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PRELIMINARY ASSESSMENT OF THE IMPORTANCE OF TURBULENT COAGULATION IN THE KUWAITI OIL FIRES

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Preliminary Assessment of the Importance of Turbulent Coagulation in the Kuwaiti Oil Fires

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This study provides a mathematical determination of the spatial distribution of aerosols due to turbulent shear coagulation and turbulent inertial coagulation, as applied to the conditions of the Kuwaiti Oil Fires (KOF) of 1991. Using an approximation from a forest fire for the normalized size distribution of aerosols, the downstream particle concentration is found by the concurrent solution of the coagulations' kinetics combined with turbulent atmospheric diffusion. The result shows the explicit dependence of the concentration on the following principal parameters: turbulent energy dissipation rate, turbulent diffusion constant, average wind speed, mass ejection from a well, Kolmorogov time scale for turbulence, and Kolmorogov length scale for turbulence. For very large values of turbulent energy dissipation rate, turbulent inertial coagulation is more effective than turbulent shear coagulation in particle growth. The spatial dependence of concentration attributed to turbulent coagulation may vary considerably. Depending on the choice of parameters, the importance of turbulent coagulation in particle transport processes may extend from less than a kilometer to tens of kilometers.
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This paper, "Preliminary Assessment of the Importance of Turbulent Coagulation in the Kuwaiti Oil Fires," is part of a larger Defense Nuclear Agency (DNA)-sponsored program dealing with the Kuwaiti Oil Fires (KOF) of 1991. This DNA program is directed toward improving our capability to predict the transport of pollutants arising from very large natural and manmade fires.

The author has received constructive comments and support from numerous individuals in the preparation of this paper. Thanks are due to Dr. Ernie Bauer (IDA) for pointing out the importance of the fractal nature of aerosols produced in these fires and the consequences of fractal considerations in coagulation; to Dr. Ed Townsley (IDA), Dr. John Cockayne (SAIC), and LCDR Harris O'Bryant (DNA) for their critical review of the equations and mathematical formalism; to Dr. Darrel Baumgardner (NCAR) for helpful comments concerning coagulation processes and the related aerosol size distribution; to Dr. Ian Sykes (ARAP) for pointing out the relationship between time averaging and plume spreading; to Drs. Robert Oliver (IDA) and Tom Vonder Haar (METSAT) for their general review; and to Dr. Ed Townsley and Mr. Peter Kysar (IDA) for their computational support. In addition, the author would also like to thank Mrs. Sharon Y. Wiley for her outstanding job of technical typing.
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EXECUTIVE SUMMARY

The Kuwaiti Oil Fires (KOF) of 1991 provided an opportunity for the Defense Nuclear Agency (DNA) to address some key questions about the atmospheric transport* of particles that are relevant to the prediction of late-time dust cloud motion. During the fires, much data were collected by numerous U.S. and foreign scientific agencies, and now the critical issue is to assemble and organize a consistent set of measurements that can be evaluated against computer models to predict small particle transport. These computer models are required for the prediction of long-range transport of nuclear dust clouds and for the prediction of smoke plumes from large oil, industrial, or urban fires that might affect electro-optical sensor performance.

The KOF included features that were common with large forest fires, but the KOF also had other characteristics that made them unique. When various long-range transport codes were exercised for KOF-like problems, certain important deficiencies were encountered. This raises the following questions: Are the mathematical and physical models of the codes unsatisfactory? Are they being applied in situations for which they are not intended?

A key requirement for predicting particle transport is the necessity to follow the motion of some "tracer" particles (Lagrangian viewpoint). To be good tracers for modeling purposes, the particles must remain physically and chemically inert during the transport process. This condition of physical and chemical invariance can be termed a "frozen" or "aged" state, and it will be reached when processes such as condensation and evaporation, accretion of water vapor or other species, combustion and other chemical reactions, and turbulent coagulation are no longer active. In this paper, we evaluate turbulent coagulation. The other factors will be addressed in subsequent investigations.

The purpose of this paper is to make a preliminary assessment of the conditions under which turbulent coagulation is or is not a significant factor. Clearly, if airborne particles continue to coagulate or otherwise grow or shrink in appreciable amounts during the transport phase, they cannot be unequivocally "tagged," and this potentially diminishes

* In this report, transport means both advection and diffusion.
the validity of a transport calculation. The importance of this investigation is that it defines the nearest spatial region where turbulent coagulation can be neglected, so that suitable comparisons can be made between transport prediction models and experimental data (notwithstanding other aging processes).

Specific reasons for evaluating turbulent coagulation in the KOF are as follows:

- Initial reported aerosol particle/mass densities are extremely high, leading to large coagulation growth rates.
- In the large Canadian forest fire of 1950, it was found that turbulent coagulation was an important consideration.
- Mass injection into the atmosphere is comparable to or exceeds that of large forest fires.
- There is a paucity of experimental data on turbulent coagulation in the atmosphere, and it may be necessary to have a means of interpreting certain KOF experimental data in this light.
- It is important to understand the dependence of turbulent coagulation on windspeed (as it relates to transport time), turbulent kinetic energy dissipation rate per unit mass, aerosol density and size distribution, and atmospheric diffusion.
- The analytic representation of collection kernels for existing turbulent coagulation theories is based on spherical particles, but this may not be valid for KOF. Therefore, it is necessary to have a simple way of estimating the changes in coagulation dynamics as a function of aerosol shape.

The approach is to develop an analytical formulation of the spatial behavior of aerosol density as the particles are carried along the turbulent wind field while simultaneously undergoing coagulation. The KOF aerosol problem is one in which the particle density of the "superplume" is formed by the merging of hundreds of individual plumes. By example, Figure S-1 shows three individual plumes merging into a superplume.

In this idealized figure, we hypothesize that there will be significant coagulation effects as the flames emerge from each individual well since the particle density (concentration) and mass density will be extremely high. As the individual plumes spread, the concentration will decrease by geometric dilution caused by atmospheric turbulence and coagulation. Ultimately, we postulate that the coagulation process becomes insignificant when compared to other processes due to decreasing concentrations. The surfaces that
define this transition are labeled "Coagulation Boundary." The shaded areas behind these boundaries are the regions where coagulation is assumed to be insignificant.

Figure S-1. Geometric Viewpoint for Coagulation Considerations

The first step in understanding the behavior of aerosol concentration in the superplume is to evaluate the contributions from an individual fire. Figure S-2 shows the mathematical model for a single plume. This model can be applied to an individual source or to the large plume provided that the appropriate initial conditions are present.

Figure S-2. Model for Individual Plume
In Figure S-2, $q_o$ is the distance from the source (oil well) where the kinetic energy of the ejected oil is no longer a factor in the transport process. $A(q_o)$ is the cross-sectional area associated with $q_o$. These dimensions are estimated from visual observations. From turbulent diffusion models, we obtain the approximation $A(q) = (\text{const}) q^2$ for $q \geq q_o$. Using $A(q)$ in conjunction with mathematical simplification of the collection kernels for turbulent shear coagulation and turbulent inertial coagulation and an approximation for the shape of the aerosol radius distribution based on the large Canadian forest fire of 1950, we have been able to derive an expression for the aerosol concentration, $N(q)$, as a function of the turbulent dissipation rate, average windspeed, and source emission rate. For the conditions assumed in this study, turbulent inertial coagulation is nearly two orders of magnitude more effective than turbulent shear coagulation.

The concentration, $N(q)$, along the center line trajectory of Figure S-2, is given by

$$N(q) = N_1(q) \Gamma_c(q) \; ,$$

where

$$N_1(q) = \frac{S_o}{U(q) A(q)}$$

is the ordinary geometric spreading and

$$\Gamma_c = \exp \left[ -l_1 \left( \frac{1}{q_o} - \frac{1}{q} \right) \right] ; q \geq q_o$$

is the part that is due to coagulation. In these equations, $S_o$ is the particle emission rate, $U(q)$ is the windspeed along $q$, $A(q)$ is the cross-sectional area of the plume, $q_o$ is the starting point for the growth of the plume, and $l_1$ is a characteristic length associated with turbulent inertial coagulation. $l_1$ is given by

$$l_1 = \frac{5.3 R_m r_I}{\rho_A \tau_k \lambda_k U^2} \; ,$$

where $R_m$ is the mass emission rate, $\rho_A$ is the atmospheric mass density, $r_I$ is a characteristic radius associated with the aerosol size shape distribution, $\tau_k$ is the Kolmogorov time scale for turbulence, $\lambda_k$ is the Kolmogorov length scale for turbulence, and $U$ is the average windspeed.
Figure S-3 shows the general behavior of $\Gamma_c$ as a function of $q$. The significant features of this curve are the initial value of 1.0, the asymptotic behavior $\exp(-l/q_0)$, and the spatial variation, which behaves as $\exp(-l/q)$. Turbulent inertial coagulation will not have an important effect on particle transport when this factor does not vary significantly over the range of interest (i.e., the spatial region where theoretical models of particle transport are to be compared with experimental observations).

As shown in Figure S-3, the rate of spatial change of $\Gamma_c$ decreases as the distance, $q$, increases. By establishing a practical criterion consistent with the prediction capabilities of particle transport codes, it is possible to establish a minimum range, $q_{\text{min}}$, beyond which the effects of coagulation on particle transport should not be a concern. The criterion is

$$\frac{\Gamma_c(2q_{\text{min}})}{\Gamma_c(q_{\text{min}})} \geq 0.9, \quad (5)$$

and this inequality leads to the minimum range,

$$q_{\text{min}} \geq 5l_1, \quad (6)$$

in the domain of interest [where $\exp(-l/q_0) \ll 1$]. $q_{\text{min}}$ is the distance beyond which $\ln \Gamma_c$ changes by less than 10 percent over $q_{\text{min}}$. 
The important point is that $q_0$ and the factors comprising $l_1$ can be determined from remote and in-situ measurements taken during the fires. Using estimated numbers for the conditions of the KOF, we find that $l_1$ can range up to several kilometers and, in some cases, can be as large as 10 kilometers. On the other hand, $q_0$ may only range up to a few hundred meters. At this time, there remains a large number of unknown factors in the computation of $l_1$. As the data reduction and analysis from the KOF continue, improved estimates for $l_1$ will become available, and we will be able to define more precisely the regions where a self-consistent set of measurements can be used to validate particle transport models.
I. INTRODUCTION AND PURPOSE

This paper is part of an IDA Task about the "Characterization of Data and Utilization of Advanced Technologies for Predicting the Atmospheric Transport of Effluents from the Kuwaiti Oil Field Fires," being performed for the Defense Nuclear Agency (DNA). The Kuwaiti Oil Fires (KOF) of 1991 provided an opportunity for DNA to address some key questions about the atmospheric transport of particles that are relevant to the prediction of late-time dust and smoke cloud motion. During the fires, much data were collected by numerous U.S. and foreign scientific agencies, and now the critical issue is to assemble and organize a consistent set of measurements that can be evaluated against computer codes to predict small particle transport. These computer codes are required for the prediction of long-range transport of nuclear dust clouds and for the prediction of smoke plumes from large oil, industrial, or urban fires that might affect electro-optical sensor performance.

The KOF included features that were common with large forest fires, but the KOF also had other characteristics that made them unique. Bauer (Ref. 1) has provided an initial assessment of the characterization of the KOF in relation to various kinds of large fires and has identified certain key aspects that should be studied further. However, the reviewers of his document pointed out that when various long-range transport codes were exercised on KOF-like problems, the agreement was often not very good. This raises the following questions: Are the mathematical and physical models of the codes unsatisfactory? Are they being applied in situations for which they are not intended?

A key requirement for predicting particle transport is the necessity to follow the motion of some "tracer" particles (Lagrangian viewpoint). To be good tracers, the particles must remain physically and chemically inert during the transport process. This condition of physical and chemical invariance can be termed a "frozen" or "aged" state and applies to those particles sufficiently far away from the sources of the plume. Some of the processes that should be evaluated to ensure that transport calculations for the frozen state are performed properly include condensation and evaporation, accretion of water vapor or other species, combustion and other chemical reactions, Brownian coagulation, coagulation in laminar shear flow, gravitational coagulation, turbulent shear coagulation, and turbulent
inertial coagulation. Baumgardner (Ref. 2) has also suggested that turbulent mixing may have an effect on aerosol growth under certain circumstances. However, since all of these processes are most active during the initial stages of mass injection into the atmosphere and simple transport codes cannot be applied to a very young plume in which the tracer particles are not yet "aged," it is important to establish a minimum range to which a transport code can be applied for particles whose mass no longer changes with distance traveled.

In this paper, we make an initial assessment of the effects of turbulent shear coagulation and turbulent inertial coagulation on aerosol growth in the KOF. The results from this analysis may be useful in defining the "aged" regions that are applicable for particle transport analysis. Specific reasons for evaluating these turbulent coagulation processes in the KOF are as follows:

- Initial reported aerosol particle/mass densities are extremely high, leading to large coagulation growth rates.
- In the Canadian fire of 1950, turbulent coagulation was an important consideration (see Sections II and III).
- Mass ejection is comparable to or exceeds that of large forest fires.
- There is a paucity of experimental data on turbulent coagulation in the atmosphere, and it may be necessary to have a means of interpreting certain KOF experimental data in this light.
- It is important to understand the dependence of turbulent coagulation on wind-speed (as it relates to transport time), turbulent kinetic energy dissipation rate per unit mass, aerosol density and size distribution, and atmospheric diffusion.
- The analytic representation of collection kernels for existing turbulent coagulation theories is based on spherical particles. While this may not be valid for KOF, it is necessary to have a simple way of estimating the changes in coagulation dynamics as a function of aerosol shape.

For mathematical simplicity, nearly all analyses involving the transport of particles from large fires have assumed that these particles have a spherical shape. Since a sphere has the smallest surface-to-volume (mass) ratio, these models usually place a lower bound on the reaction rate per mass unit. However, unlike a forest fire, in which roughly half of the condensed smoke particles are liquid and can be described as spheres, an oil fire presents a particular problem since most of the pure soot particles are long strands of solid carbon or more complex molecules and cannot really be described as spheres.
Evans et al. (Ref. 3) have examined the particle size and shape distribution for oil fires without interference from brine, water, nebulized tar, or dusty ambient aerosols and have shown that these smoke particles are an agglomeration of individual spherules that exhibit a strand-like behavior. A sample of their results is shown in Figure I-1, which indicates that large numbers of strand-like particles may have existed in the many plumes from the KOF. This particle configuration would have an important effect on the time scale for kinetic processes and chemical reactions.

In recent years, more attention has been paid to the fractal description of the irregular structures shown in Figure I-1 and on the effects of such irregularities on free molecular agglomeration (Ref. 4). However, for the purposes of this study, we will assume a spherical shape for all particles and apply a correction factor when more accurate analysis becomes available.

![Figure I-1. Electron Micrograph of a Smoke Particle From a 1.0 m Diameter Murban Crude Oil Fire (Source: Ref. 3)](image-url)
It should be emphasized that the selection of the turbulent coagulation processes as the first of the aging processes to be considered does not imply that the others have been deemed to be less important. Indeed, we plan to address the other aging processes in a subsequent publication in which we will make use of the mathematical formalism established in this study.

In Section II of this paper, we evaluate the dependence of turbulent shear coagulation and turbulent inertial coagulation on aerosol concentration and mass density, size distribution, and turbulence parameters. These considerations are then applied to the KOF in Section III, in which the effects of windspeed, atmosphere turbulence, and aerosol emission rate are taken into account. Concluding remarks are rendered in Section IV.
II. TURBULENT COAGULATION PROCESSES

Because the conditions of the KOF have not been encountered before, we have a limited data base for predicting the spatial regions and conditions for which turbulent coagulation may be important. It is also unlikely that simple transport codes will be valid for this type of assessment. One of the reasons for examining the significance of turbulent coagulation in the KOF is that the initial reported particle concentrations are extremely high. For example, Hobbs and Radke (Ref. 5) report particle concentrations frequently exceeding $10^5$ cm$^{-3}$ within a few kilometers from the fires and mass concentrations of the composite plume ~840 µg m$^{-3}$ at 20 km downwind for particles <3.5 µm in diameter. Johnson et al. (Ref. 6) report mass densities between 500-1000 µg m$^{-3}$ at distances greater than 200 km. These observations suggest that coagulation processes could be important over a significant spatial region. In this section and in Section III, we examine this conjecture.

Recently, Porch (Ref. 7) has examined the impact of turbulent coagulation on the size distribution of aerosols produced by large fires in an attempt to understand the phenomenon of Blue Moons. Porch used models where initial mass concentrations for spherical smoke particles with radii less than 1 µm ranged from $5 \times 10^{-9}$ to $5 \times 10^{-8}$ g cm$^{-3}$ (or equivalently 5000 to 50,000 µg m$^{-3}$). These estimates appear to apply over the fire itself, represented in Porch's model by a diameter of 400 km. In an earlier study, Porch, Penner, and Gillette (Ref. 8) conducted a numerical study of the effects of super-µm particles on the coagulation loss of submicron particles. The results of these studies have shown that for particles with radii less than 0.1 µm Brownian coagulation is more rapid than turbulent coagulation, while the latter mechanism generally dominates for particle sizes of a few microns (Ref. 9).

The basic model for turbulent coagulation was developed by Saffman and Turner (Ref. 10), who identified the two distinct coagulation mechanisms: turbulent shear coagulation and turbulent inertial coagulation. Pruppacher and Klett (Ref. 9) have succinctly summarized the salient features of turbulent coagulation. The first significant feature is that turbulent coagulation applies to those aerosols of radius $r \ll \lambda_k$ where $\lambda_k$ is the "Kolmorogov microscale length" (Refs. 9, 10). As shown in Table II-I, this is

II-1
satisfied for the conditions of KOF, in which virtually all of the aerosol particles have dimensions less than the indicated values of $\lambda_k$.

Table II-1. Kolmogorov Microscales as a Function of Energy Dissipation Rate, $\varepsilon$

<table>
<thead>
<tr>
<th>$\varepsilon$ (cm$^2$ s$^{-3}$)</th>
<th>$\lambda_k$ (cm)</th>
<th>$\tau_k$ (s)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>$1.8 \times 10^{-1}$</td>
<td>$1.9 \times 10^{-1}$</td>
<td>Applicable to stratus clouds where there is small mean velocity (Ref. 10)</td>
</tr>
<tr>
<td>100</td>
<td>$8.4 \times 10^{-2}$</td>
<td>$4.1 \times 10^{-2}$</td>
<td>Cloudy air (Ref. 9)</td>
</tr>
<tr>
<td>1000</td>
<td>$4.8 \times 10^{-2}$</td>
<td>$1.3 \times 10^{-2}$</td>
<td>Relevant for conditions in turbulent cumulus clouds (Ref. 10)</td>
</tr>
<tr>
<td>2000</td>
<td>$4.0 \times 10^{-2}$</td>
<td>$9.3 \times 10^{-3}$</td>
<td>Relevant to early stages of the plume in large fires (Ref. 7)</td>
</tr>
<tr>
<td>8000</td>
<td>$2.8 \times 10^{-2}$</td>
<td>$4.6 \times 10^{-3}$</td>
<td>Exceptional case of turbulent shear in high turbulence (Ref. 8)</td>
</tr>
</tbody>
</table>

Coagulation results from the velocity motion between particles. One method of achieving relative motion is through significant small-scale velocity gradients in turbulent flow. For the length scale of velocity gradients that are smaller than $\lambda_k$, the coagulation process is similar to that of the laminar shear flow case considered by Smoluchowski and described in Pruppacher and Klett (Ref. 9). If $m_1$, $m_2$ are the masses of the coagulating particles, and $r_1$ and $r_2$ are their respective radii, and $\rho$ is the material intrinsic density (assumed the same for both/all particles), and

$$m = \frac{4}{3} \pi \rho r^3,$$  \hspace{1cm} (2.1)

then the collection kernel for $turbulent$ $shear$ $coagulation$ is given by (Refs. 9, 10):

$$K_s(m_1, m_2) = 1.3(r_1 + r_2)^3 \tau_k^{-1} \text{ cm}^3 \text{ s}^{-1},$$  \hspace{1cm} (2.2)

where $\tau_k$ is the "Kolmogorov time scale" (Refs. 9, 10).

Another method for producing a relative particle velocity is due to local turbulent accelerations. Particles of different mass respond differently to these accelerations by having different viscous relaxation times associated with velocity equilibration. This leads to the following expression for the collection kernel for $turbulent$ $inertial$ $coagulation$: II-2
\[
K_1(m_1, m_2) = \frac{2\pi \rho}{\rho_A} \left( r_1 + r_2 \right)^2 \left| r_1^2 - r_2^2 \right| \tau_k^{-1} \lambda_k^{-1} \text{cm}^3\text{s}^{-1}.
\]

(2.3)

In this equation, \( \rho_A \) is the air mass density, and \( \lambda_k \) and \( \tau_k \) are the Kolmogorov microscales for length and time, which are given by

\[
\lambda_k = \left( \frac{v^3}{\varepsilon} \right)^{1/4} \text{cm}
\]

(2.4)

\[
\tau_k = \left( \frac{v}{\varepsilon} \right)^{1/2} \text{s},
\]

(2.5)

where \( \varepsilon \) is the dissipation rate of kinetic energy per unit mass \((\text{cm}^2\text{s}^{-3})\) and \( v \) is the kinetic viscosity of air \((\text{cm}^2\text{s}^{-1})\). For the altitude range used in our study, we take the U.S. Standard Atmosphere values of \( \rho_A = 10^{-3} \text{g cm}^{-3} \) and \( v = 0.172 \text{ cm}^2\text{s}^{-1} \).

As observed in Eqs. (2.4) and (2.5), the dissipation rate is a key parameter. Table II-I gives values of \( \lambda_k \) and \( \tau_k \) for selected values of \( \varepsilon \). Examination of this table shows that values of \( \varepsilon \) ranging from 2000 to 8000 \( \text{cm}^2\text{s}^{-3} \) are being considered in this study.

Some of the important mathematical and physical characteristics of the coagulation process can be presented best by considering the time behavior of the aerosol distribution in a spatially uniform medium. We let \( f(m, t) \, \text{dm} \) be the number of particles per unit volume lying in the mass range between \( m \) and \( m + \text{dm} \), where \( f(m, t) \) is defined as the mass density function \((\text{MDF})\). Using \( K(m_1, m_2) \) as the general description for either of the collection kernels [(2.2) or (2.3)] gives the following equation for the time evolution of \( f(m, t) \):

\[
\frac{df}{dt} = \frac{1}{2} \int_{m_1}^{m_2} \int_{m_1}^{m_2} K(m_1, m_2) f(m_1) f(m_2) \delta(m - m_1 - m_2) \, \text{dm}_1 \, \text{dm}_2
\]

\[-f(m) \int_{m_2}^{m} K(m, m_2) f(m_2) \, \text{dm}_2.
\]

(2.6)

The first term in Eq. (2.6) is the growth term. The \"1/2\" appearing in front of the integral is due to the fact that there is a reduction of one particle when two particles coagulate. Inclusion of the Delta function \( \delta(m - m_1 - m_2) \) selects from all collisions only those in which the mass, \( m \), equals \( m_1 + m_2 \). It is possible to integrate the growth term
over either of the dummy variables \( m_1 \) or \( m_2 \) in Eq. (2.6) and simplify the integral to the more familiar form (Ref. 7),

\[
\frac{1}{2} \int \int K(m_1, m_2) f(m_1) f(m_2) \delta(m - m_1 - m_2) \, dm_1 \, dm_2
\]

which is obtained by integration over \( m_1 \). The second term in Eq. (2.6) is the loss due to coagulation and is the integration over all possible events.

A further condition that is not explicitly represented in the expressions for \( K_S \) and \( K_I \) is the existence of an efficiency factor associated with coagulation. For usual turbulent clouds, Saffman and Turner (Ref. 10) suggest that the probability of coagulation may be close to unity when particles are about the same size but diminishes rapidly for particles of very different size. The coagulation efficiency of the KOF is not clear since the particles are not water droplets. The issue of collection efficiency is difficult to address in this preliminary assessment, particularly since many of the KOF particles may not be spherical. Hence, for the present we will not include this consideration in our model.

For this study, we assume an efficiency of unity for all colliding particles. We also assume that all particles are spherical since the collection kernels have only been modeled for this case. A preliminary estimate of the collection efficiency for nonspherical particles suggests that it would be greater than that for spherical particles of the same mass.

A more precise evaluation of the collection kernel(s) could perhaps be obtained by extending the Saffman and Turner (Ref. 10) analysis to more realistic shapes. However, until such an analysis becomes available, we will use the spherical models. At the end of the analysis, it is possible to apply a correction factor to account for enhancement in the coagulation process. Although we do not explicitly include this correction factor in this analysis, it is used in the discussion of results.

\[\text{II-4}\]
Using Eq. (2.6), we can make certain observations about the coagulation dynamics to obtain an estimate of the relevant time constants as a function of particle density and dissipation rate. We first note that the mass density, \( M \), where

\[
M = \int m f(m) \, dm ,
\]

remains invariant. This can be demonstrated by multiplying both sides of Eq. (2.6) by \( m \) and then integrating over all mass space. Thus, we have

\[
\frac{\partial M}{\partial t} = \frac{1}{2} \int_{m_1}^{m_2} \int K(m_1, m_2) (m_1 + m_2) \, dm_1 \, dm_2 
- \int_{m_1}^{m_2} m K(m, m_2) \, dm \, dm_2 .
\]

The first term in Eq. (2.9) was obtained through the integration \( \int m \delta(m - m_1 - m_2) \, dm \). Since \( K(m_1, m_2) \) and \( K(m, m_2) \) are symmetric kernels, the right-hand side of Eq. (2.9) equals zero and thus

\[
\frac{\partial M}{\partial t} = 0 \tag{2.10}
\]

as expected.

The total number of particles per \( \text{cm}^3 \) is defined as

\[
N(t) = \int f(m, t) \, dm \tag{2.11}
\]

and is determined from Eq. (2.6) by integration over mass space. We obtain

\[
\frac{\partial N}{\partial t} = \frac{1}{2} \int_{m_1}^{m_2} \int K(m_1, m_2) f(m_1) f(m_2) \, dm_1 \, dm_2 
- \int_{m_1}^{m_2} \int K(m_1, m_2) f(m_1) f(m_2) \, dm_1 \, dm_2 
= - \frac{1}{2} \int_{m_1}^{m_2} \int K(m_1, m_2) f(m_1) f(m_2) \, dm_1 \, dm_2 . \tag{2.12}
\]

Improved insight into the kinetics of coagulation is gained by introducing a normalized mass density function (NMDF), \( \phi(m, t) \), through the equation
\[ f(m, t) = N(t) \phi(m, t) \]  \hspace{1cm} (2.13)

From Eq. (2.11), we deduce the required normalization condition

\[ \int \phi(m, t) \, dm = 1 \]  \hspace{1cm} (2.14)

By introducing \( \phi(m, t) \), we can cast the analysis in terms of those issues that are attributed solely to the total aerosol concentration, \( N(t) \), and those issues that are due to the distribution in mass as given by \( \phi(m, t) \). Substituting Eq. (2.13) into Eq. (2.12) gives

\[ \frac{\partial N}{\partial t} = -\frac{1}{2} N^2 \eta(t) \]  \hspace{1cm} (2.15)

where

\[ \eta(t) = \int \int K(m_1, m_2) \phi(m_1, t) \phi(m_2, t) \, dm_1 \, dm_2 \]  \hspace{1cm} (2.16)

For the special situation in which the collection kernel is assumed to be mass-independent, that is, \( K(m_1, m_2) = K_0 \), a constant, the result is

\[ \frac{\partial N}{\partial t} = -\frac{1}{2} N^2 K_0 \]  \hspace{1cm} (2.17)

The solution to Eq. (2.17) is

\[ N(t) = \frac{N_i}{1 + (N_i K_0 t / 2)^2} \]  \hspace{1cm} (2.18)

where \( N_i \) is the initial concentration.

We will now make an estimate of \( \eta(t) \) using the analytical forms of \( K_S(m_1, m_2) \) and \( K_I(m_1, m_2) \) from Eqs. (2.2) and (2.3) in combination with certain experimental results of Radke et al. (Ref. 11) and as reported by Porch (Ref. 7), which lead to an assessment of \( \phi(m, t) \). Because \( K_S \) and \( K_I \) are easier to work with in terms of aerosol radius instead of mass, it is convenient in computing \( \eta(t) \) to express the mass distribution in terms of the aerosol radius distribution. Introducing a normalized particle radius function (NPRF), \( g(r, t) \), and applying the transformation law for probability frequency functions gives

\[ \phi(m) \, dm = g(r) \, dr \]  \hspace{1cm} (2.19)

where \( m \) and \( r \) are related by Eq. (2.1). As seen in Eq. (2.19), a knowledge of either \( \phi \) or \( g \) determines the other by differentiating Eq. (2.1).
For the turbulent shear coagulation model, the expansion of \((r_1 + r_2)^3\) in Eq. (2.2) gives

\[
K_S = (1.3) \tau_k^{-1} (r_1 + r_2)^3
\]

\[
= (1.3) \tau_k^{-1} \left( r_1^3 + 3 r_1^2 r_2 + 3 r_1 r_2^2 + r_2^3 \right) . \tag{2.20}
\]

Substituting Eq. (2.20) into Eq. (2.16) and using the property that \(K_S\) is symmetric in \(r_1\) and \(r_2\) yields

\[
\eta_s(t) = 1.3 \tau_k^{-1} \left( 2 \bar{r}^3 + 6 \bar{r}^2 \bar{r} \right) .
\]

\[
= 2.6 \tau_k^{-1} \bar{r}^3 \left( 1 + \frac{3 \bar{r}^2 \bar{r}}{\bar{r}^3} \right) , \tag{2.21}
\]

where

\[
\bar{r}^3 = \int r^3 g(r, t) \, dr = \frac{3}{4 \pi} \rho^{-1} \int m \phi(m, t) \, dm = \frac{3}{4 \pi} \rho^{-1} \overline{m} , \tag{2.22}
\]

\[
\bar{r}^2 = \int r g(r, t) \, dr , \tag{2.23}
\]

\[
\bar{r} = \int r g(r, t) \, dr , \tag{2.24}
\]

and

\[
\overline{m} = \int m \phi(m, t) \, dm . \tag{2.25}
\]

\(\overline{m}\) is the average mass of all particles as can be seen from the definition

\[
\overline{m} = \frac{\int m f(m, t) \, dm}{N(t)} = \frac{M}{N(t)} = \int m \phi(m, t) \, dm . \tag{2.26}
\]

Let us temporarily assume that \(3 \bar{r}^2 \bar{r} / \bar{r}^3\) is much less than unity [we will partially substantiate this assumption using Radke's data (Ref. 11)]. Then \(\eta_s(t)\) is approximately given by

\[
\eta_s(t) = 2.6 \tau_k^{-1} \frac{3}{4 \pi} \rho^{-1} \overline{m} . \tag{2.27}
\]

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Substituting Eq. (2.27) into Eq. (2.15) gives
\[ \frac{\partial N}{\partial t} = -\frac{0.31}{\tau_k \rho} (N \overline{m}) N \] \tag{2.28}

On the other hand, since the total mass density, \( M = N \overline{m} \), remains constant during the coagulation process, turbulent shear coagulation can be approximated by the simple equation

\[ \frac{\partial N}{\partial t} = -\frac{N}{T_S} \], \tag{2.29}

where
\[ T_S = \frac{3.2 \tau_k \rho}{M} \]. \tag{2.30}

The smallest values of \( T_S \) correspond to small values of \( \tau_k \) (high levels of turbulence) and large values of \( M \). Using \( M = 5 \times 10^{-8} \text{ g cm}^{-3} \), \( \rho = 1.0 \text{ g cm}^{-3} \), and \( \tau_k = 4.6 \times 10^{-3} \text{ s} \) (corresponding to \( \epsilon = 8000 \text{ cm}^2 \text{ s}^{-3} \)) gives an e-folding time of
\[ T_S = 2.9 \times 10^5 \text{ s} = 81 \text{ hr} \], \tag{2.31}

which appears quite long when compared to the time scales of interest in the KOF.

Let us now return to evaluating the term
\[ 3 \overline{r^2} \overline{r}/\overline{r}^3 = \Delta \], \tag{2.32}

which first appeared in Eq. (2.21) and was subsequently neglected in the derivation of Eq. (2.28). \( \Delta \) may be considered a "correction factor" and in the approximation of Eq. (2.27) has been neglected. However, as shown in Eqs. (2.22) to (2.24), each of the entities, \( \overline{r}, \overline{r^2}, \) and \( \overline{r^3} \), involved in the computation of \( \Delta \) is time dependent since \( g(r, t) \) changes with time. Essentially, \( \Delta(t) \) cannot be determined exactly without solving the problem exactly. However, it is interesting to explore the conditions under which \( \Delta \) is negligible and under which it is greater than unity, the latter condition requiring modification of Eq. (2.28).

An estimate of \( \Delta \) can be obtained using measurements of what Radke (Ref. 11) calls the *Number Size Distribution*. In his notation, this quantity is labeled "dN/dR" (see Figure II-1) and is defined as the number of particles per unit radius per unit volume. In our formalism, we express this quantity as \( N \ g(r) \) and have

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\[
\frac{dN}{dR} = N g(r) . \tag{2.33}
\]

The solid line in Figure II-1 is \(\frac{dN}{dR}\) at the beginning phases of a forest slash fire. The dashed line in this figure is an approximation of \(\frac{dN}{dR}\), which is necessary for the approximate analytical assessment used in our study. The approximation of Figure II-1 applies in the radius range extending from a lower limit of \(2 \times 10^{-6}\) cm to an upper limit of \(2 \times 10^{-2}\) cm and describes a distribution that varies as \(r^{-3}\).

We should emphasize that the Number Size Distribution of Figure II-1 is taken at an instant of time. However, during the coagulation process, \(g(r, t)\) will change with time as the size distribution shifts to larger particles (Ref. 7). In addition, the \(g(r)\) of this figure applies to a forest slash fire and would likely be different from the \(g(r)\) of the KOF. Nevertheless, the use of \(g(r)\) from Figure II-1 provides a mathematical basis for examining the importance of the radial shape function on coagulation dynamics, and, therefore, we assume that \(g(r)\) for the KOF is similar to the result for Figure II-1.
The sensitivity of the results to the analytic representation of the aerosol distribution can be explained by proposing that $g(r)$ for the KOF varies as $r^{-s}$ between a lower limit, $r_L$, and an upper limit, $r_U$. This form of the approximation for $g(r)$ is chosen because of its mathematical simplicity and because it is representative of aerosol size distributions found in nature before the onset of significant aging caused by coagulation. For this case, we have

$$g(r) = A r^{-s}, \quad (2.34)$$

where $A$ is the normalization constant determined from the requirement that $\int_0^\infty g(r) \, dr = 1$. Applying the normalization condition gives

$$A = (s-1) r_L^{s-1} \left( 1 - (r_L/r_U)^{s-1} \right)^{-1}. \quad (2.35)$$

From Figure II-1, we have $r_L = 2 \times 10^{-6}$ cm and $r_U = 2 \times 10^{-2}$ cm. The ratio $r_L/r_U$ equals $10^{-4}$ and is therefore negligible. In this case, we have the approximation

$$A = (s-1) r_L^{s-1}. \quad (2.36)$$

For expediency, we also assume that Eq. (2.36) applies for the KOF, although this assumption can readily be modified using actual data. Lastly, it should be stated that Eq. (2.34) implies a time-invariant size distribution, which is clearly not the case. Despite this limitation, Eq. (2.34) provides a basis for understanding the important features of the size distribution in regard to the coagulation process.

Equation (2.34) readily furnishes the expression for the $n$th moment:

$$\bar{r^n} = \int_{r_L}^{r_U} r^n g(r) \, dr = \left( \frac{r_L^n}{n+1-s} \right) \left( Q^{n+1-s} - 1 \right), \quad (2.37)$$

where

$$Q = (r_U/r_L). \quad (2.38)$$

The moments $\bar{r}$, $\bar{r^2}$, and $\bar{r^3}$ are readily computed from Eq. (2.37). Inserting the resulting expressions into Eq. (2.32) gives
\[ \Delta = \frac{3(4-s)}{(2-s)(3-s)} \frac{(Q^{2-s}-1)(Q^{3-s}-1)}{(Q^{4-s}-1)}. \]  

(2.39)

We now examine the dependence of \( \Delta \) on \( s \) in the range where \( Q >> 1 \). These results are shown in Table II-2 and are obtained by taking the limit of Eq. (2.39) for \( Q = r_U/r_L >> 1 \). This is the range of interest for forest fires but also appears to be the range of interest for KOF, based on preliminary data obtained from Baumgardner (Ref. 12). It should be noted that the expressions in Table II-2 cannot be applied at the integer values of \( s = 2, 3, 4 \). In order to compute \( \Delta \) at these values, the limiting form of Eq. (2.39) at \( s = 2, 3, 4 \), respectively, must be taken using L'Hopital's rule. Figure II-2 provides a broader perspective of the dependence of \( \Delta \) on \( s \) and \( Q \).

<table>
<thead>
<tr>
<th>( s )</th>
<th>( \Delta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 1 &lt; s &lt; 2 )</td>
<td>( \frac{3(4-s)}{(2-s)(3-s)} \left( \frac{r_U}{r_L} \right)^{-s} )</td>
</tr>
<tr>
<td>( 2 &lt; s &lt; 3 )</td>
<td>( \frac{3(4-s)}{(s-2)(3-s)} \left( \frac{r_L}{r_U} \right) )</td>
</tr>
<tr>
<td>( 3 &lt; s &lt; 4 )</td>
<td>( \frac{3(4-s)}{(s-2)(s-3)} \left( \frac{r_L}{r_U} \right)^{4-s} )</td>
</tr>
<tr>
<td>( 4 &lt; s )</td>
<td>( \frac{3(s-4)}{(s-2)(s-3)} )</td>
</tr>
</tbody>
</table>

Examination of Table II-2 shows that the correction factor \( \Delta \) is negligible for \( s > 2 \) since \( r_U/r_L << 1 \). On the other hand, in the range \( 1 < s < 2 \), the variation of \( \Delta \) with \( r_U/r_L \) shows that the correction factor can be several orders of magnitude greater than unity. This is also indicated in Figure II-2. Should \( g(r) \) vary as \( r^{-s} \) with \( 1 < s < 2 \), the time constant, \( T_s \), in Eq. (2.30) could be reduced by several orders of magnitude, rendering turbulent shear coagulation a very important consideration since the effective time scale would be reduced to approximately 1 hour. It is important to note that significant coagulation effects involving time scales under 1 hour and for particles greater than 1 \( \mu \text{m} \) in radius have been
computed for the large Canadian forest fire of 1950 using Radke's results in Figure II-1 (Ref. 7).

![Graph of Δ as a Function of s With Q as a Parameter]

Figure II-2. Δ as a Function of s With Q as a Parameter

Up to this point, we have only considered the contribution from turbulent shear coagulation through use of the collection kernel given by Eq. (2.2). We now consider the contribution from turbulent inertial coagulation in Eq. (2.3). Using Eq. (2.16), the corresponding parameter, η₁, is given by

\[
\eta_1 = B \int_{r_1}^{r_2} \int_{r_1}^{r_2} (r_1 + r_2)^2 |r_1^2 - r_2^2| g(r_1) g(r_2) \, dr_1 \, dr_2 ,
\]

where

\[
B = \frac{2\pi}{9} \frac{\rho}{\rho_A} \frac{1}{\tau_k^2} \
\]

\[
(2.40)
\]

\[
(2.41)
\]
In contrast to the case for turbulent shear coagulation, the double integration over \( r_1, r_2 \) is not separable due to the nonanalytic function \( |r_1^2 - r_2^2| \). However, it is possible to obtain a lower bound on \( \eta_{II} \) in the following way. Consider the integrals \( I_1 \) and \( I_2 \), where

\[
I_1 = \int_{r_1}^{r_2} \int_{r_1}^{r_2} (r_1 + r_2)^2 |r_1^2 - r_2^2| g(r_1) g(r_2) \, dr_1 \, dr_2
\]

and

\[
I_2 = \int_{r_1}^{r_2} \int_{r_1}^{r_2} (r_1 + r_2)^2 (r_1 - r_2)^2 g(r_1) g(r_2) \, dr_1 \, dr_2
\]

\[ (2.42) \]

\[ (2.43) \]

\( I_1 \) appears in Eq. (2.40). \( I_2 \) is similar and has the essential property required of \( I_1 \), namely, ensuring that no contribution comes from the domain where \( r_1 = r_2 \).

We now show that \( I_1 > I_2 \). Let

\[
a = |r_1^2 - r_2^2| = (r_1 + r_2) |r_1 - r_2|
\]

and

\[
b = (r_1 - r_2)^2 = (r_1 - r_2)(r_1 - r_2)
\]

\[ (2.44) \]

\[ (2.45) \]

and consider the difference

\[
\Omega = a^2 - b^2
\]

\[
= (r_1 + r_2)^2 (r_1 - r_2)^2 - (r_1 - r_2)^4 = (r_1 - r_2)^2 (r_1 r_2) \geq 0 \quad . \quad (2.46)
\]

Since \( a, b > 0 \), and \( a^2 \geq b^2 \), we conclude that \( a \geq b \). Therefore,

\[
I_1 > I_2 \quad . \quad (2.47)
\]

A lower bound on \( \eta_{II} \), defined as \( \eta^*_1 \), is thus given by

\[
\eta^*_1 = B I_2 \quad . \quad (2.48)
\]

Use of \( \eta^*_1 \) provides a minimum rate of coagulation by inertial turbulence. The integration of \( I_2 \) yields

\[ \Pi-13 \]
The ratio \( \theta = \left( \frac{r^2}{\mathbf{r}^4} \right) \) is deduced from Eq. (2.37) to yield

\[
\theta = \frac{\left( \frac{r^2}{\mathbf{r}^4} \right)^2}{(3-s)^2} \left( \frac{Q^{3-s} - 1}{Q^{5-s} - 1} \right) \tag{2.50}
\]

Table II-3 shows the dependence of \( \theta \) on \( s \) for the range of interest \( r_U/r_L = Q \gg 1 \), and Figure II-3 shows \( \theta \) as a function of \( s \) with \( Q \) as a parameter. It should be noted that the expressions in Table II-3 cannot be applied at the integer values of \( s = 3, 5 \). At these values of \( s \), \( \theta \) must be computed from Eq. (2.50) using L'Hopital's rule.

<table>
<thead>
<tr>
<th>( s )</th>
<th>( \theta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 &lt; ( s ) &lt; 3</td>
<td>( \frac{5-s}{(3-s)^2} \left( \frac{r_L}{r_U} \right)^{s-1} )</td>
</tr>
<tr>
<td>3 &lt; ( s ) &lt; 5</td>
<td>( \frac{5-s}{(s-3)^2} \left( \frac{r_L}{r_U} \right)^{5-s} )</td>
</tr>
<tr>
<td>( 5 &lt; s )</td>
<td>( \frac{s-5}{(s-3)^2} )</td>
</tr>
</tbody>
</table>

Since the anticipated range of interest is from \( s = 3 \) to \( s = 5 \) and for \( Q \gg 1 \), we conclude from both Table II-3 and Figure II-3 that \( \theta \) can be neglected in comparison to unity and thus use the approximation

\[
\frac{1}{2} \mathbf{r}^4 = \frac{2r^4}{(5-s)} \left( Q^{5-s} - 1 \right) \tag{2.51}
\]
Using the results from Table II-2 and neglecting the contribution from $\Lambda$ for $s > 2$ provide a comparison of the relative contribution for shear and inertial coagulation. For $s > 2$, we have

$$\eta_1 = 2.6 \tau_k^{-1} r^3$$  \hfill (2.52)

and

$$\eta^*_{1} = \frac{4\pi}{9} \frac{\rho}{\rho_A} \tau_k^{-1} \lambda_k^{-1} \frac{s}{r^4}.$$  \hfill (2.53)

and the ratio $\eta^*_{1}/\eta_{S}$ is given by

$$\frac{\eta^*_{1}}{\eta_{S}} = (0.54) \left( \frac{\rho}{\rho_A} \right) \left( \frac{4-s}{5-s} \right) \frac{\tau_L}{\lambda_k} \frac{Q^{5-s} - 1}{Q^{4-s} - 1}.  \hfill (2.54)$$

In the subsequent calculations, we use $\rho = 1 \text{ g cm}^{-3}$ for the aerosol particles, and $\rho_A = 10^{-3} \text{ g cm}^{-3}$. There is dramatic variation of $\eta^*_{1}/\eta_{S}$ as a function of $s$. This quantity...
can vary over several orders of magnitude, depending on the ratio $r_U/r_L$ (as long as $r_U/r_L >> 1$). For example, when $2 < s < 4$, Eq. (2.54) reduces to

$$\frac{\eta^*_I}{\eta^*_S} = (540) \frac{(4-s)}{(5-s)} \frac{r_U}{\lambda_k} , \quad (2.55)$$

while for $s > 5$ we have

$$\frac{\eta^*_I}{\eta^*_S} = (540) \left( \frac{s-4}{s-5} \right) \frac{r_L}{\lambda_k} . \quad (2.56)$$

The ratio between Eqs. (2.55) and (2.56) is approximately given by $r_U/r_L$, which may range several orders of magnitude (e.g., $r_U/r_L = 10^4$ in Figure II-1).

It is interesting to examine $\eta^*_I/\eta^*_S$ in the range $2 < s < 4$ for a representative large value of dissipation rate. Using the value $\lambda_k = 4 \times 10^{-2} \text{ cm}$ from Table II-1, corresponding to $\varepsilon = 2000$, and $r_U = 2 \times 10^{-2}$ and the value $s = 3$ deduced from Figure II-1 gives

$$\frac{\eta^*_I}{\eta^*_S} = \frac{(540)}{4} = 135 . \quad (2.57)$$

If $\eta^*_I$ were to be used in the formalism of Eqs. (2.28) to (2.30), the time constant would be $T_I$ instead of $T_S$ and would be given by

$$T_I = \frac{T_S}{135} = \frac{2.9 \times 10^5}{135} = 2.1 \times 10^3 \text{ s} = 35 \text{ min} . \quad (2.58)$$

The results show that inertial coagulation is the dominant mechanism for particle frequency functions that contain a substantial fraction of particles greater than 1 $\mu$m. This conclusion is also consistent with the general observations made by Pruppacher and Klett (Ref. 9).
III. APPLICATION TO KOF

The purpose of this section is to examine the geometric boundary(ies) of KOF where turbulent coagulation no longer contributes to the aging of aerosols. When combined with the results of other aerosol mass changing processes, this analysis will contribute to defining the overall spatial region(s) of KOF where aerosols no longer experience mass changing. These are the only regions where it is meaningful to compare the concentration prediction capability of current-generation particle transport codes (which do not include aging effects) to experimental results.

Figure III-1 depicts the KOF plume behavior for turbulent coagulation considerations. This figure portrays three individual plumes merging into a superplume. The actual KOF aerosol problem is one in which the particle concentration of a superplume is forged by the merging of hundreds of individual plumes.

![Diagram of geometric viewpoint for coagulation considerations](image)

Figure III-1. Geometric Viewpoint for Coagulation Considerations

In this figure, we hypothesize that there will be significant coagulation effects as the flames emerge from each individual well since the concentration/mass density will be extremely high. As the individual plumes spread, the concentrations will decrease by geometric dilution caused by atmospheric turbulence and coagulation. Ultimately, we propose that the coagulation process becomes insignificant due to decreasing particle concentration.
concentrations. The surfaces that define this transition are labeled "Coagulation Boundary." The shaded areas beyond these boundaries are the regions where coagulation is assumed to be insignificant.

Figure III-1 suggests the possibility that the merging plumes may again produce conditions for coagulation; hence, another Coagulation Boundary is shown in the superplume. However, Sykes (Ref. 13) has questioned the possibility of a secondary coagulation boundary for the merged plumes on the grounds that this could not occur without some mechanism for concentrating particles locally. This issue will be resolved when detailed and reliable data are available from the field experiment.

The specific objective of this section is to develop a mathematical model for predicting the downwind aerosol density for either an individual fire or a merged plume and, from this model, to determine the Coagulation Boundaries shown in Figure III-1. The analytical model is developed by combining the turbulent coagulation kinetics described in Section II with particle transport and incorporating advection and turbulent diffusion.

In Section II, we showed that the aerosol concentration for a spatially uniform medium is given by

$$ \frac{dN}{dt} = -\frac{1}{2} N^2 \eta(t) \ , \quad (3.1) $$

where $\eta(t)$ can be written as

$$ \eta(t) = \int_{r_1}^{r_2} \int_{r_1}^{r_2} K \left[ m_1(r_1), m_2(r_2) \right] g(r_1, t) g(r_2, t) \, dr_1 \, dr_2 \ . \quad (3.2) $$

Equation (3.2) is the same as Eq. (2.16) but is now written in terms of the radius variables. Summarizing the results from Section II, we have the following for turbulent shear coagulation:

$$ \eta_s = 2.6 \tau_k^{-1} \bar{r}^3 (1 + \Delta) \ , \quad (3.3) $$

where

$$ \Delta = 3 \bar{r}^3 \bar{r} \bar{r}^3 \ . \quad (3.4) $$

For turbulent inertial coagulation, we have

$$ \eta_i^* = 4\pi \frac{9}{\rho} \frac{\rho}{\rho_A} \frac{\bar{r}^3}{\tau_k^3} \bar{r} \left( 1 - \theta \right) \ , \quad (3.5) $$
where

$$\theta = \left( \frac{\tau^2}{r^4} \right).$$  \hspace{1cm} (3.6)

It should be noted that Eq. (3.3) is a restatement of Eq. (2.21) and Eq. (3.5) is a restatement of Eq. (2.48) using the exact expression of \( I_2 \) of Eq. (2.49) instead of neglecting \( \theta \).

As indicated in Eqs. (3.3) to (3.6), a knowledge of the first four moments of \( g(r, t), \bar{r}, r^2, r^3, \) and \( r^4 \), precisely determines the time evolution of the aerosol density. It should be stressed that these quantities apply to any functional form of \( g(r, t) \). Thus, any measurements of aerosol properties that could furnish information regarding the first four moments would be sufficient to determine the aerosol dynamics. The situation becomes simplified, somewhat, when we have further knowledge of \( \Delta \) and \( \theta \).

Although it is not explicitly stated, Eq. (3.1) applies to the case in which an initial number of particles, \( N_0 \), are inserted into the system at time \( t = 0 \). The concentration at subsequent times is given by

$$N = \frac{N_0}{1 + N_0 \tau/2},$$  \hspace{1cm} (3.7)

where

$$\tau = \int_0^t \eta (\tau') \, dt'.$$  \hspace{1cm} (3.8)

Equations (3.7) and (3.8) are deceptively simple because they assume a knowledge of \( g(r, t) \). As we mentioned at the outset of Section II, \( g(r, t) \) cannot be determined without solving the entire problem. However, estimates of the significant times scales involved in the coagulation process can be obtained by using approximate forms of \( g(r, t) \) or real-time measurements of \( g(r, t) \). It is through the latter that we consider turbulent coagulation in the context of the KOF.

In the remainder of this section, we will calculate the aerosol concentration as the particles emerge from a single well. The mathematical model developed for this case is directly applicable to the superplume, with the appropriate change of coordinates.

III-3
When the small-scale turbulent fluctuations are averaged out, the Eulerian equation for the aerosols is given by the extension of Eq. (2.6) to now include convection. We have

$$f(m, m_1, m_2) \frac{dm_1 dm_2}{dm_2}$$

$$\text{where} \ \bar{U} \ \text{is the average velocity and} \ \nabla \ \text{is the gradient operator. The right-hand side of Eq. (3.9) is the same as that of Eq. (2.6). For brevity, we have suppressed the explicit dependence of} \ f \ \text{on the space variable,} \ \bar{R}, \ \text{and on time,} \ t. \ \text{For example,} \ f(m) \ \text{is really} \ f(m, \bar{R}, t).$$

For an individual plume, Eq. (3.9) is solved by matching the solution to the source emission at the origin (location of the burning well). If the source emission does not vary with time, Eq. (3.9) will also be time invariant and, hence,

$$\frac{\partial f(m, \bar{R}, t)}{\partial t} = 0 \ . \ \ (3.10)$$

The space dependence of \( f(m, \bar{R}) \) is found by constructing a simplified geometric model of the plume as shown in Figure III-2. In this figure, we approximate the plume as a tube of variable cross section, \( A(q) \), in which all the particles move with average velocity, \( \bar{U} \), normal to the surface, \( A \). The concentration is assumed to be uniform across the area. The coordinate system is constructed so that the \( q \)-axis is always directed along the direction of \( \bar{U} \). If \( |\bar{U}| \) is the magnitude of \( \bar{U} \) along the \( q \)-axis, the divergence term in Eq. (3.9) becomes

$$\nabla \cdot [\bar{U} f(m, \bar{R})] = \frac{2}{A(q)} \left[ U(q) f(m, q) \right] \ \ (3.11)$$

since, by construction, \( f(m, q) \) does not vary in directions normal to \( q \).

We now let

$$f(m, q) = N(q) \phi(m, q) \ \ (3.12)$$

where \( N(q) \) is the aerosol concentration (cm\(^{-3}\)) and \( \phi(m, q) \) is the NMDF (expressed as a function of the space coordinate, \( q \), instead of a function of time).
Using Eqs. (3.10) and (3.11) in Eq. (3.9) and then integrating over mass space using the techniques developed in Section II gives

\[
\frac{d}{dq} \left( \text{ANU} \right) = -\frac{(\text{ANU})}{U T(q)},
\]

where the q-dependent time "constant," \( T(q) \), is given by

\[
T(q) = \frac{2}{N(q) \eta(q)}
\]

and

\[
\eta(q) = \int_{r_1}^{r_2} \int_{r_2}^{r_3} K \left[ m_1(r_1) m_2(r_2) \right] g(r_1, q) g(r_2, q) dr_1 dr_2.
\]

The expressions for \( \eta \) corresponding to turbulent shear and turbulent inertial coagulation are given by Eqs. (3.3) and (3.5), respectively. As in Section II, we have also introduced an NPRF, \( g(r, q) \), through the transformation

\[
\phi(m, q) dq = g(r, q) dr.
\]

Further simplification is possible by using the conservation of mass. Multiplying Eq. (3.9) by \( m \) and then integrating over mass space and area gives
\[ \frac{\partial}{\partial q} \left( U(q) \bar{M}(q) A(q) \right) = 0, \quad (3.17) \]

where \( \bar{M}(q) \) is the mass per cm\(^3\). Equation (3.17) is analogous to Eq. (2.10). \( \overline{UMA} \) is the total mass emission rate (g s\(^{-1}\)) from a burning well, and as long as the flux tube of Figure III-2 encompasses all the mass, we must have the equality

\[ U(q) \bar{M}(q) A(q) = R_m, \quad (3.18) \]

where \( R_m \) is the mass emission rate from the well expressed in g s\(^{-1}\).

The next step in determining \( N(q) \) is to evaluate \( A(q) \). We assume a circular plume with radius, \( R_o \), and area,

\[ A(q) = \pi R_o^2(q), \quad (3.19) \]

\( R_o^2 \) is approximated by the equation

\[ R_o^2 = 2 \int_0^t D(t') \, dt', \quad (3.20) \]

where \( D \) is the turbulent diffusion constant evaluated along the path. Following basic theoretical arguments discussed by Tennekes and Lumley (Ref. 14), we assume the relationship

\[ D(t) \sim R_o(t) U(t), \quad (3.21) \]

which applies for times long enough for many independent eddies to establish fully developed turbulence.

With this assumption, Eq. (3.20) can be written as a precise equation:

\[ D = a R U, \quad (3.22) \]

where "\( a \)" is an experimentally deduced constant, as yet undetermined. Inserting Eq. (3.21) into Eq. (3.20) and differentiating the latter gives

\[ \frac{dR_o}{dt} = a U(t). \quad (3.23) \]

Integrating Eq. (3.23) along the path yields

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\[ R_0(q) = a \int_0^t U(t) \, dt = a \, q \quad (3.24) \]

For the turbulent diffusion case considered, the radius of the plume is observed to be proportional to the distance traveled.\(^1\)

In the regions described by Eq. (3.21), it is recognized that Eq. (3.24) is an approximation since the diffusion rates for horizontal and vertical diffusion are different. For the KOF, it has been observed that the plumes are typically constrained between 1 and 3 km in thickness. When the top of plume slows its rise and stabilizes from the initial buoyancy effect, the path variable \( q \) can be replaced by the horizontal axis in the direction of flow, \( x \); the vertical dimension can be replaced by a constant value, \( \Delta h = 2 \) km; and the horizontal dimension normal to the flow can be allowed to grow at a turbulent diffusion rate given by

\[ y = bx \quad (3.25) \]

where \( b \) is another experimentally deduced quantity. If it is desired, the approximation for \( A(q) \) could be improved by using real-time KOF experimental data.

However, until such KOF experimental data become available, we will use the circular model described by Eq. (3.24) with the constant "a" determined from previous experimental data. Figure III-3 shows the horizontal mean cloud half-width, \( \sigma_y(x) \), for a stack plume as a function of downwind distance, \( x \), from the source (Refs. 15, 16). Lines A through F correspond to different levels of stability. Taking the "average" of the curves gives the approximate relationship

\[ \sigma_y = 0.1 \, x \quad (3.26) \]

Identifying the \( \sigma_y \) and \( x \) of Eq. (3.26) with the \( R_0 \) and \( q \), respectively, of Eq. (3.24) gives

\[ R(q) = 0.1 \, q \quad (3.27) \]

and

\[ A(q) = \pi R^2 = 3.14 \times 10^{-2} \, q^2 \, \text{cm}^2 \quad (3.28) \]

\(^1\) I. Sykes (Ref. 13) has pointed out that the linear behavior of Eq. (3.24) is an "early time" result but may well extend to dimensions near global scales. For very long transport times, the horizontal growth will vary with the square root of the distance.

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during the initial stages of plume growth from the burning well. For numerical simplicity, the path length can be measured in kilometers instead of centimeters. We therefore write \( q = 10^5 q_k \), where \( q_k \) is the distance in kilometers from the source, and use the equation
\[
A(q) = 3.14 \times 10^8 q_k^2 .
\]  
(3.29)

We now write \( U = 28 U_w \), where \( U_w \) is the windspeed in km/hr, and insert Eq. (3.29) into Eq. (3.18) to obtain
\[
\overline{M} = \frac{(1.14) \times 10^{-10} R_m}{U_w q_k^2} .
\]  
(3.30)

At this point, it is interesting to estimate the mass densities calculated from Eq. (3.30) for an "average" burning well and compare them with the values used by Porch.
If \( R_M \) is the aerosol emission rate for all the burning wells and \( W \) is the total number of burning wells, then the average mass emission rate per well, \( \bar{R}_m \), is

\[
\bar{R}_m = \frac{R_M}{W} \text{ g s}^{-1}. \tag{3.31}
\]

We do not currently have information regarding the maximum emission rates for the largest individual fires, and there appears to be some uncertainty regarding both the total emission rate, \( R_M \), and the average emission rate. Bauer (Ref. 1) has estimated \( R_M \) to be 45,000 metric tons per day, while Hobbs and Radke (Ref. 5) have estimated \( R_M \) to be 3400 metric tons per day. However, it is not clear whether these numbers apply to the same number of burning wells and other similar conditions. For lack of more complete information, we take the average of the two references and use an emission rate of 24,000 metric tons per day, which converts to \( R_M = 2.8 \times 10^5 \text{ g s}^{-1} \). If we assume that 300 wells are burning,\(^2\) the average emission rate per well is

\[
\bar{R}_m = \frac{2.8 \times 10^5 \text{ g s}^{-1}}{300} = 930 \text{ g s}^{-1}. \tag{3.32}
\]

We may anticipate that all wells will not be burning at the same rate and that some wells may have an emission rate far exceeding that given by Eq. (3.32).

In order to examine a hypothetical "worst-case" scenario, we use a value of \( R_m^* = 9.3 \times 10^3 \text{ g s}^{-1} \) in the calculations, which is an order of magnitude higher than the average of Eq. (3.32). At a distance of 1 kilometer from the well (\( q_k = 1 \)), with a windspeed of 10 km/hr (\( U_w = 10 \)) and well emission rate \( R_m = R_m^* = 9.3 \times 10^3 \text{ g s}^{-1} \), the mass density, \( M \), equals \( 1.06 \times 10^{-7} \text{ g cm}^{-3} \), which is a factor of 2.12 higher than the maximum value of \( 5 \times 10^{-8} \text{ g cm}^{-3} \) used by Porch (Ref. 7). For the same values of \( q_k \) and \( U_w \), but with \( R_m = R_m^* = 930 \text{ g s}^{-1} \), \( M = 1.06 \times 10^{-8} \text{ g cm}^{-3} \), which is consistent with Porch's (Ref. 7) range of interest, \( 5 \times 10^{-9} \) to \( 5 \times 10^{-8} \text{ g cm}^{-3} \). Using \( U_w = 10 \), \( R_m = R_m^* \), but this time using \( q_k = 10 \), gives \( M = 1.06 \times 10^{-9} \text{ g cm}^{-3} \), which is a factor of 4.7 lower than Porch's (Ref. 7) minimum of \( 5 \times 10^{-9} \text{ g cm}^{-3} \). The net result of this discussion is the reassuring observation that the values of \( M \) calculated for a realistic range of conditions are consistent with the values previously considered in a large forest fire.

We now return to the main thrust of this section, which is the determination of aerosol density, \( N(q) \), from Eq. (3.13). Using the result

\[ \text{In the May/June 1991 time period, it is estimated that between 500-600 wells were burning.} \]
in Eq. (3.18) gives
\[ N(q) = \frac{3R_m}{4\pi \rho r^3 \text{AU}} \]  \hspace{1cm} (3.34)

Substituting Eq. (3.34), in conjunction with the expressions for \( \eta(q) \) obtained from Eqs. (3.3) and (3.5), respectively, in Eq. (3.14) gives the following expressions for \( T(q) \) for turbulent shear \( (T_S) \) coagulation and turbulent inertial \( (T_I) \) coagulation:
\[ T_S(q) = \frac{8\pi \rho \text{AU} \tau_k}{7.8 R_m f_s} \] \hspace{1cm} (3.35)
\[ T_I(q) = \frac{6\rho \lambda_k \lambda_k \text{AU}}{R_m r_1} \] \hspace{1cm} (3.37)

where
\[ f_s = 1 + \Delta \] \hspace{1cm} (3.36)

and
\[ r_1 = \frac{r^4 (1 - \theta)}{r^3} \] \hspace{1cm} (3.38)

Using either of the expressions \( T_S(q) \) or \( T_I(q) \) for \( T(q) \) in Eq. (3.13) yields the density along the path. Solving Eq. (3.13) for "ANU" gives
\[ N(q) = \left( A_0 \frac{N_0 U_0}{U(q) A(q)} \right) \Gamma_c \] \hspace{1cm} (3.39)

where
\[ \Gamma_c = e^{-\Lambda(q)} \] \hspace{1cm} (3.40)

and
\[ \Lambda(q) = \int_{q_0}^{q} \frac{dq'}{U(q') T(q')} \] \hspace{1cm} (3.41)
The initial values $A_0$, $N_0$, and $U_0$ are taken at the location $q_0$, where the approximation of Eq. (3.27) is valid or, equivalently, where the initial jetting action and buoyancy effects of the well are dissipated. That is, our turbulent diffusion model only considers those effects attributed to usual atmospheric processes.

For the purposes of this analysis, we must make an assumption about the way $U(q')$ and $T(q')$ are treated in the integration of Eq. (3.41). Until experimental data become available, we approximate these quantities as "path-averaged" constants. On the other hand, we do include the variation of $A(q)$ from Eq. (3.27). The results for turbulent shear ($S$) coagulation and turbulent inertial ($I$) coagulation are as follows:

$$
\Lambda_S = \frac{l_S}{q_0} - \frac{l_S}{q},
$$

(3.42)

where

$$
l_S = \frac{10 R_m f_s}{U^2 \tau_k \rho},
$$

(3.43)

and

$$
\Lambda_I = \frac{l_I}{q_0} - \frac{l_I}{q},
$$

(3.44)

where

$$
l_I = \frac{5.3 R_m r_1}{\rho \tau_k \lambda_k U^2}.
$$

(3.45)

In the absence of coagulation, the spatial distribution of density is given by

$$
N_1(q) = \frac{A_0 N_0 U_0}{U(q) A(q)}.
$$

(3.46)

This spreading behavior of the plume is attributed to atmospheric diffusion. $N_1(q)$ can be estimated from the mass ejection using the equation

$$
\overline{M} = \overline{m} N
$$

(3.47)

in Eq. (3.18). The result is

$$
N_1(q) = \frac{R_m}{\overline{m} U(q) A(q)}.
$$

(3.48)

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$N_1(q)$ is the particle density that would exist in the absence of coagulation. Since $\Gamma_c \leq 1$, coagulation reduces the aerosol concentration beyond ordinary geometric dilution. Coagulation is quantitatively manifested by the parameters $I_S$ or $I_I$, which pertain to turbulent shear coagulation and turbulent inertial coagulation, respectively. If we temporarily let $l = I_S$ or $I_I$, then the coagulation contribution to density can be written as

$$\Gamma_c = \exp\left[-\left(\frac{Uq_o - Uq}{Uq_o}ight)\right],$$

(3.49)

which is observed to be less than or equal to unity for the prescribed range $q \geq q_o$. The density at $q_o$ is

$$N(q_o) = N_1(q_o) = \frac{R_m}{\bar{m} U(q_o) A(q_o)}.$$

(3.50)

It is difficult to theoretically predict $q_o$ since the processes describing the aerosol behavior as the oil emerges from the well are extremely complex. However, estimates of $q_o$ can be obtained from visual observations. While a knowledge of $q_o$ is required for determining the absolute level of aerosol concentration for $q \geq q_o$, it does not appear to be a critical factor in determining whether to consider coagulation effects when interpreting transport behavior. What is important is the spatial variation of $\Gamma_c$ as a function of $q$ when compared to other transport processes.

It is now worthwhile to examine certain features of $N(q)$, including predictions of aerosol density close to the source and the apparent limiting effects of coagulation. Based on crude visual interpretations of the KOF photographs, we estimate the initiation of "conventional" atmospheric diffusion processes to begin between 0.1 and 1.0 kilometers from an individual burning well. For orientation, we compute $N_1(q_o)$ at $q_o = 10^5$ cm (1 km) for a windspeed $U_w = 10$ km hr$^{-1}$, for $R_m = R_m^*$, and for the average mass, $\bar{m} = \left(4\pi \rho r^3/3\right) = 6.7 \times 10^{-13}$ g, as determined from the size distribution defined by Eq. (A.6) of Appendix A. Inserting these values in Eq. (3.48) gives $N_1$ (at 1 km) = $1.6 \times 10^5$ particles per cm$^3$. This value of density appears to be consistent with the results of Hobbs and Radke (Ref. 5) at close-in ranges.

As a matter of theoretical interest, it is worthwhile to examine the behavior of aerosol density as a function of emission rate, $R_m$. By the way of example, we evaluate the asymptotic behavior of $N(q)$. In the limit where $q >> l$ and $q \geq q_o$, the coagulation
function $\Gamma_c$ approaches the constant $\Gamma'_c = \exp(-l/\rho_0)$. From Eqs. (3.43) and (3.45), we note that both $l_S$ and $l_I$ are proportional to $R_m$. Therefore,

$$l = \alpha R_m \quad \text{(3.51)}$$

where $\alpha$ is the proportionality constant [for example, $\alpha = (5.3 \, \tau_1 / \rho \tau_s \lambda U^2)$ in the case of turbulent inertial coagulation]. Substituting Eq. (3.51) into $\Gamma'_c$ yields the following result for the asymptotic behavior:

$$N_{as} = \left( \frac{R_m}{m U(q) A(q)} \right) \exp \left[ - \frac{R_m}{\lambda} \right] \quad \text{(3.52)}$$

where

$$\lambda = \frac{q_0}{\alpha} \quad \text{(3.53)}$$

Keeping all other factors the same, $N_{as}$ has a maximum with respect to $R_m$. This maximum results from the competing factors of increased emission and reduction by coagulation. $N_{as}$ maximizes at $R_m = 1$, with the maximum value given by

$$\left( N_{as} \right)_{max} = \frac{(0.37) \, \lambda}{m U(q) A(q)} \quad \text{(3.54)}$$

As we indicated earlier, the main issue regarding the relevance of turbulent coagulation is to examine its effect on the interpretation of transport behavior. More specifically, we want to ensure that $N(q)$ does not change appreciably due to turbulent coagulation over distances that are significant in the interpretation of transport. This means that we would like the function $\Gamma_c$ to remain essentially constant. This condition is examined most easily by plotting $\Gamma'_c$ as a function of $q$ with $q_0$ and $l$ as parameters.

To obtain an estimate of values of $l$ that are of interest, we calculate $l_S$ and $l_I$ for the following set of parameters $^3$:

- $R_m = R_m^* = 9.3 \times 10^3 \, \text{g s}^{-1}$  
- $\rho = 1 \, \text{gm cm}^{-3}$  
- $U = 278 \, \text{cm s}^{-1} \left( U_w = 10 \, \text{km hr}^{-1} \right)$  

$^3$ Improved values for these parameters will be available when the measured KOF data are consolidated.
Inserting these values into Eqs. (3.43) and (3.45) gives

\[ l_s = 2.6 \text{ m} \]  \hspace{1cm} (3.56)

and

\[ l_I = 0.49 \text{ km} \]  \hspace{1cm} (3.57)

For the conditions of Eq. (3.55), the small value of \( l_s \) indicates that turbulent shear coagulation will not be a factor in transport since \( \Gamma_c \) is essentially constant at \( \Gamma_c = 1 \) for all values of \( q \) of interest. Moreover, it is hard to imagine a set of realistic parameters that could change the foregoing conclusion.

On the other hand, Eq. (3.57) shows that \( l_I \) is in the kilometer range. Considering the approximations and the unknown factors that have gone into the calculation of \( l_I \), it is not inconceivable that \( l_I \) could be as high as 10 km under some set of circumstances. This could have an impact on the selection and interpretation of data used for transport predictions. Turbulent coagulation will have an effect on the prediction and interpretation of concentration if \( \Gamma_c \) varies significantly over the range of interest.

The precise criterion about what constitutes an acceptable degree of spatial variation remains to be defined for the Kuwaiti Oil Fire Field Experiment (KOFFE). From the analytical form of \( \Gamma_c \) (Eq. 3.49), there will be some minimum value of \( q \), defined as \( q_{\text{min}} \), beyond which variations of \( \Gamma_c \) with \( q \) will be very small. Moreover, since the dependence of \( \Gamma_c \) on \( q_o \) only affects the magnitude of concentration through the factor \( \exp[-l/l_{q_{o}}] \), it follows that \( q_{\text{min}} \) will depend only on \( l \).

John Cockayne (Ref. 17) has suggested a possible criterion for determining \( q_{\text{min}} \). Cockayne's criterion is that turbulent coagulation will not be important in the range \( q \geq q_{\text{min}} \) if we have the condition.
\[
\frac{\Gamma_c(2q_{\min})}{\Gamma_c(q_{\min})} \geq 0.9 , \quad (3.58)
\]

where \( q_{\min} \) is the distance beyond which \( \ln \Gamma_c \) changes by less than 10 percent over \( q_{\min} \).

Using Eq. (3.49) in Eq. (3.58), we easily deduce

\[
1.1 \geq e^{l/2q_{\min}} = 1 + l/2q_{\min} , \quad (3.59)
\]

which leads to the result

\[
q_{\min} \geq 5\ l . \quad (3.60)
\]

Thus, if \( l = l_1 = 0.49 \) km is used from Eq. (3.57), we have \( q_{\min} = 2.45 \) km. This value of \( q_{\min} \) indicates that turbulent inertial coagulation ceases to be important for the average single well only 2.45 km downstream. On the other hand, if \( l_1 \) is on the order of 10 km, then turbulent inertial coagulation could be very important at the formation of the superplume.

In the analytical model of this study, we have reduced the dependence of the spatial variation of turbulent coagulation to the two parameters \( q_0 \) and \( l_1 \). Recognizing that there is some uncertainty in the numerical values of these quantities, it is instructive to visually examine the dependence of \( \Gamma_c(q) \) on \( q_0 \) and \( l_1 \). This will provide a preliminary indication of the range of values for \( q_0 \) and \( l_1 \) that may be important in assessment of transport issues. The results are shown in Figures III-4 and III-5. All of these curves begin at \( q = q_0 \).

Figure III-4 shows the behavior of \( \Gamma_c \) as a function of \( q \) for assumed values of \( q_0 = 0.1 \) km and \( l_1 = 0.01, 0.1, \) and 1.0 km, respectively. As predicted, increasing the value of \( l_1 \) for a fixed value of \( q_0 \) leads to increasing reduction of the asymptotic value of \( \Gamma_c \). When \( (l_1/q_0) = 10 \), the asymptotic value of \( \Gamma_c \) is less than \( 10^{-4} \) of its initial value. The case for \( q_0 = 0.1 \) and \( l_1 = 10.0 \) is not shown because the excessive reduction in aerosol density for this set of values diminishes interest in this calculation.

Figure III-5 shows the behavior of \( \Gamma_c \) as a function of \( q \) for assumed values of \( q_0 = 1.0 \) km and \( l_1 = 0.1, 1.0, \) and 10.0 km. These results scale similarly to those in Figure III-4 since the ratios of \( l_1/q_0 \) are the same in each case. In addition, the respective asymptotic values of \( \Gamma_c = \exp - [(l_1/q_0)] \) are also equal. However, as shown in the latter set, the scale is stretched out and leads to significant changes in density extending into the tens-of-kilometers range.
Figure III-4. $\Gamma_c$ as a Function of $q$ for $q_o = 0.1$ km With $\lambda$ as a Parameter

Figure III-5. $\Gamma_c$ as a Function of $q$ for $q_o = 1.0$ km With $\lambda$ as a Parameter
IV. CONCLUSION

In this paper, we focus on the KOF of 1991 to investigate one of several possible mass-changing processes that can limit the applicability of the current generation of transport codes, i.e., turbulent coagulation. The purpose of this paper is to make a preliminary assessment of the conditions where turbulent coagulation is or is not an important factor. Clearly, if airborne particles continue to coagulate in appreciable amounts during the transport phase, they cannot be unequivocally "tagged," and this potentially diminishes the validity of a transport calculation.

The approach is to develop an analytical formulation of the spatial behavior of aerosol density as the particles are carried along the turbulent wind field while simultaneously undergoing coagulation. Using approximations for the shape of the aerosol radial size distribution based on the large Canadian forest fire of 1950, we have been able to compute the time and space behavior of aerosol density for turbulent shear coagulation and turbulent inertial coagulation. The latter is nearly two orders of magnitude more effective than the former for the conditions of the KOF.

For a single source, the analysis shows that the upstream density can be written as $N = N_1N_2$, where $N_1$ is that part due to ordinary atmospheric transport processes and $N_2$ is that part attributed to coagulation. $N_2$ depends on the source of emission, windspeed, atmospheric turbulence, and the characteristics of the coagulation process and behaves as $\exp(-l_1/q)$, where $l_1$ is the characteristic dimension associated with coagulation and $q$ is the upstream distance.

Using approximate numbers for the conditions of the KOF, $l_1$ can range up to several kilometers and, in some cases, may be as large as 10 km. However, in the computation, there are a large number of unknown factors that can be determined from the experimental data. More accurate data would improve the estimate of the coagulation distance and, hence, define more precisely the regions where a self-consistent set of measurements can be integrated with valid transport models.

The theoretical models for the turbulent collection kernels are based on spherical shapes for the aerosol particles. In oil fires, however, the soot particles are usually not
spherical but are more strand-like. Perhaps these particles can be better described using the theory of fractals.

Preliminary evaluation suggests that strand-like particles tend to interact more aggressively. In turn, this interaction would lead to a larger collection kernel that would manifest itself in a large $k_1$ and would make turbulent coagulation an important factor in defining the spatial regions where transport codes can be compared with theoretical models.

Improvements to this initial theory should include fractal considerations and improved approximations for the aerosol size distribution function, turbulence levels, inhomogeneities within the plume, and collection efficiency.
REFERENCES


12. D. Baumgardner (National Center for Atmospheric Research), personal communication.


17. J. Cockayne, (Science Applications International Corporation), personal communication.
APPENDIX A

CALCULATION OF MOMENTS OF NORMALIZED PARTICLE RADIUS FUNCTION WHEN $s = 3$
When $r_U >> r_L$, the normalizing constant, $A$, is given by the approximation

$$A = (s - 1) r_L^{s-1}. \quad (A.1)$$

For $s = 3$, we obtain

$$A = 2 r_L^2 \quad (A.2)$$

and, hence,

$$g(r) = \frac{2r^2}{r^3}. \quad (A.3)$$

Using the fact that $r_U >> r_L$ gives the following results for the first four moments:

$$\bar{r} = A \int_{r_L}^{r_U} \frac{dr}{r^2} = 2 r_L \quad (A.4)$$

$$\bar{r}^2 = A \int_{r_L}^{r_U} \frac{dr}{r} = 2 r_L^2 \ln \frac{r_U}{r_L} \quad (A.5)$$

$$\bar{r}^3 = A \int_{r_L}^{r_U} dr = 2 r_L^2 r_U \quad (A.6)$$

$$\bar{r}^4 = A \int_{r_L}^{r_U} r dr = r_L^2 r_U^2. \quad (A.7)$$

The values of $r_L = 2 \times 10^{-6}$ cm and $r_U = 2 \times 10^{-2}$ cm are used in all of the calculations.