, rather than the mass transfer rate, D , which gives the equilibration timescale. The timescale is typically much smaller than a second; for example for water droplets in air at 2 bar and 100°C and an aerosol mass density of 1 gm^{-3} , we have $t_E = 0.023 \bar{R}^3/\bar{R}s$, where the radii are measured in μm . This means that, unless the RHS of equation (16) is chosen initially to vanish, there will be a fast transient with the timescale given by equation (17). Following this transient, redistribution processes and possible changes in T will lead to a much slower time dependence of all the moments occurring in (16), and consequently slow changes in u .

For the case of a heat loss by radiation from the aerosol only, we have for the source term q ,

$$q = - \int Q_R n(R) dR = -N\bar{Q}_R = 4\pi kT^2 RG(R)/\beta. \quad (18)$$

The terms in equation (16) containing q and $\overline{RG(R)}$ now combine and, omitting the Kelvin and solute terms, equation (16) can be reduced to

$$\frac{du}{dt} + 4\pi DuN\bar{R} \frac{Le + Cn(1-c_e)}{1 + Cn(1-c_e)} = \frac{(1-c_e)(Le-1)\bar{Q}_RN}{\rho L(1 + Cn(1-c_e))}. \quad (19)$$

It is easy to show that the corresponding 'equilibrium' value of u gives the supersaturation, $S-1$, and growth rate, \bar{R} , obtained previously (Barrett and Clement, 1990, equations (22) and (23)).

Another physical case corresponds to the requirement of constant temperature which we previously examined (Barrett and Clement, 1990) with a constant amount of vapour plus aerosol. This corresponds to $q = -L\dot{m}_v$, and the modifications to equations (15) and (16) are

$$\frac{du}{dt} = -(1-c_e-u) \frac{\dot{m}_v}{\rho}$$

$$= - \frac{4\pi DN\bar{R}(1-c_e)Cn}{Cn(1-c_e)+1} \left[u + c_e \frac{\mu_v - (\mu_v - \mu_g)c_e}{\mu_v} \left(-\frac{R_y}{\bar{R}} + \frac{bR_s^3\bar{R}^{-2}}{\bar{R}} + \frac{\overline{RG(R)}}{\bar{R}} \right) \right]. \quad (20)$$

The equilibrating timescale is increased from the value given by the final form in equation (17) by a factor Cn^{-1} for $Cn \ll 1$. The equilibrium corresponds to no mass transfer between the aerosol and vapour-gas mixture, i.e. $\dot{m}_v \approx 0$.

Instead of obtaining equations for u , we could work in terms of the saturation, S , when equation (15) is replaced by

$$\frac{dS}{dt} = - \frac{\mu_v}{\rho c_e Cn[\mu_v - (\mu_v - \mu_g)c]} \left\{ \left[Cn(1-c) + \frac{cLe}{c_e} \right] \dot{m}_v + \frac{c}{c_e} \frac{Leq}{L} \right\}. \quad (21)$$

This equation with $q=0$ and $c \ll 1$ was used to discuss nucleation in a vapour-gas mixture (Barrett and Clement, 1991).

2.4. Aerosol size distribution and moments

Starting from an initial size distribution $n_0(R_0, t=0)$, a formal solution of equation (3) is obtained in the Appendix:

$$n = n_0(R_0) \left(\frac{\partial R_0(R, t)}{\partial R} \right)_t \exp \left[- \int_0^t \lambda(R'(R_0, t')) dt' \right]. \tag{22}$$

The time dependence of \bar{R} is determined by moments over the distribution at time t :

$$\begin{aligned} \overline{f(R)} &= \int_0^\infty f(R) n(R) dR / N \\ &= \int_0^\infty dR_0 f(R(R_0, t)) n_0(R_0) \exp \left[- \int_0^t \lambda(R'(R_0, t')) dt' \right] / N, \end{aligned} \tag{23}$$

where we have used equation (22) to express the moment as an integral over R_0 . This form of the integral enables us at all times to use the initial distribution which we take to be log-normal:

$$n_0(R_0) = \frac{N_0}{(2\pi)^{1/2} \ln \sigma_g} \exp \left\{ - \frac{1}{2} \left[\frac{\ln(R_0/R_g)}{\ln(\sigma_g)} \right]^2 \right\} \frac{1}{R_0}. \tag{24}$$

Fixed points for numerical integration in R_0 can be chosen once and for all which cover the main peak of the distribution.

Being able to calculate the moments of the distribution, we have a direct way of calculating actual changes in u (or S) from mass conservation. From equation (3) we easily obtain by multiplying by R^3 and integrating over R :

$$\frac{4}{3} \pi \rho_\ell \frac{d}{dt} (N \overline{R^3}) = \dot{m}_v - \frac{4}{3} \pi \rho_\ell N \overline{\lambda R^3}. \tag{25}$$

We can determine \dot{m}_v from one or both of equations (1) and (3). For the case $q=0$, we obtain, using equation (15) in equation (25) and integrating,

$$u(t) - u(0) = - \frac{1}{\rho} \frac{Cn(1 - c_e) + Le}{Cn} \frac{4\pi\rho_\ell}{3} \left[N \overline{R^3}(t) - N_0 \overline{R_0^3} + N \int_0^t dt' \overline{\lambda R^3}(t') \right]. \tag{26}$$

For the case $T = \text{constant}$, assuming $c \ll 1$, we use equation (20) in (25) and integrate to obtain,

$$u(t) - u(0) = - \frac{4\pi\rho_\ell}{3\rho} \left[N \overline{R^3}(t) - N_0 \overline{R_0^3} + N \int_0^t dt' \overline{\lambda R^3}(t') \right]. \tag{27}$$

For large Cn the results are identical, but for $Cn \ll 1$ the change in u is formally larger by a factor Le/Cn .

3. CALCULATIONS

We have performed calculations for the various effects in turn using the water data given in Table 1. Trajectory equations were integrated numerically using a fourth order Runge-Kutta method with moments found from equation (36). Integrals were performed using Simpson's rule with 101 points. As noted previously (Barrett and Clement, 1990), in cases such as radiative redistribution where a lower part of the distribution evaporates, inaccuracies can arise at later times. It is important to note, however, that the removal of such inaccuracies has only a limited physical value. If the bulk of an initial distribution disappears either by evaporation or removal leaving only a residue from the wings of the distribution, the results will be sensitive to the initial specification of these wings. The common practice, continued here, of using lognormal distribution (equation (37)), may not be adequate to describe accurately the wings of physical distributions, so that excessive numerical accuracy in calculating their evolution is unwarranted.

3.1. *Removal alone*

With no growth or redistribution, the solution for n is

$$n(R, t) = n_0(R) e^{-\lambda_0 R^2 t}. \tag{28}$$

We can define a ‘‘half-life’’ $t_{1/2}$ for removal as the time taken for the aerosol volume to reduce by a factor 1/2, so that

$$\int R^3 n_0(R) e^{-\lambda_0 R^2 t_{1/2}} dR = \frac{1}{2} \int R^3 n_0(R) dR. \tag{29}$$

For a monodisperse aerosol $t_{1/2} = \ln 2 / \lambda_0 R^2 = 5780 h / R^2$.

Figure 1 shows the half life vs log-normal parameter R_g for a monodisperse aerosol ($\sigma_g = 1$) and for log-normal (L-N) distributions with $\sigma_g = 1.5$ and 2. The lines are very close to being parallel, indicating that L-N distributions with finite widths act like monodisperse aerosols with suitably chosen monodisperse radii $\bar{R} > R_g$. In fact, the dashed lines are plots of

$$t_{1/2} = \frac{5780 h}{\bar{R}_g^2 \exp[12.5 \times 0.5 (\ln \sigma_g)^2]}, \tag{30}$$

which accurately represents the numerical data. The factor in the denominator of equation (30) is written in the form shown because, for a L-N distribution

$$\bar{R}^m = R_g^m \exp[m^2 \times 0.5 (\ln \sigma_g)^2]. \tag{31}$$

Thus the denominator is related to $\bar{R}^{\sqrt{12.5}} \simeq \bar{R}^{3.5}$. Although it is not unreasonable that the removal rate should be proportional to some higher moment of the size distribution, we have not been able to derive (30) analytically; nor have we tested it for size distributions other than L-N.

3.2. *Kelvin effect alone (Ostwald ripening)*

As noted previously (Barrett and Fissan, 1989), simulations with the Kelvin effect give reasonable results at short times but poor results for longer times due to the necessity to determine $S(t)$ very accurately as $S \rightarrow 1$. However, the asymptotic theory (Lifshitz and Slezov, 1959, 1961) of Ostwald ripening with $R \gg R_y$ is well known, although some derivations of the results are confusing. We shall follow the derivation of Dunning (1973),

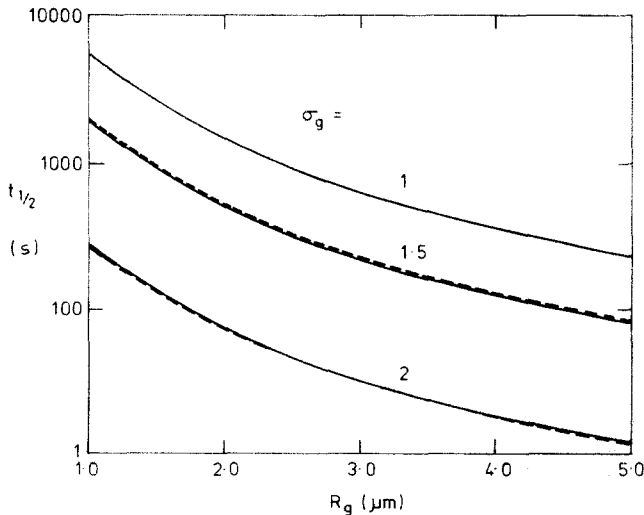


Fig. 1. The half-life $t_{1/2}$ for gravitational fallout as a function of R_g for various σ_g specifying log-normal size distributions. The dashed lines are given by the approximate formula in the text.

referred to as I below, which removes some of the confusion, although it contains numerous typographical errors and some omissions.

To determine the change of saturation with time, we integrate equation (21) with $q=0$, the approximation of $c=c_e$ on the RHS, and all terms on the RHS, other than \dot{m} , assumed to be constant:

$$S_0 - S = \hat{K} (\overline{NR^3}(t) - N_0 \overline{R_0^3}), \quad (32)$$

where

$$\hat{K} = \frac{4\pi\rho_l}{3\rho_{ve}(T)} \frac{\mu_v}{\mu_v - (\mu_v - \mu_g)c_e(T)} \left[1 - c_e(T) + \frac{Le}{Cn} \right]. \quad (33)$$

Equation (32) is an alternative form of equation (26) without the removal term, and may be identified with equation (21) of I, apart from a different \hat{K} and $N_0=0$ in I. It should be possible to follow Dunning's derivation in I, the crucial step being the derivation of equation (I-40) which in our notation should read:

$$\bar{R}^3 - \frac{S_\infty R_\gamma}{2\hat{K} N \bar{R}^3(t_0)} (\bar{R}^2 + 2R_\gamma \bar{R}) = \frac{AR_\gamma t}{\gamma N} + \text{const.}, \quad (34)$$

where $N\bar{R}^3(t_0)$ gives the aerosol volume at time t_0 in the ripening period. A factor M/d , which appears in equation (I-37) appears to have been omitted from the second term in equation (I-40) and subsequent equations.

The factor γ_v in (34) takes the value 9/4, characteristic of the asymptotic distribution, and A is our constant (7) specifying \dot{R} (α in I).

At long times, the solution of equation (34) reduces to the simpler expression of Wagner (1961):

$$\bar{R} \approx \left(\frac{AR_\gamma t}{\gamma_v} \right)^{1/3}. \quad (35)$$

By choosing a typical mean radius, \bar{R} , we thus obtain a timescale for Ostwald ripening

$$t_{OR} \approx \gamma_N \bar{R}^3 / (AR_\gamma). \quad (36)$$

The remainder of the derivation in I proceeds unchanged with the asymptotic form of the size distribution being given by equation (I-61). Using the constants for water from Table 1, we have $t_{OR} = 17.4 \bar{R}^3 (\mu\text{m})\text{s}$ at $T=20^\circ\text{C}$ and $t_{OR} = 7.7 \bar{R}^3 (\mu\text{m})\text{s}$ at $T=100^\circ\text{C}$. An asymptotic containment water aerosol at 100°C is hardly likely to have $\bar{R} < 5 \mu\text{m}$, for which $t_{OR} = 16$ min, and more likely nearer $\bar{R} = 10 \mu\text{m}$, for which $t_{OR} \approx 2$ h. However the timescale does indicate that the Kelvin effect is clearly important for water aerosols contained for periods of the order of an hour or more.

3.3. Solute effect alone

In this case we allow no heat losses so that $q=0$ and omit the Kelvin and radiative terms in \dot{R} given by equation (5). The aerosol tends to an equilibrium in which salt particles with initial radius, R_s , have a final radius, R_∞ , given by

$$R_\infty = \left(\frac{b}{1-S_\infty} \right)^{1/3} R_s. \quad (37)$$

The final distribution is given by (equation (22))

$$\begin{aligned} n(R_\infty, t \rightarrow \infty) &= n_0(R_s) \frac{\partial R_s}{\partial R_\infty} = \left(\frac{1-S_\infty}{b} \right)^{1/3} n_0 \left(\left[\frac{1-S_\infty}{b} \right]^{1/3} R_\infty \right) \\ &= \frac{N_0}{(2\pi)^{1/2} \ln \sigma_g} \exp \left\{ -\frac{1}{2} \left[\frac{\ln(1-S_\infty)^{1/3} R_\infty / (b^{1/3} R_s)}{\ln \sigma_g} \right]^2 \right\} \frac{1}{R_\infty}, \end{aligned} \quad (38)$$

for an initial L-N distribution.

The final distribution is then also log-normal with the same value of σ_g , but R_g replaced by $b^{1/3} R_g / (1 - S_\infty)^{1/3}$. Consequently, the moments are related by

$$\overline{R_x^n} = \left(\frac{b}{1 - S_\infty} \right)^{n/3} \overline{R_s^n}. \quad (39)$$

We can now find S_∞ from the mass conservation condition (32) by using equation (39) with $n=3$:

$$1 - S_\infty = \frac{1}{2} \{ -(S_i - 1 + K_i) + [(1 - S_i - K_i)^2 + 4K_i b]^{1/2} \}, \quad (40)$$

where $K_i = \hat{K} N_0 R_g^3 \exp[4.5(\ln \sigma_g)^2]$.

We have used S_i rather than S_0 in equation (40) to represent the initial saturation before any water condensation takes place. Generally, following a sudden advent of water vapour or steam the aerosol will equilibrate in two stages (Clement, 1988), the first timescale being given by equation (17). During this initial rapid condensation in which $\dot{R} \sim 1/R$, particles of radius R_s grow to $R_0 = (R_s^2 + R_G^2)^{1/2}$, where R_G is a constant likely to be 1–5 μm . The saturation or u will now be at an approximate equilibrium. A redistribution of the water mass will then occur over a longer timescale to accommodate the salt content of the droplets. It is our purpose now to discover the properties of this redistributive process.

If the initial saturation for the redistributive calculation is S_0 , which we taken to be 1 for the calculations, the mass balance corresponding to equation (40) for the initial aerosol growth gives

$$S_i - S_0 = K_i \left[\frac{(R_s^2 + R_G^2)^{3/2}}{R_s^3} - 1 \right]. \quad (41)$$

This difference corresponds to the water mass in the aerosol which will usually be much greater than the salt mass so that $S_i - 1 \gg K_i$ and equation (40) can be expanded to give

$$1 - S_\infty \approx \frac{K_i b}{S_i - 1} \approx \frac{b R_s^3}{(R_s^2 + R_G^2)^{3/2}}, \quad (42)$$

where the final form follows from equation (41) with $S_0 = 1$. In Figs 2(a) and (b) we show the variation of the mean \bar{R} and standard deviation σ for L-N CsOH aerosol with $R_g = 0.15 \mu\text{m}$, $\sigma_g = 1.5$, with initial water coatings of 1 μm (Fig. 2a) and 5 μm (Fig. 2b). Note also that $N = 10^{11} \text{m}^{-3}$ in Fig. 2a and 10^{10}m^{-3} in Fig. 2b. In Table 2 we show the asymptotic values, \bar{R}_∞ and σ_∞ calculated by the theory just discussed; these agree with the numerical results shown in Fig. 2. The redistribution takes place in very different timescales in Figs 2a and 2b for which we can obtain estimates by putting $S = S_\infty = \text{const.}$ in the growth rate equation (5) so that

$$\dot{R} = A \left(\frac{(S_\infty - 1) + b R_s^3 / R^3}{R} \right). \quad (43)$$

We write $R = R_\infty + y(t)$, where y is assumed small, expand equation (43) to first order in y and solve to obtain

$$y \approx \exp \left(\frac{3A(1 - S_\infty)t}{R_\infty^2} \right). \quad (44)$$

Table 2. Initial and calculated parameters for the redistributions calculations with solute

R_G (μm)	N (m^{-3})	$S_i - 1$	$1 - S_\infty$ equation (22)	$1 - S_\infty$ equation (25)	$\left(\frac{b}{1 - S_\infty} \right)^{1/3}$	\bar{R}_∞ (μm)	σ_∞ (μm)
1	10^{11}	0.0170	4.87×10^{-3}	6.25×10^{-3}	5.76	0.94	0.40
5	10^{10}	0.204	5.26×10^{-5}	5.25×10^{-5}	26.05	4.24	1.79

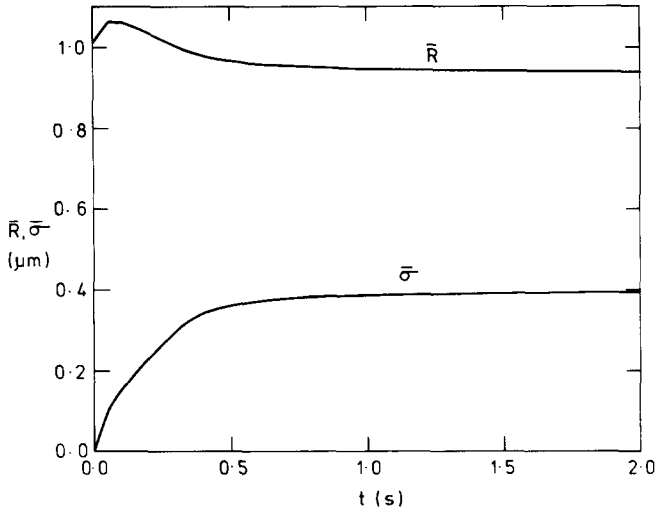


Fig. 2a. Mean radius \bar{R} and standard deviation $\bar{\sigma}$ as a function of time during water redistribution on an initial salt aerosol with $R_g=0.15 \mu\text{m}$, $\sigma_g=1.5$ having a water coating of $R_G=1 \mu\text{m}$.

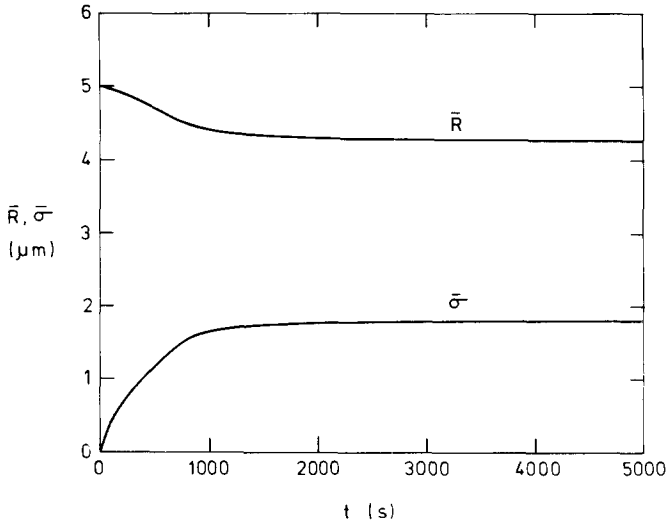


Fig. 2b. As for Fig. 2a but with $R_G=5 \mu\text{m}$.

An estimate for the mean relevant solute redistributive timescale is thus

$$t_{\text{sol}} \approx \frac{\bar{R}_\infty^2}{3A(1-S_\infty)} \tag{45}$$

For the distributions calculated in Figs 2a and 2b, $t_{\text{sol}}=0.15 \text{ s}$ and 820 s , respectively, which are comparable to the numerical results shown.

Figure 3 shows the size distribution at various times calculated from equation (22). The initial distribution is the same as in Fig. 2a and is close to a δ function. The dashed line shows the asymptotic distribution which is a log-normal with mean $[b/(1-s_\infty)]^{1/3}$ $R_g \approx 0.864 \mu\text{m}$ and $\sigma_g = 1.5$. Figure 4 shows the variation in the distribution of concentration $C = \text{vol salt/vol water}$ for this case. Initially the distribution of C is quite broad but it rapidly tends to a delta function distribution $n(C, \infty) = \delta(C - C_\infty)$ where the asymptotic concentration is

$$C_\infty = \frac{R_s^3}{(b/(1-s_\infty))R_s^3 - R_s^3} = \frac{1}{5.76^3 - 1} = 0.00526. \tag{46}$$

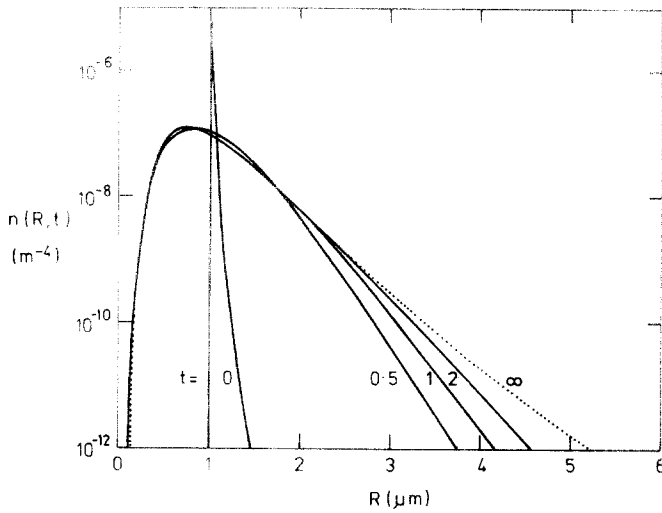


Fig. 3. Size distributions, $n(R, t)$, as functions of radius R at different times for the case shown in Fig. 3a.

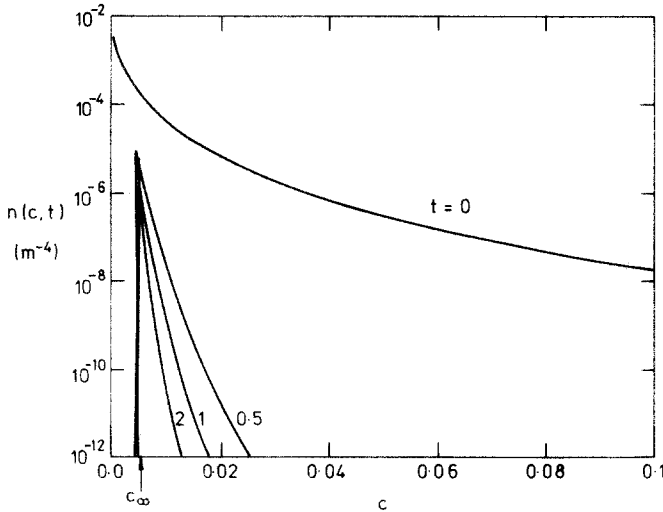


Fig. 4. Distributions of salt concentration, $n(C, t)$, where $C = \text{salt volume/total volume}$, at different times for the case shown in Fig. 3a.

3.4. Radiative redistribution and removal

For an aerosol which is radiating, we have to specify the total heat transfer q to or from the system, and previously (Barrett and Clement, 1990) and in section 2.3 we have considered the following cases: (i) pure radiative cooling: q given by equation (29) when the temperature of the system falls, and (ii) constant temperature with constant amount of vapour plus aerosol apart from a settling loss where u satisfies equation (32).

In case (i) we compare results which allow for redistribution through the R dependence of $G(R)$ with calculations which allow for the same amount of total cooling, but have all the aerosol growing with \dot{R} proportional to $1/R$.

To be explicit, the alternative forms for \dot{R} for case (i), which may be obtained from equations (18) and (19), or equations (22) and (23) of Barrett and Clement (1990), are as follows.

Redistribution with cooling

$$\dot{R} = \frac{A}{R} \left[B_1 \frac{\overline{RG(R)}}{\bar{R}} + G(R) \right]. \tag{47}$$

Cooling alone

$$\dot{R} = \frac{A}{R} B_2 \frac{\overline{RG(R)}}{\bar{R}}, \quad (48)$$

where

$$B_1 = B_2 - 1 = \frac{\text{Cn}(\text{Le} - 1)(1 - c_e)}{\text{Le} + (1 - c_e)\text{Cn}}. \quad (49)$$

Clearly, if $G(R)$ is independent of R , the growth rates (47) and (48) are identical.

From equation (21) of Barrett and Clement (1990) or, alternatively, equations (11), (18), (48) and (49), we have $\overline{Q_R} = L[1 + (1 - c_e)\text{Cn}/\text{Le}] \dot{m}_v$.

Thus, in case (i) from equations (2) and (29), the fall in temperature is given by

$$\rho \bar{c}_p \frac{dT}{dt} = -(1 - c_e) \frac{\text{Cn}}{\text{Le}} L \dot{m}_v. \quad (50)$$

Now we substitute for \dot{m}_v from the mass conservation equation (25) and integrate, ignoring the change of physical properties with t or T , to obtain

$$T - T_0 = -B_3 \left[\overline{NR^3(t)} - N_0 \overline{R_0^3} + \int_0^t dt' N \overline{\lambda R^3(t')} \right], \quad (51)$$

where

$$B_3 = \frac{4\pi\rho_l(1 - c_e)\text{Cn}L}{3\rho\bar{c}_p\text{Le}}. \quad (52)$$

Calculations have been performed with the growth rates (47) and (48) using the parameters given in Table 1. Results at 100°C are shown in Figs 5a and 5b. The water aerosol grows rapidly in size so that its density initially increases before reaching a peak and then falling rapidly as settling begins to dominate over growth. Meanwhile the temperature falls at an initially increasing rate, because of the growth in droplet size, but then levels off as the aerosol becomes depleted. The only significant difference that redistribution appears to make in these calculations is that of keeping a significant width to the size distribution, as shown in Fig. 5a. At longer times there is slightly more airborne aerosol density with redistribution, probably because it keeps more aerosol with smaller radii. The results (not shown) at 20°C show larger differences between the cooling only and cooling plus redistribution cases. This difference arises because of the much larger negative B , which is proportional to Cn . A much larger fraction of the bottom end of the size distribution is then evaporating at 20°C and we find a slight increase in $\bar{\sigma}$ with t and a lower airborne mass ρ_a .

We now discuss some results for radiative redistribution and gravitational removal in case (ii). The growth rate is given by

$$\dot{R} = \frac{A}{R}(S - 1 + G(R)). \quad (53)$$

Using eqn (6) (with the approximation $c = c_e(T)$ in the denominator), together with equation (27) gives an expression for $S(t)$ in terms of moments of the distribution. We choose the initial saturation S_0 to have the value which gives no net condensation or evaporation, i.e. $\dot{m}_v = 0$. From equations (11) and (53), we find

$$S_0 = 1 - \frac{\overline{RG(R)}}{\bar{R}}. \quad (54)$$

Radiative redistribution causes large droplets (those with $G(R) > 1 - S$) to grow and small ones to evaporate. The aerosol is then no longer in net equilibrium with the vapour and more vapour condenses leading to a reduction in S and an increase in total aerosol mass. However, gravitational settling removes some of the aerosol, reducing the total suspended mass. The net effect may therefore be an increase or a decrease in suspended mass initially,

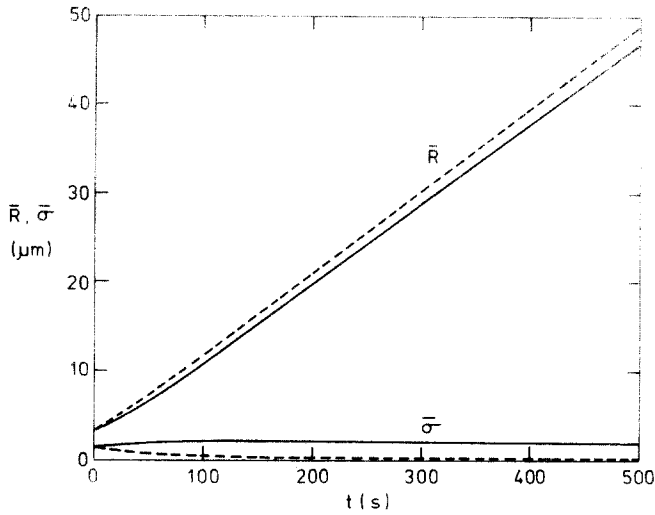


Fig. 5a. Mean radius \bar{R} and standard distribution $\bar{\sigma}$ as functions of time for pure radiative cooling and redistribution at 100°C of an initial aerosol specified by $R_g = 3 \mu\text{m}$, $\sigma_g = 1.5$, $N_0 = 10^8$. The solid line is with, and the dashed line without, redistribution.

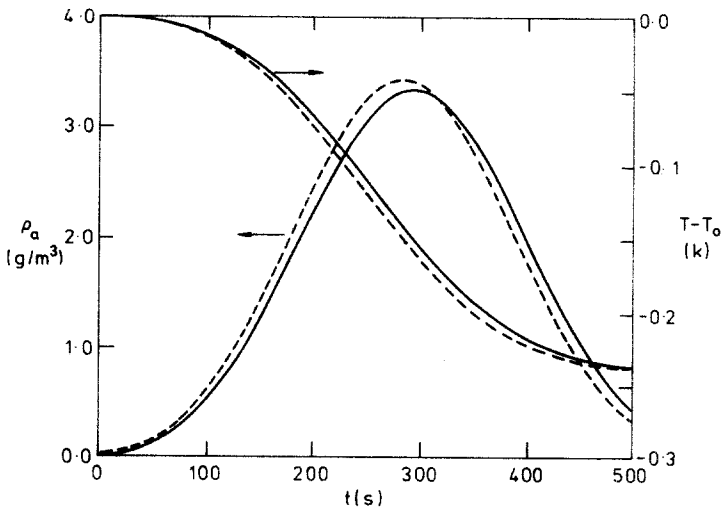


Fig. 5b. The suspended aerosol mass density, ρ_a , and mixture temperature change, $T - T_0$, as functions of time for the case shown in Fig. 5a.

as shown in Fig. 6. For the case illustrated at 100°C , redistribution and condensation exceed removal initially and the suspended volume increases. However, as the aerosol grows the removal term (which, with $\lambda(R)$ given by equation (4), varies as the fifth moment of the size distribution) increases, leading to a rapid fall in suspended volume for longer times. At 20°C the vapour density is much less than at the higher temperature and condensation never exceeds removal in the cases considered here. Also shown by the dashed line in Fig. 6 is the suspended volume if only removal operates and there is no radiative redistribution. It is clear that the radiative redistribution of vapour onto larger droplets greatly enhances gravitational removal of aerosol.

3.5. Solute effect and removal

Finally, we again consider the hygroscopic aerosols discussed in section 3.3 but now also include the effect of gravitational removal. The simplest cases are those where the redis-

tribution timescale, given approximately by equation (45), is much shorter than the timescale for significant fallout to occur. We can then treat the two processes separately using the final distribution due to hygroscopic redistribution, given by equation (38), as the initial distribution, $N_0(R)$, in the equation for evolution by gravitational removal alone, equation (28). Such a case is illustrated by Fig. 7 which shows the effect of removal on a case previously considered in section 3.3 (Fig. 2a). At first there is very rapid condensation, reducing S from its initial value of 1 to close to its equilibrium value S_∞ given by equation (40) (see also Table 2). Subsequent redistribution on a timescale $\sim s$ causes little change in suspended aerosol mass but leads to a much wider distribution which then falls out much more rapidly than the initial, almost monodisperse, distribution. The dashed lines show the amount of salt suspended (assuming the salt density is the same as water): after 5 h more than twice as much salt has fallen out than would be predicted by gravitational removal

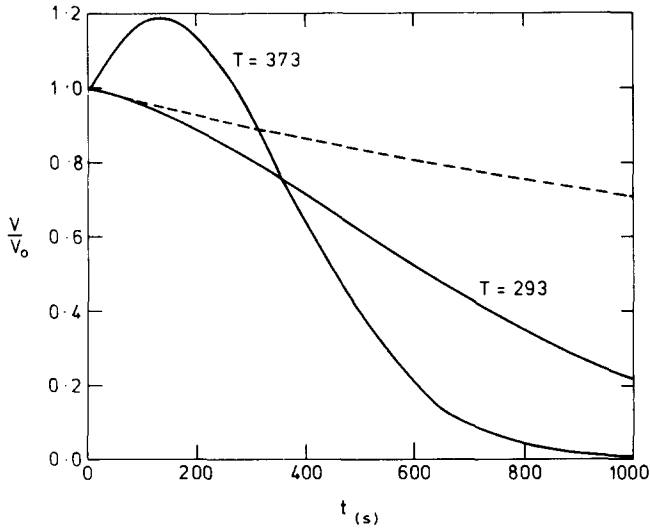


Fig. 6. Aerosol volume/initial volume as a function of time for radiative redistribution and removal at constant temperatures ($T=293$ K and 373 K) for an aerosol with initial distribution: $R_g=3 \mu\text{m}$, $\sigma_g=1.5$, $N_0=10^{10} \text{ m}^{-3}$. The solid line is with, and the dashed line without, redistribution.

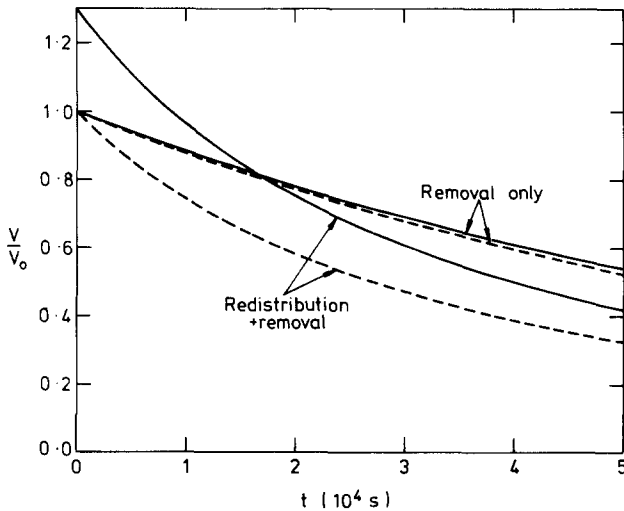


Fig. 7. Aerosol volume/initial volume as a function of time for water redistribution on a salt aerosol with $R_g=0.5 \mu\text{m}$, $\sigma_g=1.5$, $N_0=10^{11} \text{ m}^{-3}$ and $R_G=1 \mu\text{m}$ water coating. Also shown (dashed lines) are salt volume/initial salt volume vs time.

alone. For the second case considered in section 3.3, the two processes cannot be treated separately and a full numerical calculation is necessary. The results (not shown) display a similar enhancement of fallout by redistribution although the effect is not as large as in Fig. 7.

4. CONCLUSIONS

We have described redistributive and gravitational settling which affect a water cloud in a well-mixed volume or containment. In order to specify the supersaturation in the volume we have coupled the aerosol growth to both the vapour concentration and temperature, heat transfer, but no mass transfer, being allowed to the walls. Our conclusions from the theory and calculations performed are the following.

(A) As was originally pointed out for the water case with $C_n < 1$ (Clement, 1985), the coupling between heat and mass transfer is essential. The neglect of heat transfer essentially means neglecting the dominant Le/C_n terms in equations (15) and (26). In physical terms, release of latent heat and consequent temperature change is the controlling factor on the supersaturation following condensation with redistribution in a water cloud, just as it is in condensation without redistribution.

(B) An accurate form (17), has been given for the initial equilibration timescale between the aerosol and the surrounding vapour–gas mixture. Following this equilibration, which will usually occur in seconds or less, redistributive processes take place with the supersaturation maintained at a slowly changing quasi-equilibration value given by one of the equations (26) or (27) in the special cases considered.

(C) A formal solution (equation (22)) has been given for the aerosol size distribution, which leads to a convenient way to calculate its moments as a function of time. Numerical problems in calculating trajectories can arise, particularly if much of the distribution evaporates or falls out. However, in this case it is likely that results will be sensitive to forms assumed for the wings of an initial size distribution which may be poorly determined physically.

(D) Analytic timescales have been obtained for gravitational settling alone, Ostwald ripening from the Kelvin effect, and redistribution arising from solute in droplets. In this last case an initial log-normal aerosol size distribution will tend to a final equilibrium distribution of a similar form. The timescale can be used to see which process is likely to dominate in a given situation: they will often occur in sequence because, for example, the solute redistribution can be quite fast whereas settling and Ostwald ripening are slow.

(E) Condensation on a hygroscopic aerosol can occur in two stages: rapid growth followed by solute driven redistribution. Numerical calculations performed for the redistributive phase show how the salt concentration tends to uniformity and the influence that this can have in enhancing gravitational fallout.

(F) For radiative redistribution, results have been presented for the two cases of pure radiative cooling and cooling but the mixture maintained at a constant temperature. In the pure cooling case the redistributive process has only a relatively small effect on the settling rate, so that models which neglect the explicit radiative terms in the droplet growth rate would give reasonable results. In the second case, however, these models would fail as they would predict only a slow settling rate, whereas the results show that the airborne mass can be rapidly depleted by the redistribution of mass to larger droplets and their subsequent fallout.

Current codes used to calculate aerosol behaviour coupled with thermal hydraulics in a PWR containment have several deficiencies when compared to the results obtained here, although they do take account of aerosol agglomeration and interactions with walls not considered here. For example, no codes include the advanced code ITHACA (Ketchell *et al.*, 1992) yet include the radiative term in the Mason equation for aerosol growth. The work here shows that its inclusion could significantly increase gravitational settling and thus reduce radioactive source terms predicted for practical situations. The MAAP model jumps

straight to a final equilibrium radius coupled to supersaturation for a hygroscopic aerosol (Jokiniemi, 1992) without considering whether redistributive processes could occur first. We hope that the present results may be used to improve the models and give some insight into timescales and redistributive effects in various experiments and situations which could occur in practice.

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APPENDIX A. TRAJECTORY SOLUTIONS

The aerosol equation (3) with growth and removal terms has trajectories from $(n_0, R_0, t=0)$ to (n, R, t) which are solutions of

$$\dot{R} = \frac{dR}{dt} \equiv v(R, t), \quad (\text{A1})$$

$$\frac{dn}{dt} = -\left(\lambda(R) + \frac{\partial v}{\partial R}\right)n. \quad (\text{A2})$$

Taking the partial derivative with respect to R_0 of the formal integral solution of equation (A1), we have

$$y(R_0, t) = \left(\frac{\partial R}{\partial R_0}\right)_t = 1 + \int_0^t \frac{\partial v}{\partial R'} y(R_0, t') dt'. \quad (\text{A3})$$

Differentiating (A3) with respect to t at constant R_0 , and then solving the resulting differential equation gives

$$y = \exp\left[\int_0^t \frac{\partial v}{\partial R}(R_0, t') dt'\right]. \quad (\text{A4})$$

Using this result, we obtain for the solution of (A2)

$$\begin{aligned} n &= n_0 \exp\left[-\int_0^t dt' \left(\lambda + \frac{\partial v}{\partial R}(R_0, t')\right) dt'\right] \\ &= n_0(R_0) \left(\frac{\partial R_0}{\partial R}\right)_t \exp\left[-\int_0^t \lambda(R'(R_0, t')) dt'\right]. \end{aligned} \quad (\text{A5})$$

The need to calculate $\partial v/\partial R$ numerically can cause problems in the numerical calculation of the trajectory using equation (A2). We derive an alternative equation which avoids the problem for redistribution of hygroscopic aerosols where the trajectory follows R_s to R_0 to R :

$$\frac{\partial v}{\partial R} = \frac{v}{R} + \frac{A 3bR_s^3}{R^3} \left[\frac{1}{R_s} \frac{\partial R_s}{\partial R} - \frac{1}{R} \right], \quad (\text{A6})$$

$$\frac{\partial R_s}{\partial R} = \frac{\partial R_s}{\partial R_0} \frac{\partial R_0}{\partial R} = \frac{R_0}{R_s} \exp \left[- \int_0^t \frac{\partial v}{\partial R} dt' \right]. \quad (\text{A7})$$

Eliminating the exponential in (A7) using the solution (A5) with $\lambda=0$, and substituting in (A2) gives

$$\frac{dn}{dt} = -n \left[\frac{v}{R} + \frac{A 3bR_s^3}{R^3} \left(\frac{1}{R_s n_s(R_s)} - \frac{1}{R} \right) \right]. \quad (\text{A8})$$