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Stability effects on aerosol size and height distributions

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ABSTRACT

The usual description of the vertical distribution of aerosol concentrations assumes neutral atmospheric stratification. This work examines the influence of atmospheric stability on vertical aerosol concentration profile. A model is developed to describe the aerosol transport in the atmospheric surface layer. The model describes a balance between upward aerosol flux due to atmospheric turbulence and downward flux due to gravitational deposition. Monin-Obukhov similarity theory describes the turbulent profiles of wind, temperature and specific humidity. An approximate equilibrium expression relates particle size to ambient relative humidity.

To isolate the role of transport and growth, we first use the model to describe the surface layer concentration of inert particles unaffected by relative humidity. The calculations show that growth phenomena must be included to correctly describe aerosol concentrations in the presence of relative humidity gradients. Complete calculations of aerosols concentrations, including growth effects, show that the concentration of large particles (radius greater than 5 μm) can vary by an order of magnitude depending on stability. With light winds, the stability effect on large aerosols substantially affects the slope of the expected aerosol size distribution. With strong winds the aerosol distribution is not affected by changes in temperature and humidity lapse rates, but instead is governed by wind-shear-produced turbulent mixing.

1. Introduction

Atmospheric stability has been recognized as an influence on aerosol transport in continental situations. In modeling aerosol transport over the ocean, stability effects are usually neglected. Junge (1957) examined transport of ocean aerosol, assuming two different diffusion coefficients unrelated to bulk atmospheric parameters. Toba (1965) conducted an intensive examination of sea-salt aerosol transport, with the important assumption of neutral atmospheric stratification. The object of this work is to describe those atmospheric conditions where stability considerations affect vertical transport of sea-salt aerosols, and to evaluate the sensitivity of vertical profiles of aerosol size distribution within 100 m of the ocean surface to stability.

Atmospheric stability is determined both by wind speed and temperature and humidity lapse rate. With strong winds (>10 m s^{-1}) atmospheric stability is approximately neutral for the typical range of near surface lapse rates in the marine environment. With light winds, the stability is strongly affected by the temperature and humidity lapse rate. The following discussion mainly concerns low wind speed conditions, where thermal lapse rate is expected to be an important influence on vertical aerosol mixing. The light wind aerosol profiles are compared to the profiles in strong wind conditions.

We develop the general formulation of surface layer turbulent transport of aerosols in Section 2. This formulation is applied in Section 3 to the specific case of dry particles unaffected by humidity. In Section 4 we evaluate the general formulation for transport of moist, growing particles. We apply the theory to aerosol size distributions in Section 5.

2. Vertical profiles in a non-neutral atmosphere

The equilibrium distribution of aerosol particles in the surface boundary layer (depth 100 m)

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consists of a balance between the aerosol fallout flux and the turbulent upward flux, i.e.,

$$D\frac{dn}{dz} + wn = 0 \tag{1}$$

where D is the eddy diffusivity, n is the aerosol number density, and w is the aerosol fall velocity. The eddy diffusivity is

$$D = kU_* z/\phi(z/L) \tag{2}$$

where U_* is friction velocity; k, von Karman constant; z, height; L, Monin-Obukhov length; and ϕ, a semi-empirical stability function (Businger et al., 1971). We chose the stability function describing heat transport rather than momentum transport. The former function describes the flux of moisture and temperature, which affect the equilibrium size of the sea-salt aerosol.

The Monin-Obukhov length, L, describes the degree of stability of the air layer above the ocean surface. It is defined by

$$L = -\frac{U_*^3 T_v}{gk\overline{w' T_v'}}$$

where T_v is the mean virtual temperature of the atmospheric layer, g is acceleration due to gravity, and $\overline{w' T_v'}$ is the virtual heat flux in the layer. The Monin-Obukhov length can be evaluated from values of wind, humidity, air temperature and sea surface temperature by several means, e.g., the method of Barker and Baxter (1975).

The solution for the equilibrium aerosol distribution is

$$\frac{n}{n_0} = \exp\left[-\int_{z_0}^{z} \frac{w\phi(z'/L)}{U_* kz'}\,dz'\right] \tag{3}$$

where n_0 is the concentration of aerosols at the surface roughness height, z_0.

3. Transport of inert particles

The transport of aerosols in an environment where growth is not a factor follows from an approximate solution to (3) by assuming that the fall velocity of particles is independent of humidity variations and thus, height. Such a situation would occur, for example, in a very dry environment or in one with a uniform humidity profile. The solution is

$$\frac{n}{n_0} = \exp\left\{-\frac{w}{kU_*}\left[\ln\frac{z}{z_0} - \psi\left(\frac{z}{L}\right)\right]\right\} \tag{4}$$

where (Paulson, 1970)

$$\psi\left(\frac{z}{L}\right) = \int_{z_0/L}^{z/L} \frac{1-\phi(\zeta)}{\zeta}\,d\zeta \tag{5}$$

Equation (4) shows that the stability influence on transport of non-growing particles can be described by a linear correction to a logarithmic profile.

For neutral conditions, $\psi(z/L)$ is zero. Equation (4) can be manipulated to yield the following expression for the influence of stability on the ratio between non-neutral (n) and neutral (n_N) concentrations of inert particles at a given level z and a constant value of U_*

$$\ln\frac{n}{n_N} = \frac{w}{kU_*}\psi\left(\frac{z}{L}\right) \tag{6}$$

In Section 6, results using this relationship are presented. The next section discusses the situation of a growing aerosol.

4. Transport of growing aerosol

A realistic model of vertical aerosol profiles must include the influence of aerosol growth upon fall velocity. The fall velocity of small aerosol particles can be represented assuming viscous flow and using Stokes' formula,

$$w = \frac{2}{9}\frac{\rho r^2 g}{\eta} \tag{7}$$

where ρ is particle density, g is acceleration due to gravity, r is particle radius, and η is dynamic viscosity. A simple dependence of equilibrium sea-salt aerosol size on the relative humidity was presented by Kasten (1969),

$$r = r_d(1-f)^{-0.25}$$
$$f = \begin{cases} \mathrm{RH}/100, & \mathrm{RH} \le 99.5\% \\ 0.995, & \mathrm{RH} \ge 99.5\% \end{cases} \tag{8}$$

where r_d is the radius of the dry aerosol, and RH is relative humidity. The variation of growth rate is neglected and all particles are assumed to attain their equilibrium size instantaneously (Toba, 1965).

Representing the density of water by ρ_w, and the density of sea salt by ρ_d, the density of a sea-salt solution droplet of radius r is

$$\rho = \rho_w + (\rho_d - \rho_w)(1 - f)^{0.75} \tag{9}$$

The concentration of wet aerosols at height z, relative to their surface concentration, is found by substituting eqs. (7), (8) and (9) into eq. (3) with the result

$$\ln\left(\frac{n}{n_0}\right) = -\frac{2g\rho_w}{9\eta U_* k} r_d^2 \times \int_{z_0}^{z} \frac{\phi\left(\frac{z'}{L}\right)}{} dz' \times \left\{(1-f)^{-1/2} + \left(\frac{\rho_d}{\rho_w} - 1\right)(1-f)^{0.25}\right\} \tag{10}$$

The function in brackets in eq. (10) is the modification of the aerosol concentration by including growth. This factor varies in magnitude from 2.7 to 14.5 over the range of RH from 40% to 100%. The effect of this modification on $\ln n/n_0$ is equivalent to considering particles different in radius by up to a factor of 3. Clearly when the marine near-surface layer is characterized by a substantial humidity lapse rate, particle growth must be considered for a correct formulation of the aerosol concentration.

5. Stability and wind influences on aerosol distributions

Differential aerosol transport alters the aerosol size distribution for different height, wind, and stability conditions. A frequently used formula for a maritime aerosol distribution is the Junge distribution

$$n(r) = \frac{dN}{d(\log r)} = N_c\left(\frac{r}{r_c}\right)^{-\beta} \tag{11}$$

where $n(r)$ is number of aerosols (cm^{-3}) per unit log radius interval; N_c, r_c and β are constants. Generally the value of β is found to be near 3.

To study the influence of wind and stability on the aerosol size distribution, we now make a simplifying assumption. We assume that the aerosol distribution at the surface follows eq. (11) with $\beta = 3$. Prior to this, we have only been concerned with the relative height distribution, $n(z)/n_0$ of aerosols, and not with the aerosol size distribution. We now specify the form of the surface aerosol distribution, n_0, as the Junge distribution, eq. (11). We can now compute from eq. (3) what the form of the distribution must be at various heights for a given stability and wind. The general expression for the aerosol distribution is

$$n = \left(\frac{n(z)}{n_0}\right) N_c\left(\frac{r}{r_c}\right)^{-\beta} \tag{12}$$

From eq. (3) it is clear that the relative number distribution, n/n_0, of a given sized particle is not solely a function of stability, z/L. The wind, through U_*, plays a major role. At higher wind speeds, the marine surface layer generally is near neutral ($z/L \sim 0$) stratification and n/n_0 is governed by mechanical, wind-shear produced turbulence. Under these conditions, the vertical transport descriptions of Junge (1963) and Toba (1965) apply. At lower wind speeds, the influence of the stability function, $\psi(z/L)$, becomes more evident.

Under the conditions of strong turbulent mixing, we expect the size distribution to change little with height. Under light wind, stable conditions, however, we expect the distribution to change substantially with height. With these weak mixing conditions, increasing numbers of the large particles fallout relative to the small particle concentration. Results from such numerical experiments are discussed in the next section.

6. Results

Fig. 1 shows the relation between the scaled height, z/L, and the ratio of particle concentration in non-neutral stability to particle concentration at neutral stability. The results in this figure assume inert particles, $z_0 = 1 \times 10^{-4}$ m, and $U_* = 0.15$ m s^{-1} (corresponding to a wind speed at 10 m of 5 m s^{-1} under neutral conditions). Extension to other values of U_* is straightforward. In an unstable boundary layer ($z/L < 0$), the aerosol number density at level z is enhanced over its value at neutral stability. The relative enhancement is greater for the larger particles since smaller particles are already quite well mixed at neutral

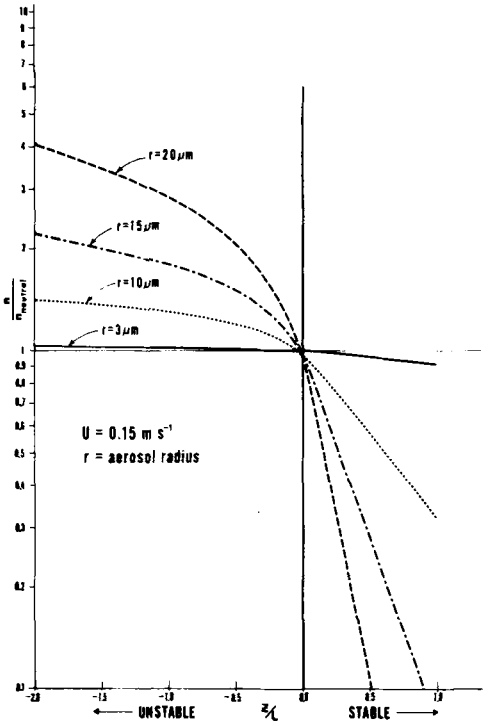

Fig. 1. Ratio of neutral to non-neutral aerosol concentrations as a function of stability-scaled height (z/L) for various inert aerosol particle sizes. This is evaluated for $U_* = 0.15$ m s^{-1} corresponding to a 10 m wind of 5 m s^{-1} in neutral conditions.

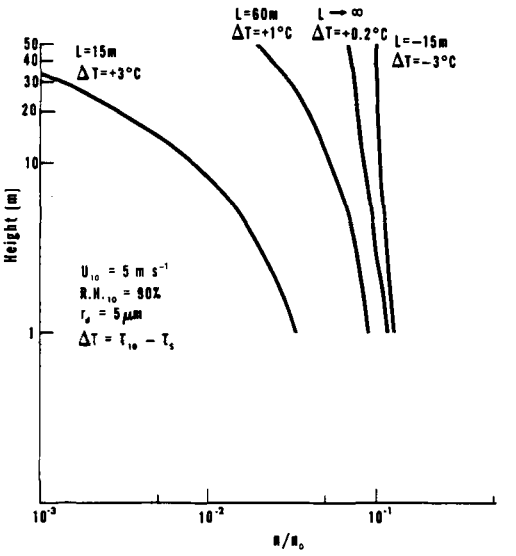

Fig. 2. Vertical concentration profiles of 5 μm aerosols at different stabilities. Each curve is identified by an air–sea temperature difference ΔT, and Monin-Obukhov length L. The relative humidity, RH, and 10 m wind speed, U_{10} are indicated.

stability. In the stable boundary layer ($z/L > 0$), the aerosol number density is less than its neutral value, with the relative decrease in aerosol content being more pronounced for the larger particles. This results from the reduced upward turbulent transport being less able to balance the fallout of the larger particles.

The next case includes aerosol growth with results calculated by numerical integration of (10). Relative aerosol concentrations are shown in Fig. 2 for a wind speed of 5 m s^{-1} and a relative humidity of 90%, both evaluated at a height of 10 m. Aerosols having a dry radius, r_d, of 5 μm are considered in this case. It is clear that particle concentration is strongly dependent on thermal stability. The difference between the stable and neutral concentrations can be greater than an order of magnitude at a height of 20 m.

The effects of boundary layer stability on a Junge aerosol size distribution are calculated using eq. (12). The aerosol size distribution is evaluated for a high and a low wind speed, 15 m s^{-1} and 3 m s^{-1}, and for a high and low relative humidity, 95% and 75%. For each case, the size distribution is calculated for an air–sea temperature difference of −2 °C, 0 °C and +2 °C, corresponding to an unstable, neutral, and stable thermal lapse rate. Although much stronger near-surface lapses occur overland, these values are rather typical for the marine surface layer. As we anticipated, at high wind speeds the thermal stability plays a very minor role in determining the size distribution. Fig. 3a shows the size distributions at a height of 20 m for these three different lapse rates. It is clear that mechanical mixing is dominant, with the stable and unstable distributions being nearly identical. A change in relative humidity (Fig. 3d) at this wind speed makes negligible difference.

At lower wind speeds, the influence of thermal lapse and humidity becomes evident. Figs. 3b and 3c show a pronounced change in the size distribution with stability, particularly for stable stratification. This difference in size distribution is enhanced by changes in relative humidity, since the latter change equilibrium aerosol size. Figs. 3b and 3c show the change in size distribution induced by a change in relative humidity from 75% to 95%.

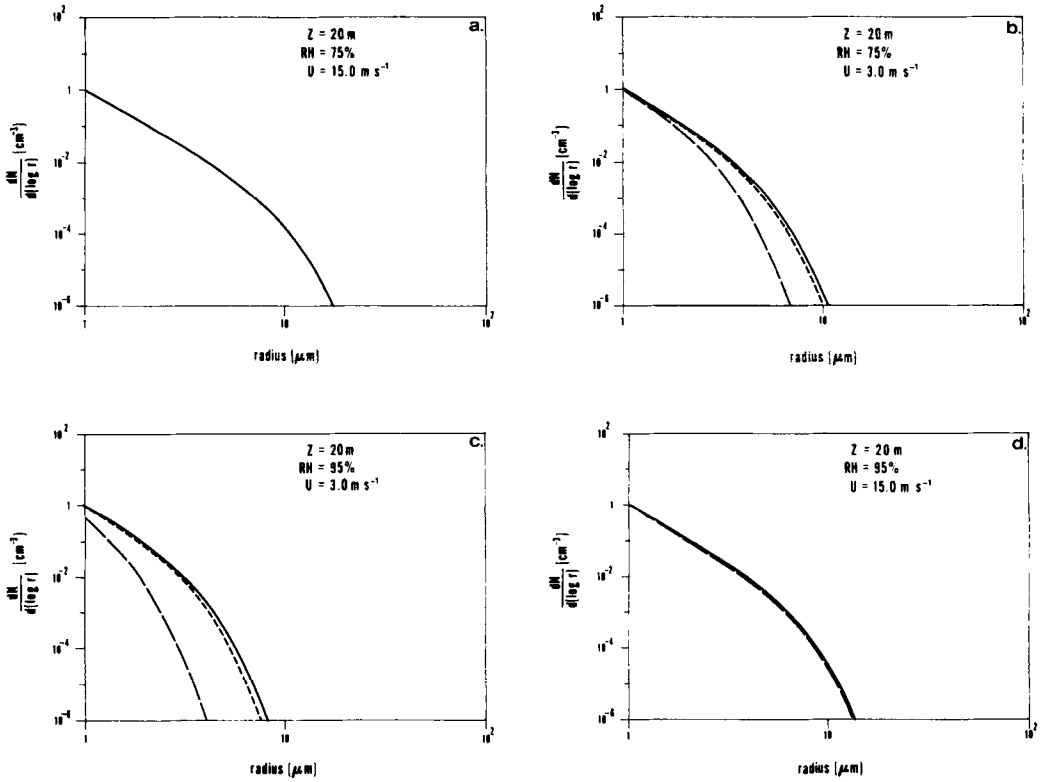

Fig. 3. Stability effects on a Junge aerosol size distribution. Solid curve is for air-sea temperature difference $\Delta T = -2\,°C$ (unstable), short dash is $\Delta T = 0$ (neutral), and long dash is $\Delta T = +2\,°C$ (stable). The curves indicate that stability and relative humidity have a large effect on aerosol concentration in light winds. Note that in 3a and 3d, the high wind cases, the curves for different stabilities overlap.

7. Conclusions

The transport of aerosol particles has been treated by a simple model of gravitational settling balanced by upward turbulent transport. The influence of thermal lapse upon the number density of measured aerosol is important, particularly in stable vertical profiles. The concentration of aerosols at any given height can vary by an order of magnitude because of stability.

Wind speed and stability are found to exert strong influences upon the aerosol size distribution. At high wind speeds, the turbulent mixing governing the particle distribution is generated primarily by wind shear. Thus, at high wind speeds, the distribution is unchanged in the range of typical lapse rates of humidity and temperature found in the marine surface layer. At lower wind speeds, however, these lapse rates play a significant role in altering the character of the distribution. Relative humidity affects aerosol concentration because equilibrium particle size depends on the ambient relative humidity. Fallout flux, being dependent on particle size, increases as relative humidity increases. Hence, in the surface layer, the concentration of large particles decreases with increasing humidity.

It should be understood that this model does not include certain other effects which may be dominant under specific atmospheric conditions. For example, the only source of aerosols is at the lower (sea-surface) boundary. Other sources, such as evaporated cloud drops, can prevail over surface generation under conditions of low wind. These and other problems will be addressed in future work.

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ВЛИЯНИЕ УСТОЙЧИВОСТИ НА РАСПРЕДЕЛЕНИЕ АЭРОЗОЛЯ ПО РАЗМЕРАМ И С ВЫСОТОЙ

Обычное описание вертикального распределения концентраций аэрозоля подразумевает нейтральность стратификации атмосферы. В работе исследуется влияние атмосферной устойчивости на вертикальный профиль аэрозольной концентрации. Построена модель для описания переноса аэрозоля в приземном слое атмосферы. Модель списывает баланс между потоком аэрозоля вверх из-за атмосферной турбулентности и его потокам вниз из-за гравитационного оседания. Теория подобия Монина-Обухова описывает турбулентные профили ветра, температуры и удельной влажности. Приближенное соотношение связывает в равновесии размер частиц с относительной влажностью окружающего воздуха.

Чтобы изолировать роль переноса и роста, мы сначала используем модель для описания в приземном слое концентрации инертных частиц, не реагирующих с относительной влажностью. Вычисления показывают, что явления роста должны быть включены для правильного описания концентрации аэрозоля в присутствии градиентов относительной влажности. Полные расчеты аэрозольной концентрации, включающие эффекты роста, показывают, что концентрации больших частиц (с радиусом больше 5 мкм) могут меняться на порядок в зависимости от устойчивости. При небольших ветрах влияние устойчивости существенно определяет наклон ожидаемого распределения аэрозольных частиц по размерам. При сильных ветрах распределение аэрозоля не зависит от изменений в вертикальных градиентах температуры и влажности, но определяется турбулентным перемешиванием, порождаемым сдвигом ветра по высоте.