

Size spectra and aggregation of suspended particles in the deep ocean

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Abstract—Work of the last 10 years has demonstrated that oceanic particle size distribution by volume tends to be flat at mid-water depths (equivalent to a cumulative particle number distribution with a slope of -3) and is peaked in nepheloid layers with active resuspension and in surface waters with active biological production. The observed loss of fine peaks from the suspensions to yield flat distributions requires aggregation of the material, as the fines settle slowly. Mechanisms leading to particle collision are examined, for interactions between particles of similar size. Brownian motion dominates below 1.5 to $8\ \mu\text{m}$. However, if large particles (such as 'marine snow') are present at realistic concentrations, they become important in the removal of fine particles by shear-controlled coagulation. The coagulation times calculated for shear are too long for steady state to be presumed while the size distributions evolve under the influence of coagulation mechanisms. Therefore suggestions that the flat size distributions are quasi-stationary results of shear-controlled coagulation are rejected, and the notion that there is sub-equal production of particles at different points in the spectrum is favoured. Such production and the subsequent scavenging of small particles by large settling ones confers great importance on components of biological origin in both providing elements of the total size spectrum and determining the distribution and sedimentation of others of lithogenic origin. In surface waters, filtration rates by zooplankton indicate that aggregation rates of particles above submicron sizes are biologically determined.

INTRODUCTION

THE removal of particles from surface and mid-water depths in the ocean normally requires their aggregation into larger units having a settling speed greatly in excess of the original particles. The argument has been presented and corroborated by several authors including REX and GOLDBERG (1958), SCHRADER (1971), McCAVE (1975), HONJO (1976), and in papers based on sediment-trap data. The size distribution of suspended particles is a function of several variables including source and nature of the particles, physical or biological processes of aggregation and 'age' of the suspension. In this paper the physical processes controlling formation of aggregates are examined, their rates are compared, and their relative importance evaluated. A large body of theory and experience concerning particle interactions has been developed by aerosol physicists and by chemical engineers treating aqueous suspension. The work has been made easily accessible in books by FRIEDLANDER (1977), TWOMEY (1977), and PRUPPACHER and KLETT (1978), and has been presented by LERMAN (1979) for natural waters. Another purpose of this paper is to examine the implications of the aerosol and experimental work for the hydrosols of the deep sea.

There are few measurements of particle size spectra from the ocean. The principal sources give Coulter Counter® data (BRUN COTTAN, 1971, SHELDON *et al.*, 1972; McCAVE,

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1975, LERMAN *et al.*, 1977; PAK *et al.*, 1980, RICHARDSON, 1980; McCAVE, 1983) Counting and sizing particles by microscopy is tedious and few data have been obtained in that way (WELLERSHAUS *et al.*, 1973; HARRIS, 1977, BAKER *et al.*, 1979, LAMBERT *et al.*, 1981). Wellershaus *et al.* used light microscopy while Harris used transmission and the others scanning electron microscopy to examine the particles. The particle number data can, in nearly all cases, be fitted by a power-law distribution over a large part of the measured range. Expressed as cumulative number N as a function of particle diameter d this is $N = k d^{-\beta}$. A value of $\beta = 3$ signifies equal particle volumes in logarithmically increasing size grades. This was found by Brun-Cottan, Sheldon, McCave, Lerman *et al.*, and Pak *et al.* (cited above) in regions well away from the bed or, if close to the bed (Lerman *et al.*), regions of very low concentrations (Fig 1). In more concentrated nepheloid layers near the bed the sizes show a

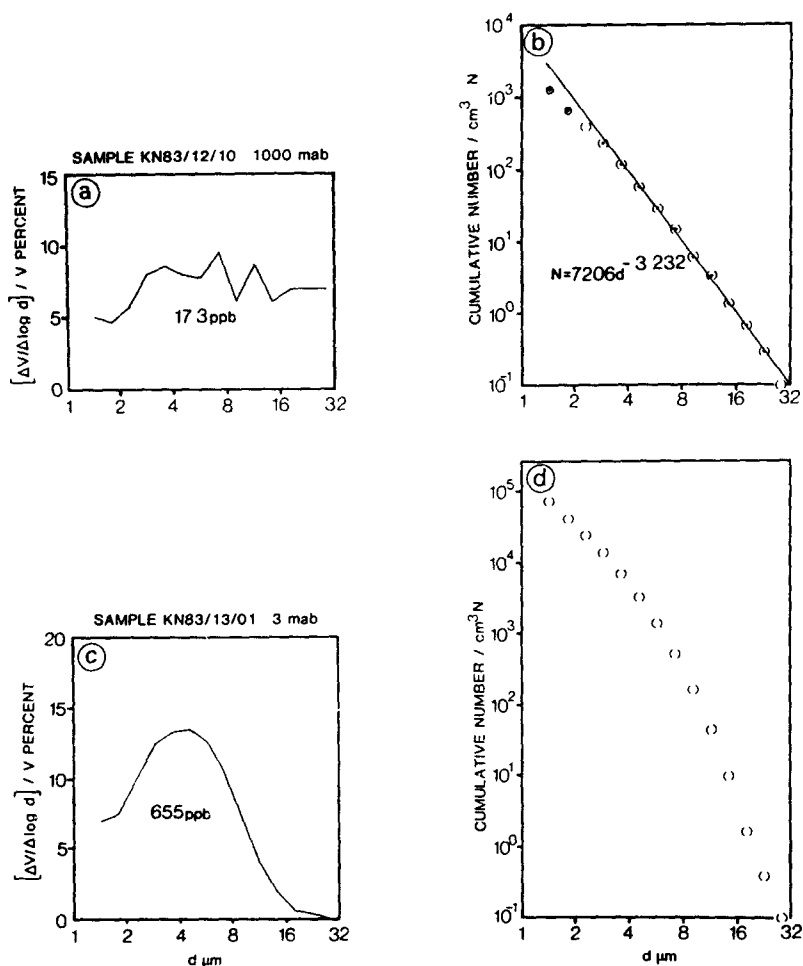


Fig 1 Examples of size distributions that are relatively flat in volume (a) with the equivalent number distribution showing a slope close to -3 (b) [sample obtained from 3900 m at ~ 1000 m above the bed over the Nova Scotian Continental Rise (Sta 12 of McCAVE, 1983)] and (c) peaked from a nepheloid layer with a number distribution (d) that is not a straight line. Size analysis by Coulter Counter[®] with 70- μ m aperture

peak around 3 to 4 μm and the number distribution cannot be fitted by a straight line (RICHARDSON, 1980; MCCAVE, 1983). LAMBERT *et al* (1981) also maintained that the distributions of individual components (e.g., aggregates, goethite, aluminosilicate) they observed are not of power-law type but are log-normal by number. They found a distinct fall-off at the finest end of the distribution whereas HARRIS (1977) counted the finest particles down to 0.1 μm and showed a power-law distribution with $\beta = 1.62$ in the region $d < 1 \mu\text{m}$. WELLERSHAUS *et al* (1973) also found a decrease in numbers of fine particle but said that it is an artifact of the counting method. The sum total of the components shown by LAMBERT *et al* (1981, Fig. 1) however, seems to be fitted by a straight line over a good part of the measured range with $\beta \sim 3$. The only results from the Pacific (BAKER *et al.*, 1979) also are fitted by the higher value of $\beta = 3.29$ for $3 < d < 8 \mu\text{m}$ from mid-water samples.

In summary, most workers show data implying $\beta \sim 3$ in the size range 1 to 100 μm in clear water. There is no consensus as to what is the distribution of submicron particles. Relatively young (i.e., freshly eroded) suspensions in nepheloid layers have peaked volume size distributions, but old ones are flat (i.e., $\beta \sim 3$) according to MCCAVE (1983). And there is one view that the overall distribution, whatever it may be, is made up of log-normal number distributions of several different components. The flat $\beta = 3$ distribution may extend to sizes well beyond 100 μm shown by SHELDON *et al* (1972).

The recent surge of interest in rapidly settling particles coupled with the demonstration that the first choice, faecal pellets, could only account for a small fraction of the measured vertical flux (HONJO, 1980), has prompted a re-examination of 'marine snow' as described by the Japanese in the early 1950s (SUZUKI and KATO, 1953; NISHIZAWA *et al.*, 1954; SHANKS and TRENT, 1980). The effectiveness of such aggregates in collecting particles as they settle and the mechanisms for production of smaller flocs in the ocean are still unclear, and it is intended that this paper will set out some of the physical principles involved.

PARTICLE DENSITY AND SETTLING VELOCITY

For model calculations some assumptions about density are necessary to obtain settling velocity. It is clear that density decreases with increasing aggregate size and there is a wide spread of values for any given size, partly because of the diversity of particle types. Earlier data were summarized by MCCAVE (1975) and a simple mean law was assumed. More recently there have been measurements of settling velocity by CHASE (1979), TAMBO and WATANABE (1979), and KAWANA and TANIMOTO (1979) [subsequently re-analysed by HAWLEY (1982)], all of which suggest (on incomplete data) that for Stokes' settling, the densities given earlier are too low. Also, from comparison of the particle density data of LAGVANKAR and GEMMELL (1968) with the settling data of Tambo and Watanabe, it appears that either the densities are low or that the particles settle faster than Stokes' Law would allow, a central point in Chase's arguments. To avoid problems here I shall follow the suggestions of Tambo and Watanabe. They showed fall-off of density for clay-iron and clay-magnesium flocs to be as $d_p^{-1.1}$ approximately and for biological flocs $d_p^{-1.65}$. Here I assume $d_p^{-1.3}$ dropping to a density contrast $\Delta\rho = 0.003 \text{ g cm}^{-3}$ at $d_p = 0.12 \text{ cm}$ derived from Tambo and Watanabe's data but assuming particle sphericity of unity.

The value 0.003 is assumed constant for larger (non-Stokesian because $Re > 1$) particles. With a fall-off of $d_p^{-1.3}$ there must be a change to a lesser slope of $\Delta\rho$ vs d_p at some size $< 100 \mu\text{m}$ as surmised by Tambo and Watanabe. I shall assume a break at 50 μm (Fig. 2) and an origin at $\Delta\rho = 1 \text{ g cm}^{-3}$ at $d_p = 1 \mu\text{m}$. This is denser than assumed by MCCAVE (1975), but

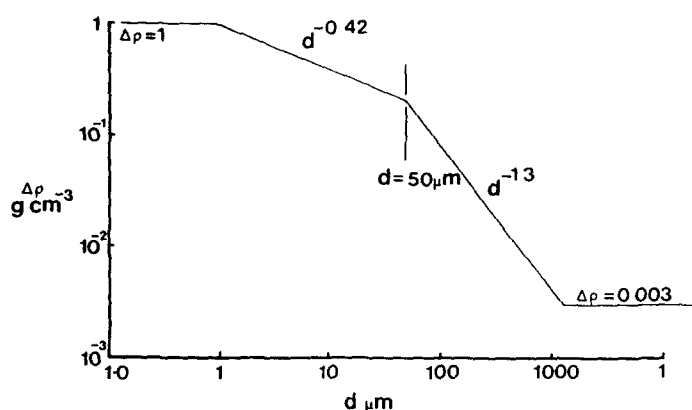


Fig 2 Model particle density contrast as a function of size for aggregates assumed here After TAMBO and WATANABE (1979)

Table 1 Model particle parameters*

d (μm)	$\Delta\rho^\dagger$ (g cm^{-3})	w_s (cm s^{-1})	Re^\ddagger
0.1	1	3.63×10^{-7}	2.54×10^{-10}
0.2	1	1.45×10^{-6}	2.03×10^{-9}
0.5	1	9.08×10^{-6}	3.18×10^{-8}
1	1	3.63×10^{-5}	2.54×10^{-7}
2	0.746	1.08×10^{-4}	1.51×10^{-6}
5	0.506	4.60×10^{-4}	1.61×10^{-5}
10	0.378	1.37×10^{-3}	9.59×10^{-5}
20	0.282	4.09×10^{-3}	5.73×10^{-4}
50	0.191	0.0174	6.09×10^{-3}
100	0.0776	0.0282	0.0197
200	0.0315	0.0458	0.0641
500	9.58×10^{-3}	0.0870	0.305
1000	3.89×10^{-3}	0.1410	0.98
2000	3×10^{-3}	0.1447	2.03
5000	3×10^{-3}	0.1563	5.47
10^4	3×10^{-3}	0.1736	12.2

* Compared with Table 2 of McCAVE (1975) w_s is greater

$\dagger \Delta\rho = (\rho_s - \rho)$, where ρ_s is particle *in situ* bulk density and ρ is fluid density (here taken as 1.05 g cm^{-3})

$\ddagger Re = w_s d / \nu$ with $\nu = 0.0143$ Stokes

recent investigations of nepheloid layers (McCAVE, 1983) show higher apparent densities at high concentrations, where there is presumably less low-density organic matter and less aggregation. Below $d_p = 1 \mu\text{m}$ a constant $\Delta\rho$ is assumed (Table 1, Fig 1)

THEORY OF PARTICLE INTERACTIONS

Introduction

Although the interaction of particles must play an important role in controlling oceanic particle size spectra, there has been little work on the subject. There has, however, been

extensive work on atmospheric aerosols and, as most of the processes are similar and the equations are presented in non-dimensional form, substantial use has been made of this body of experience in the present treatment (TWOMEY, 1977, FRIEDLANDER, 1977, PRUPPACHER and KLETT, 1978). SHOLKOVITZ (1976) and EDZWALD *et al* (1974) substantiated KRONE's (1962, 1972, 1978) contention that flocculation is important in estuaries and thus there is every reason to believe particle aggregation occurs in the deep ocean, albeit more slowly because of the lower concentration. Krone showed that under estuarine conditions Brownian motion controlled aggregation of particles $<1 \mu\text{m}$ in diameter, and shear controlled aggregation of larger ones. The formation of aggregates involving both organic and inorganic materials has been experimentally demonstrated by KRANCK and MILLIGAN (1979). Within the water column and on the sea floor there are several types of organic-inorganic aggregates including those known as 'marine snow' and faecal pellets. Some such organic aggregates result from an organism actively gathering and consolidating sediment and organic matter (e.g., faecal pellets), whereas others result from the passive collision of particles, whether organic or inorganic. In the latter case an organic component may simply provide a sticky substrate for particle accumulation. It may increase the coalescence efficiency (i.e., the fraction of collisions that result in particle sticking) but the organic matter does not play any active role in increasing the frequency of collisions. The efficiency is often assumed to be ~ 1 in atmospheric aerosol work because it is difficult to obtain independently and no great discrepancy between calculation and measurement is caused by the assumption (TWOMEY, 1977, p. 126). However, SWIFT and FRIEDLANDER (1964) determined efficiency of ~ 0.37 for both Brownian and shear coagulation of polystyrene latex in salt solution, and EDZWALD *et al* (1974) showed the efficiency to be only about 0.1 for natural sediment from the seaward end of an estuary in artificial seawater. Thus there is room for improvement of coalescence efficiency by sticky organic coatings (mucus). Nevertheless, work on estuarine and coastal suspensions has shown that particles tend to be electrically uniform with the same electrophoretic mobility (HUNTER and LISS, 1982), a property most likely conferred by particle coatings of dissolved organic matter (NIEHOFF and LOEB, 1972, 1974, LOEB and NIEHOFF, 1977, HUNTER, 1980). The surface similarity of particles should decrease the chance of their coalescence because of mutual repulsion and could explain the smaller efficiency reported by EDZWALD *et al* (1974) than that of SWIFT and FRIEDLANDER (1964). Still, the principal control on aggregation is the frequency with which particles come into contact, which is controlled by physical mechanisms outlined in a later section.

The size distribution function

The continuous size distribution function $n(v)$ is defined by $dn = n(v)dv$, where dn is the number of particles of volume between v and $v + dv$ per unit water volume. The related size distribution function $n(d_p)$ is defined by $dn = n(d_p) d(d_p)$, where dn is the number of particles per unit volume of suspension in the size (diameter) interval d_p to $[d_p + d(d_p)]$. Because particulate material has many more small particles than large ones a distribution of the form $dn = n(x) d(x)$ is generally used where $x = \log d$, so $n(\log d_p) = dn/d(\log d_p)$. Because $d(\log d_p) = d_p^{-1} d(d_p)$ the distribution functions are related by

$$n(\log d_p) = \frac{dn}{d(\log d_p)} = d_p \frac{dn}{d(d_p)} \quad (1)$$

Data are often acquired in such a way that the cumulative distribution, the number of particles N larger than a given size, is commonly used for presentation, $\log N$ being plotted vs $\log d_p$, where

$$N = \int_{d_p}^{\infty} n(d_p) d(d_p) = \int_{\log d_p}^{\infty} n(\log d_p) d(\log d_p)$$

for a given size d_p (TWOMEY, 1977, pp 4-7). From this it can be seen that

$$\frac{dN}{d(\log d_p)} = n(\log d_p) = \frac{dn}{d(\log d_p)} \quad (2)$$

and

$$\frac{dN}{d(d_p)} = n(d_p) = \frac{dn}{d(d_p)}$$

The slope of the cumulative number distribution is often a straight line of slope $d(\log N)/d(\log d_p) = -\beta$, where $\log N = -\beta \log d_p + \log k$. Also for such a distribution the slope of $dN/d(\log d_p)$ vs d_p on log-log axes is $-\beta$, and the slope of $dn/d(d_p)$ vs d_p commonly plotted in aerosol work and by LERMAN *et al.* (1977), BAKER *et al.* (1979), and PAK *et al.* (1980) in the oceans is $-(\beta + 1)$ (BADER, 1970)

Plots of particle volume as $dv/d(\log d_p)$ vs $\log d_p$ are also commonly used and are employed here in a discrete form normalized by total particle volume, $[\Delta v/\Delta(\log d_p)]/V$, where the size interval $\Delta(\log d_p)$ used is $\Delta \log_{10} d = 0.1003$. In this scheme the size classes are in a volume doubling sequence, i.e., grain size doubles every third class. This is the built-in scheme of the Coulter Counter® with a $2^{1/3}$ size sequence, or a one-third phi interval scale in geological parlance (SHELDON, 1969). The advantage of using the volume distribution function $n(v)$ is that volume is conserved when solid particles collide, while the distribution function using number $n(d_p)$ is usually plotted.

The general dynamical equation

FRIEDLANDER (1977, chap. 7) and JEFFREY (1982) presented the general dynamical equation or population balance equation for particles in flow systems. A two-dimensional time-averaged form of the equation appropriate to the case of particles in mid-water oceanic depths is

$$\frac{\partial \tilde{n}}{\partial t} + \bar{U} \frac{\partial \tilde{n}}{\partial x} = \frac{\partial}{\partial x} \left[(D + \varepsilon_x) \frac{\partial \tilde{n}}{\partial x} \right] + \frac{\partial}{\partial z} \left[(D + \varepsilon_z) \frac{\partial \tilde{n}}{\partial z} \right] - \frac{\tilde{n} \partial w_s(v)}{\partial z} + w_s(v) \frac{\partial \tilde{n}}{\partial z} \quad (i) \quad (ii) \quad (iii) \quad (iv)$$

$$+ \frac{1}{2} \int_0^{v_j} K(v_i, v_j - v_i) n(v_i) n(v_j - v_i) dv_i - \int_0^{\infty} K(v_j, v_i) n(v_j) n(v_i) dv_i \quad (3)$$

$$(v) \quad (vi)$$

$$+ \int_0^{\infty} J(v_j + v_i, v_j, v_i) n(v_j + v_i) dv_i - \int_0^{v_j} J(v_j; v_i, v_j - v_i) n(v_i) dv_i,$$

$$(vii) \quad (viii)$$

where \tilde{n} is the volume distribution function $n(v)dv$. The terms are (i) non-steady, (ii) horizontal advection, (iii) diffusion including both Brownian diffusion with the coefficient $D = kT/3\pi\mu d$, where k is the Boltzmann constant, T the absolute temperature, μ dynamic viscosity, d particle diameter, plus turbulent diffusivities ϵ_x , ϵ_z , (iv) settling of particles with settling velocity distribution $w_s(v)$, (v) increase of particle numbers in certain sizes (v_j) due to coagulation of smaller ones v_i and $(v_j - v_i)$, (vi) loss of particles of size v_j by coagulation, (vii) gain of v_j sized particles from breakup of larger ones and (viii) loss of v_j sized particles by breakup to give smaller ones. In (v) and (vi) the terms $K(v_i, v_j - v_i)$ and $K(v_j, v_i)$ are the coagulation kernels. The probability that a particle of volume v_j will encounter a particle of volume v_i is proportional to the number of v_i particles and is given by $K(v_j, v_i) n(v_i) dv_i$ (TWOMEY, 1977, p. 123). The analogous J terms for breakup are such that

$$J(v_j, v_i, v_j - v_i) n(v_j) dv_j dv_i dt$$

gives the number of flocs of size v_j that break into flocs of sizes v_i and $(v_j - v_i)$ in time dt and are due to JEFFREY (1982).

There are several other terms that could be added to the equation, namely deposition sink and source terms. The sink of deposition on the bed can be expressed as

$$-\tilde{n}_\delta w_s(v) [1 - \tau_0/\tau_i(v)],$$

where \tilde{n}_δ is the volume distribution function of particles within the viscous sublayer of thickness δ , and $\tau_i(v)$ is the distribution of limiting shear stress for deposition as a function of particle volume (EINSTEIN and KRONE, 1962, McCAYE and SWIFT, 1976). Large particles of high settling velocity that might be deposited rapidly would, if broken up into pieces, have a slower rate of deposition. Finally, near the bed there is a possible source from erosion. Little is known about this source in terms of numbers of particles produced and their size distribution.

Coagulation mechanisms

As was pointed out above, coagulation mechanisms are ways of bringing particles together. They are discussed in greater detail by PRUPPACHER and KLETT (1978, p. 304) and in other aerosol texts. The mechanisms are however physically general and have been verified for aqueous suspensions by SWIFT and FRIEDLANDER (1964), BIRKNER and MORGAN (1968), HAHN and STUMM (1970), DELICHATSIOS and PROBSTEN (1975), and HUNT (1982a, b). Five mechanisms are discussed below and their relative effectiveness is assessed.

Brownian motion. Spherical particles undergoing Brownian diffusion have a probability of colliding given by the kernel K_{Bij} times the number of particles. The kernel with dimensions $L^3 T^{-1}$ is

$$K_{Bij} = 2\pi D_{ij} d_{ij} = \frac{2kT}{3\mu} \frac{d_i^2}{d_i d_j}, \quad (4)$$

where $D_{ij} = (D_i + D_j)$, the sum of the diffusion coefficients ($D_i = kT/3\pi\mu d_i$), and $d_{ij} = (d_i + d_j)$, the sum of the diameters of particles of size d_i and d_j . If all the particles are the same size at the beginning of coagulation (a monodisperse suspension) then the coagulation time t_c , the time to reduce the initial number N_0 of particles per unit volume by a half, is $t_c = 3\mu/4kTN_0E$. Using appropriate values for deep-ocean nepheloid layers, $\mu = 0.015$ poise, $T = 275^\circ\text{K}$, $N_0 = 4 \cdot 10^5$ particles cm^{-3} in the size range 0.5 to 1 μm , then $t_c \sim 8.6$ days assuming efficiency (E) of unity. With more realistic efficiency $E = 0.10$ from EDZWALD *et al.* (1974) $t_c \sim 3$ months, which could be reduced to about 10 days for the concentrated suspension.

reported by BISCAYE *et al* (1980). Thus we expect appreciable Brownian pumping of particles from submicron size into aggregates a few μm in diameter on time scales of a week to a few months to occur initially after production of a nepheloid layer by erosion of the bed. However, well away from the boundaries where the number of small particles is about 100 times less, the coagulation time is ~ 20 years. Thus at mid-water depths, Brownian coagulation of small particles is very slow.

Laminar and turbulent shear. The laminar and turbulent shear kernels are similar:

$$K_{LSy} = \Gamma \frac{d_y^3}{6} \quad (5)$$

for laminar shear with $\Gamma = d\bar{U}/dz$ and

$$K_{TSy} = 0.163 d_y^3 (\epsilon/\nu)^{1/2} \quad (6)$$

for turbulent shear with the shear rate Γ given in terms of the turbulent dissipation rate ϵ and kinematic viscosity ν (SAFFMAN and TURNER, 1956). The relationship has been verified for aqueous suspensions by DELICHATSIOS and PROBSTEN (1975) though they had the constant as 0.105. The coagulation time corresponding to equation 6 for a monodisperse suspension ($d_i = d_j$) is $t_c = 0.693 \pi/4VT$, where $V = \pi d^3 N_0/6$, the total solid particle volume conserved in the collisions. In the deep sea, turbulence levels are generally low, and I shall assume values no higher than a dissipation rate of $10^{-4} \text{ cm}^2 \text{ s}^{-3}$ in the bottom mixed layer (NEWBERGER and CALDWELL, 1981) and $10^{-6} \text{ cm}^2 \text{ s}^{-3}$ in clear mid-ocean depths (OAKLEY, personal communication). Higher values may occur in surface waters (10^{-3} to $10^{-2} \text{ cm}^2 \text{ s}^{-3}$). Using temperature and viscosity as above with a water density of $\sim 1.05 \text{ g cm}^{-3}$ gives kinematic viscosity $\nu = \mu/\rho$ of ~ 0.0143 stokes. The implied shear rates $\Gamma = (\epsilon/\nu)^{1/2}$ are 0.084 and 0.0084 s^{-1} . These values characteristic of the deep ocean will be used from here on. The relative importance of Brownian and shear coagulation is approximately given by K_{By}/K_{LSy} or $2kT/\mu d^3 \Gamma$, and so for equal importance with $\Gamma = 0.0084 \text{ s}^{-1}$ in a mono-disperse suspension $d \sim 8 \mu\text{m}$, but with $\Gamma = 1.0 \text{ s}^{-1}$ ($\epsilon = 1.4 \times 10^{-2} \text{ cm}^2 \text{ s}^{-3}$), $d = 1.7 \mu\text{m}$. So Brownian coagulation dominates over shear in suspensions with low turbulence intensity for particles of diameter $< 8 \mu\text{m}$, but in more turbulent suspensions shear dominates down to $1.7 \mu\text{m}$. With variable stresses the grain size region 1.5 to $8 \mu\text{m}$ is thus a transition region from Brownian to shear-dominated behaviour of particles relative to their near neighbours in size.

In a concentrated nepheloid layer the size range 4 to $8 \mu\text{m}$ ($\bar{d} = 5.65 \mu\text{m}$) has less particles than the range $< 1 \mu\text{m}$, and we shall take the value $N_0 = 3 \times 10^3$ (McCAVE, 1983) giving $V = 2.83 \times 10^{-7} \text{ cm}^3$ and thus $t_c = 222$ days with $\Gamma = 1 \text{ s}^{-1}$ and efficiency 0.1 . This could be shortened by higher shear (but sustained high shear is not a feature of the abyssal environment), and lengthened to 7 years for $\Gamma = 0.084 \text{ s}^{-1}$. In low concentrations at mid-depth, N_0 is 100 times less and this suggests coagulation times of ~ 7000 years if the efficiency is 0.1 and $\Gamma = 0.0084 \text{ s}^{-1}$. Because of the term d_y^3 in the shear coagulation kernel polydisperse suspensions (more than one particle size) coagulate more rapidly than the examples given here. Where there are a few large particles they may grow rapidly because of their large value of d_y^3 when combined with any other particle. Such cases need to be examined by numerical solutions such as those of JUNGE (1957, 1963), JUNGE and ABEL (1965, as reported in PRUPPACHER and KLETT, 1978, Fig. 12-2), and WALTER (1973). Nevertheless it appears that Brownian coagulation will create aggregates of a few μm diameter faster than shear coagulation will remove them to larger sizes, at least during the initial stages of aggregation. It must be noted that, as will be seen later, coagulation of similar sized particles in the size range 5 to $50 \mu\text{m}$ is

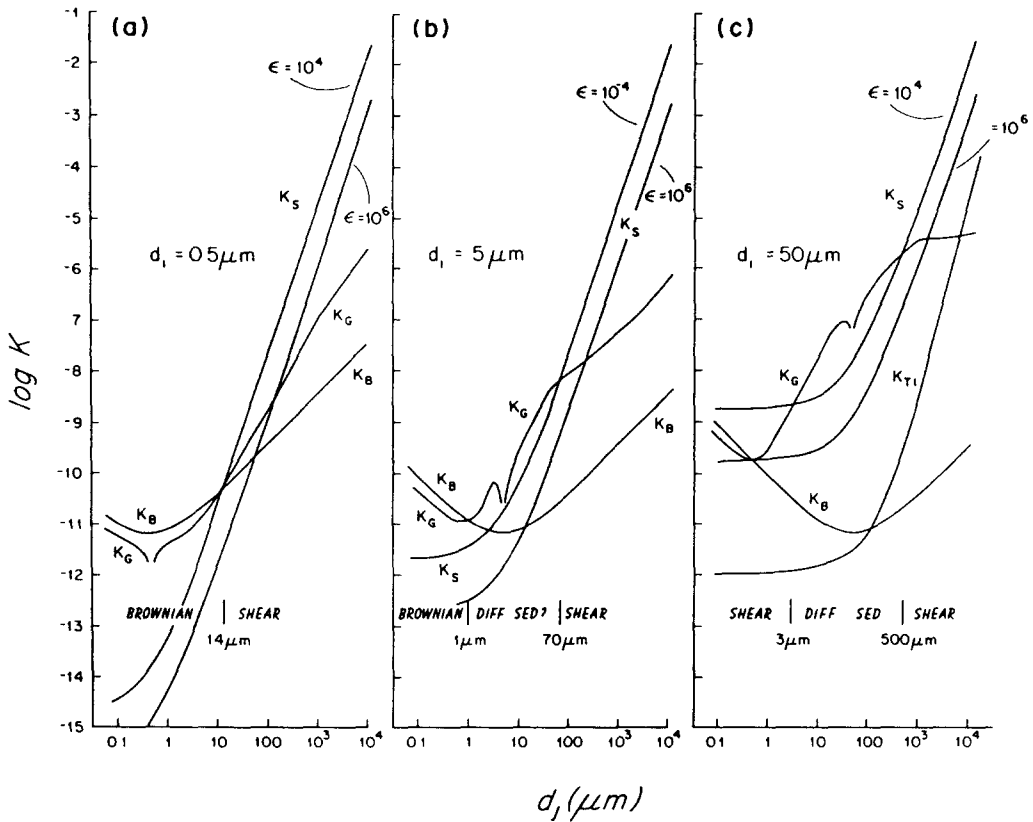


Fig 3 Coagulation kernels K for collection of 0.5, 5, and 50- μm particles plotted for oceanic conditions ($\rho = 1.05$, $\mu = 0.015$, $\epsilon = 10^{-4}$ and 10^{-6} cgs units, $T = 275^\circ\text{K}$) in the manner of FRIEDLANDER (1965). Subscripts are B for Brownian, G for differential settling (gravitational), and S for shear.

not dominated by shear but by differential sedimentation with a kernel about four times as large (Fig. 3). Even with this value, however, the rate at which particles are pumped into the size range 1 to 5 μm by Brownian aggregation is greater than their removal up the size spectrum. Subsequently, as the number of submicron particles becomes relatively depleted, the 'bump' in the size distribution at values of a few μm will be dissipated by transfer of particles to larger sizes (MCCAVE, 1983).

Turbulent inertial coagulation. In turbulent flow local turbulent accelerations produce relative particle velocities for particles of unequal mass (PRUPPACHER and KLETT, 1978, p. 375). The resulting coagulation kernel is

$$K_{TIj} = \frac{\pi d_j^2}{4} (t_j - t_i) e^{3/4} v^{-1/4}, \quad (7)$$

where t_i and t_j are the viscous relaxation times, the time scale for the particle in Stokes flow, given by $t_j = d_j^2 \Delta \rho_j / 18\mu$, where $\Delta \rho_j$ is the *in situ* density contrast of particles with diameter d_j . If the particle is travelling at a speed U_0 then its stop distance is $U_0 d_j^2 \Delta \rho_j / 18\mu$. The relative

importance of the two turbulent mechanisms is

$$\frac{K_{TI}}{K_{TS}} \approx \frac{\Delta \rho_j (d_j - d_i)}{4 \rho \lambda_k} \approx \frac{(d_j - d_i)}{4 \lambda_k} \quad (8)$$

for $\rho_{si} = \rho_{sj} \sim \rho$ with the Kolmogorov microscale length $\lambda_k = (\nu^3/\epsilon)^{1/4}$. Using $\nu = 0.014 \text{ cm}^2 \text{ s}^{-1}$ and ϵ in the range 10^{-3} to $10^{-4} \text{ cm}^2 \text{ s}^{-3}$, λ_k is 0.23 to 0.4 cm and possibly ~ 0.1 cm in higher stress ($u_* \sim 2 \text{ cm s}^{-1}$) events. This shows that turbulent inertial collection is normally only important when particles differ in diameter by more than $4\lambda_k$ or more than 1 cm. Thus the mechanism may aid impaction of very small on very large particles (clay onto 'marine snow'), but will not influence collision of most particles in the ocean for which $d \ll 1$ cm. In fact the assumption $\rho_{si} = \rho_{sj}$ is not good and the plotted values of the kernel (Fig. 3c) are two to three orders of magnitude less than those for turbulent shear.

Differential settling. The faster settling of larger particles through a suspension leads to collisions with slower settling particles and their capture to form aggregates. The kernel is given simply by the area swept out times the relative velocity, the collision efficiency E_{Cij} , and the efficiency of diffusive collection E_{Dij}

$$K_{Gij} = \frac{\pi d_j^2}{4} (w_{sj} - w_{si}) (E_{Cij} + E_{Dij}) \quad (9)$$

The collision efficiency is the ratio of the number of particles with which the settling particle makes contact to the number in the volume it sweeps out as it sinks. The ratio is small (think of trying to catch small particles on your finger in a bowl of water, most of them elude capture). The collision efficiency is given by PRUPPACHER and KLETT (1978) as

$$E_{Cij} = \frac{(d_i/d_j)^2}{2(1 + d_i/d_j)^2} \quad (\text{where } d_j > d_i) \quad (10)$$

and by FRIEDLANDER (1957, 1965) as

$$E_{Cij} = 1 - \frac{3}{2(1 + d_i/d_j)} + \frac{1}{2(1 + d_i/d_j)^3} \quad (11)$$

When the Reynolds number of the settling particle $w_s d/\nu$ is $\gg 1$ the efficiency is also given by the Stokes number, the ratio of the stop distance for the small particle at a relative speed $U = (U_j - U_i)$ to the linear dimension d_j of the large particle (TWOMEY, 1977, p. 59)

$$St = \frac{\Delta \rho_i d_i^2 U}{9 \mu d_j} \quad (12)$$

Small particles are also transferred by Brownian diffusion to the surface of the larger settling particle. The collection efficiency by diffusion is the ratio of the diffusive flux of small particles onto the catcher to the flux of particles moving past it due to its settling. This is related to the Peclet number by $E_{Dij} = 4Pe^{-1} (1 + 0.36 Re^{1/6} Pe^{1/3})$ in the diffusive range and $E_{Dij} = 1.7 Pe^{-2/3}$ in the transition from the diffusive to the inertial range, where $Pe = d_j (w_{sj} - w_{si}) D_i$ (TWOMEY, 1977, p. 146, FRIEDLANDER, 1967, HAMPL *et al.*, 1971). The transition is in the region $Re \sim 1$ to 10.

Values of the various formulae for efficiency have been plotted in Fig. 4 for oceanic conditions. The expressions of Friedlander and of Pruppacher and Klett for the viscous range have

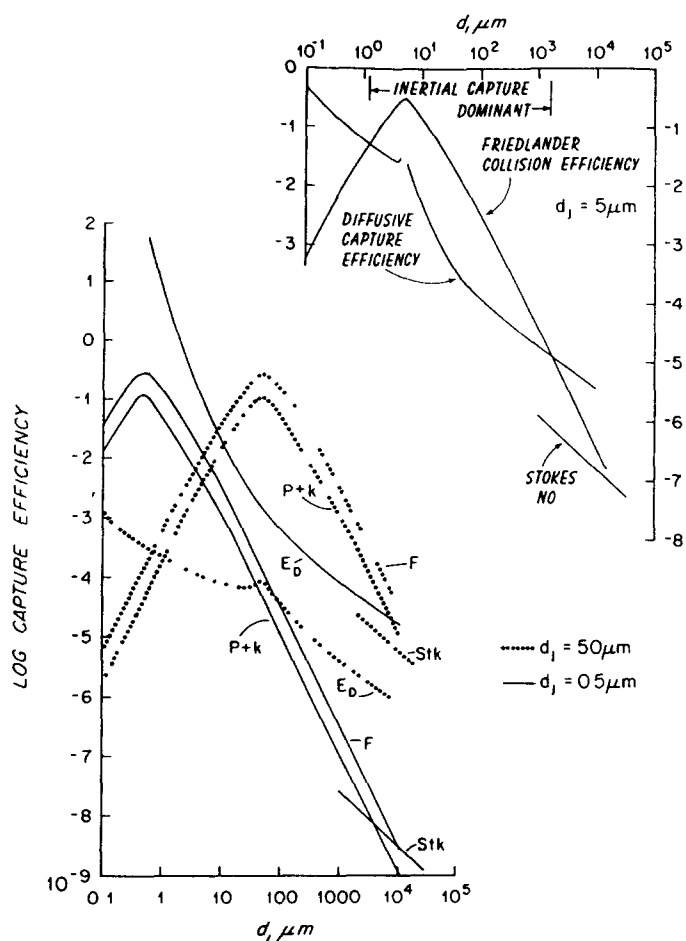


Fig 4 Collection efficiencies for settling particles by Brownian diffusion (E_D) and by collision according to FRIEDLANDER (1957) (F), PRUPPACHER and KLETT (1978) (P + K) and the Stokes number (Stk). These will only yield the actual overall capture efficiencies if the coalescence efficiency is unity.

a similar trend but data do not indicate which is correct. The former is used here. The Stokes number merges well with the other curves for sizes < 1 cm ($Re \geq 10$).

There are several cases of interest that can be illustrated, the capture of small particles ($d = 0.5$ and $5 \mu\text{m}$) by medium sized ($d \sim 50 \mu\text{m}$) and large ($d \sim 5$ mm) ones, and the capture of medium by large particles (Table 2). The efficiency for diffusive capture of $0.5\text{-}\mu\text{m}$ particles is always greater than that for collision, while for $50\text{-}\mu\text{m}$ particles most interactions are dominated by collision (Fig. 4). For $5\text{-}\mu\text{m}$ particles also most of the range of interest is dominated by collision save for particles $< 1 \mu\text{m}$, where diffusion is most important. The figures (Table 2) show also that inertial capture of small particles by 'marine snow' is most inefficient and is not important, but that diffusive capture is relatively efficient. Settling through mid-water where there are perhaps 3×10^4 particles cm^{-3} of size 0.35 to $0.7 \mu\text{m}$ a piece of 'marine snow' of diameter 5 mm and cross-sectional area 0.2 cm^2 sinking through 1000 m would collect 1700 particles in that size range assuming 10% coalescence efficiency.

Table 2 *Settling capture efficiencies*

Caught (μm)	Catcher			
	5 mm snow	50 μm aggregate		
	E_D	E_C	E_D	E_C
50	1.43×10^{-6}	<u>1.47×10^{-4}</u>	—	—
5	<u>6.16×10^{-6}</u>	1.5×10^{-6}	5.8×10^{-4}	<u>1.2×10^{-2}</u>
0.5	<u>2.85×10^{-5}</u>	1.5×10^{-8}	<u>2.66×10^{-3}</u>	1.47×10^{-4}

Underlined values are the most efficient for interactions of that pair of particles E_D = diffusive capture, E_C = collision capture

This may be important for the individual aggregate, but to sweep the water column clean of such particles would require about 35,000 more particles following the path of the first! In fact, the 'snow' catches the 50- μm aggregate more efficiently and the 50- μm aggregate catches the 5- μm one most efficiently. However, it will be seen below that turbulent agitation of large particles leads to more contacts with small ones than does any mechanism simply related to their sinking.

Clearly the efficiency of inertial capture increases as the relative sizes of the catching and caught particles become closer. Thus, small particles are most efficiently coagulated into aggregates a little larger and so on through the spectrum up to particles of millimetre dimensions or more in estuaries (BIDDLE and MILES, 1972). Thus, there could be a cascade of particles from small into large sizes which, judging from the calculations for monodisperse suspensions, would take many years to hundreds of years to achieve flat volume distributions in the deep sea. The larger particles have settling velocities increasing as d^2 in the Stokes region and so are increasingly rapidly lost from the system. It must also be noted that the larger particles, particularly 'snow', are not produced by aggregation of small ones, they start out with a large organic substrate. With evolution of size spectra to flatness there is also a decrease in concentration. Eventually, far from erosional sources and at mid-water depths far from surface sources, some large sinking particles may contribute small particles by breaking up while others continue to sweep their way through the water column, perhaps resulting in a steady state.

Biogenic aggregation processes It is possible to write down a coagulation kernel for active particle catching and aggregate production by animals. Basically they sweep or filter a certain volume of water each second, $V = AU$, where A is the area of water column swept at speed U , and catch particles with an efficiency E_{Bij} , which depends on the type of organism j , the size d_i of the particle, and its perceived food value f . The kernel would have the form $K_{Bij} = AUE_{Bij}$.

Microzooplankton with flagellae or cilia appear to act as filters (FENCHEL, 1980, JORGENSEN, 1983). For the Tintinnids, data of STOECKER and GUILLARD (1982) suggest filtration rates of $\sim 10 \mu\text{l h}^{-1}$ per animal. With populations of, for example, 10^2 to 10^4 l^{-1} (HEINBOCKEL and BEERS, 1979), fractional filtration rates are 10^{-1} to 10^{-3} h^{-1} giving complete filtration times for the water of 0.5 to 50 days. Mucous membranes and nets employed by other, large, zooplankton (ALLDREDGE and MADIN, 1982) also operate as filters (FLOOD and FIALA-MEDIONI, 1981, JORGENSEN, 1983). In several species of ascidian the efficiency of such mucous nets is 100% for particles over 3 μm in diameter (RANDLOV and RIESGARD, 1979). Data summarized by MADIN (1982) and ALLDREDGE and MADIN (1982) for gelatinous zooplankton using mucous nets and films show filtration rates of 6 to 60 ml h^{-1} for

Appendicularia, 5 to 12 ml h⁻¹ for Doliolids, and a remarkable range from 3 up to 5000 ml h⁻¹ for salps. Salp population densities can be as high as 65 m⁻³ in swarms (WIEBE *et al.*, 1979), but they are often more sparsely distributed, 8×10^{-3} m⁻³ as measured by MADIN (1982). At a rate of $\sim 3 \times 10^{-3}$ m³ h⁻¹ per animal, a *Salpa aspera* swarm would completely filter the surface water in 5 h. The lower population density would still give significant aggregation rates of larger particles compared with rates due to inorganic processes alone.

Copepods it now appears are suspension but not filter feeders as often supposed (KOEHL and STRICKLER, 1981, PAFFENHÖFER *et al.*, 1982, JORGENSEN, 1983). They are quite selective about the particles they capture and do not take many small particles. They process appreciable volumes of water however (4 to 8 ml h⁻¹ per animal from data summarized in ALLDREDGE and MADIN, 1982). Population densities of copepods in surface waters are highly variable but a representative range for oceanic waters from lower to higher productivity (eastern North Atlantic to Gulf Stream) is 10 to 100 m⁻³ (GRICE and HULSEMAN, 1965, GRICE and HART, 1962). This decreases to <1 m⁻³ below 1000-m depth and may be as low as 0.1 m⁻³ at 4000 to 5000 m (GRICE and HULSEMAN, 1965). At the filtration rates given above the denser populations result in fractional filtration of 0.02 day⁻¹ or complete processing in 50 days. By contrast the deep populations give very small fractional filtration rates ($\sim 0.4\%$ per year). At the surface the animals can scavenge and aggregate a significant fraction of the bigger particles ($\geq 5 \mu\text{m}$) and convert them into rapidly settling faecal pellets (KOMAR *et al.*, 1981).

The data show that animals are likely to have the major impact on particle spectra in the upper layers of the ocean where there is most food. It is not clear what the effects will be at great depths in mid-water and in the nepheloid layer, where even the population densities are not well known (WISNER, 1980).

Particle breakup

In equation (3) the breakup terms seven and eight due to JEFFREY (1982) were inserted. Earlier VALENTAS and AMUNDSON (1966) and VALENTAS *et al.* (1966) proposed breakup terms, which SPIELMAN (1978) summarized as

$$\frac{\partial n(v_i)}{\partial t} = \int_{v_j}^{\infty} J(v_i, v_j) n(v_j) dv_j - n(v_i) \int_0^{v_i} \frac{v_j}{v_i} J(v_j, v_i) dv_j \quad (13)$$

The breakup function $J(v_i, v_j)$ gives the rate of production of v_i sized particles from breakup of v_j sized particles as $J(v_i, v_j) n(v_j) dv_j dv_i$. However it is by no means clear what should be the functional form of the J terms of JEFFREY (1982) or the other authors. They depend to some extent on the modes of particle breakup.

PARKER *et al.* (1972) proposed that particles either are fractured or are eroded particle-by-particle through surface shear. Their analysis showed that, for particles smaller than the Kolmogorov microscale λ_k , the maximum size stable to surface erosion should be $d_m \propto \Gamma^{-1}$, and for $d > \lambda_k$ it should be $d_m \propto \Gamma^{-2}$, while in both regimes the breakup size should be $d_m \propto \Gamma^{-1/2}$. PARKER *et al.* (1972) cited experimental support for such dependencies. Examination of floc size distributions frequently shows many flocs larger than λ_k , which PARKER *et al.* (1972) conclude must be undergoing surface erosion but are also maintained by high rates of aggregation. The rate of production of particles by surface erosion below the microscale was shown to be $dN/dt = K_e \Gamma^2$, where K_e is the floc breakup rate coefficient.

Different relationships for floc fracture were proposed by TAMBO and HOZUMI (1979). They

suggested that $d_m \propto \epsilon^{-1/2(3+k_p)}$ in the viscous subrange and $d_m \propto \epsilon^{-1/(1+k_p)}$ in the inertial subrange, k_p is the exponent in the relationship between floc size and density having a range $k_p = 1$ to 1.5 , so $d_m \propto \epsilon^{-(0.38 \text{ to } 0.31)}$ and $d_m \propto \epsilon^{-(0.5 \text{ to } 0.4)}$. If we say for the viscous subrange $d_m \propto \epsilon^{-1/3}$, then $d_m \propto u_*^{-1}$ or $\tau_0^{-1/2}$ rather than τ^{-1} as suggested by KRONE (1962). TAMBO and HOZUMI's relationships for the viscous range are supported by photographic size data from a stirred flocculator. They found $d_m \sim 1.5$ mm at $\epsilon = 2 \cdot 10^{-2} \text{ cm}^2 \text{ s}^{-3}$ and $200 \mu\text{m}$ at $\epsilon = 10 \text{ cm}^2 \text{ s}^{-3}$ ($\Gamma = 1.4$ and 32 s^{-1}) for clay-aluminium flocs. This suggests that in the generally low shear environment of the sea, flocs of several millimetres and more will be stable while in higher shear zones near the bed 100 to $200 \mu\text{m}$ may be the limit. Under depositional conditions $\tau_0 \leq 0.1 \text{ Pa}$ ($u_* < 0.98 \text{ cm s}^{-1}$) yielding, in the viscous sub-layer, a shear rate $dU/dz \leq 100 \text{ s}^{-1}$. Equating this with the local turbulent shear rate $(\epsilon/\nu)^{1/2}$ in the vicinity of suspended particles suggests that an equivalent condition is $\epsilon \leq 100 \text{ cm}^2 \text{ s}^{-3}$. Extrapolating the results of TAMBO and HOZUMI (1979) indicates particles of diameter up to $100 \mu\text{m}$ would be stable, and for deposition under $\tau_0 = 0.031 \text{ Pa}$ ($u_* = 0.55 \text{ cm s}^{-1}$) particles up to $200 \mu\text{m}$ would be stable.

The above values of size depend on floc strength, and clay-aluminium flocs are not particularly strong. For example, TAMBO and HOZUMI (1979) found that the addition of 2 ppm of an organic polymer flocculent increased floc strength by a factor of 48 and raised the maximum diameter at $\epsilon = 10 \text{ cm}^2 \text{ s}^{-3}$ from 0.2 to 0.6 mm, and, at $\epsilon = 0.4 \text{ cm}^2 \text{ s}^{-3}$, from 0.54 to 1.6 mm. Organically produced polymers are also capable of increasing the efficiency of coagulation (decreasing stability of the suspension) and are used in wastewater treatment to promote flocculation (BUSCH and STUMM, 1968; GREGORY, 1978; O'MELIA, 1978).

Numerical models such as that of IVES and Bhole (1973) show that breakup may exert a controlling influence on size distributions. Differing distributions are produced by breakup into 2, 3, or 4 fragments. None of the theoretical treatments yet goes much beyond total disintegration or breakage in two. PARKER *et al.* (1972), TAMBO and HOZUMI (1979), and JEFFREY (1982) all consider some floc breakage mechanism by a straining flow resisted by floc strength. However no kernels for breakup (J terms in equations 3 and 13) have yet been proposed.

SOLUTIONS TO THE GENERAL DYNAMICAL EQUATION

The general dynamical equation cannot be solved other than numerically even if simplifying assumptions such as $\partial/\partial t = 0$, $\partial/\partial x = 0$, $D \ll \epsilon_{xx}$, and $w_s(v)$ is constant with z are made. One complex part of the equation is solution of the coagulation terms. Two principal approaches have been made by Friedlander and his co-workers (FRIEDLANDER, 1977; see also TWOMEY, 1977, p. 132 and PRUPPACHER and KLETT, 1978, p. 399). A 'self-preserving distribution' (FRIEDLANDER and WANG, 1966) is an asymptotic solution to the coagulation equation where, after a long time, the coagulating suspension acquires a size distribution whose shape is related to the mechanism governing coagulation. The solution to the equations is achieved by using similarity transformations that reduce the coagulation equation to a function of one independent variable. The conditions for a self-preserving distribution to fit a measured one are that the distribution should have undergone a long evolution by coagulation (when 'long' is of order $10 t_c$), and should have neither gained nor lost particulate matter. Such distributions are not applicable to the oceans because of sedimentation and dissolution and because on the time scales involved, thousands of years at mid-ocean depths and a few years close to the bed, there would be considerable departure from constant oceanic conditions. A similar

lack of applicability was found for the atmosphere by JUNGE (1969) The 'steady-state solution' (FRIEDLANDER, 1960a, b) is not exactly a solution but through dimensional analysis gives the form of the distribution for different subranges dominated by different coagulation mechanisms

FRIEDLANDER (1960a, b) proposed that for distributions where there was production of particles at the fine end and removal by sedimentation at the coarse end, the form of the distribution is determined by the parameters describing the coagulation and sedimentation mechanisms and the flux of matter through the spectrum In this he drew analogy with the theory underlying spectral analysis of turbulence, and the form of the distributions deduced are termed 'quasi-stationary' In equilibrium the size spectrum displays several subranges each dominated by a single coagulation mechanism There is a flux of particulate matter through the distribution from smaller to larger sizes ending in sedimentation of large particles In the atmosphere there is a source of small particles from condensation but in the ocean such input at the fine end is more sporadic and largely confined to erosion of the bed and input of aerosols and biogenic particles at the surface The subranges are dominated by Brownian motion, shear, and differential sedimentation An aspect of the concept of a subrange pointed out by JEFFREY (1981) is that collision between particles of similar size should make most contributions to term (vi) of equation (3) Again, if there is net loss of material from a distribution or if it changes shape with time it is not stationary and Friedlander's method is inapplicable

In the subrange where Brownian motion dominates the particle spectrum, dimensional analysis of $n(d)$, d , $K = 2kT/3\mu$, and E_m the material flux through the distribution, with dimensions of

$$n(d) = L^{-3} l^{-1}, d = l, K = L^3 T^{-1},$$

and

$$E_m = l^3 L^{-3} T^{-1},$$

where l and L are particle and fluid length dimensions respectively, yields the result

$$n(d) = A \left(\frac{E_m}{K} \right)^{1/2} d^{-5/2}. \quad (14)$$

Thus the cumulative distribution $\Delta \log N / \Delta \log d = n(\log d)$ varies as $d^{-3/2}$ or $\beta = 3/2$ It is of considerable interest that HARRIS (1977) measured particle number in the range 0.1 to 1 μm in a nepheloid layer and showed a variation of cumulative number as $\beta = 1.65$ suggesting a Brownian-controlling mechanism in the submicron range.

HUNT (1980) applied the same similarity argument to additional subranges dominated by shear and differential sedimentation coagulation. For the cumulative number distribution he finds the following dependencies on d ; for shear $\beta = 3$ and for differential sedimentation $\beta = 3.5$. In addition for relatively high shear conditions, $\Gamma = 3 \text{ s}^{-1}$, he suggested that the shear subrange occupied the region $d = 1$ to 100 μm and that Brownian and differential sedimentation dominated at lower and higher diameters, respectively. The prediction of a flat volume distribution when shear is dominant was verified experimentally by HUNT (1982a, b).

For valid application of the similarity arguments E_m must be constant with respect to size and time, so particle input and removal by non-coagulative processes act against maintenance of constant E_m . As there is only episodic fine particle production in nepheloid layers and as there is likely to be deposition of a substantial part of the size range for most of the time that

$u_* \leq 0.8 \text{ cm s}^{-1}$ (McCAVE and SWIFT, 1976), the occurrence of quasi-stationary distributions may not be common. In mid-water situations the possibility of a source of fine particles is even less likely and one would not expect a Brownian subrange with a $d^{-1.5}$ slope there.

Several authors have solved the coagulation equation numerically for particular aerosol problems. LINDAUER and CASTLEMAN (1971) examined the case of Brownian coagulation with gravitational fallout assuming an initial log-normal number size distribution. With time the mean size increases at first and then decreases—particle growth by coagulation is overtaken by fallout. In WALTER's (1973) numerical experiment, condensation nuclei ($d_p = 1.2 \text{ nm}$) are coagulated and show a steep fall-off with size and development of a secondary peak at 0.1 to 0.3 μm . If larger particles (0.1 μm) are introduced they rapidly scavenge out the smaller ones and leave a gap in the spectrum for $d_p < 0.01 \mu\text{m}$. Similar numerical experiments would be illuminating for the case of marine particles.

PHYSICAL PARTICLE INTERACTIONS IN THE OCEAN

The regions dominated by the different coagulation mechanisms are shown in Fig 3 in a form based on the original suggestions by FRIEDLANDER (1965). The coagulation rate constants from the kernels are plotted for interactions with particles of size $d_i = 0.5, 5, \text{ and } 50 \mu\text{m}$. The conditions assumed are those of low shear, $\Gamma = 0.084 \text{ to } 0.0084 \text{ s}^{-1}$, and deep-sea density and viscosity mentioned above with particle properties of Table 1. Brownian motion dominates interactions of fine particles, followed by an intermediate range where differential sedimentation is most effective, and finally shear dominates for large particles because of the d_v^3 term in the kernel. This differs from the sequence Brownian—shear—differential sedimentation proposed by HUNT (1980) using $\Gamma = 3 \text{ s}^{-1}$ ($\epsilon \sim 0.1 \text{ cm}^2 \text{ s}^{-3}$) appropriate for some coastal waters. According to the plots shown here, differential sedimentation should dominate coagulation between similar-sized particles over most of the range of interest in the sea (1 to 100 μm). However, the 'giant' particles represented by marine snow appear to be dominated by shear in their interactions.

Such a plot may seem to reinforce the notion of a cascade of particle volume through the size distribution. Particle collisions with near neighbours (in size) are favoured so small aggregates join to make bigger ones and so on up to large sizes. However, the time scales calculated for the processes are very long with 10% efficiency; Brownian coagulation times for 0.5- μm particles are 3 months to 20 years for nepheloid layer and mid-water concentrations, respectively, and corresponding shear-coagulation times for 5- μm particles are from 7 months to 7000 years. This makes it unlikely that small particles can aggregate into big ones other than through biological intervention to produce faecal pellets. Apparently the mill grinds very slowly.

But does it? A feature of the turbulent shear kernel is that it attains high values for interaction involving large particles. One way of examining the effect of this is to calculate particle removal rate as suggested by LERMAN (1979, pp. 288–299) [cf. Hidy's (1973) Tables 4 to 6 for aerosols]. This is given by the kernel times the numbers of interacting particles $K n(v_i) n(v_j)$ with units $\text{cm}^{-3} \text{ s}^{-1}$. Table 3 shows the results of calculations for nepheloid layer and mid-water concentrations. The numbers of smaller particles are based on data from McCAVE (1983). The numbers of giant (2 to 4 mm) particles are preliminary data from the photographic observation system of HONJO *et al* (1984) at a low-productivity North Atlantic station ($34^\circ 17' \text{N}$, $73^\circ 27' \text{W}$, 4140 m deep) (HONJO and ASPER, personal communication). The values are about 100th those reported by HONJO *et al* (1984) under the productive California

Table 3 Particle removal rates* $-dn/dt$ ($\text{cm}^{-3} \text{s}^{-1}$)

	Nepheloid layer ($\Gamma = 0.084 \text{ s}^{-1}$)			Mid water ($\Gamma = 0.0084 \text{ s}^{-1}$)		
$d_i =$	0.5 μm	5 μm	50 μm	0.5 μm	5 μm	50 μm
L_B	6.74	6.07×10^{-5}	2.43×10^{-12}	6.74×10^{-4}	6.07×10^{-9}	1.22×10^{-14}
L_S	1.37×10^{-2}	1.23×10^{-4}	4.93×10^{-9}	1.37×10^{-7}	1.23×10^{-9}	1.23×10^{-12}
$L_{S(2-4)}$	1.86×10^{-2}	5.61×10^{-5}	1.18×10^{-8}	6.20×10^{-6}	1.87×10^{-8}	1.97×10^{-11}
$L_G(50)$	5.68×10^{-4}	8.85×10^{-6}	—	2.84×10^{-7}	4.44×10^{-9}	—
$L_{G(2-4)}$	2.42×10^{-3}	2.34×10^{-6}	1.35×10^{-8}	8.06×10^{-6}	7.80×10^{-9}	2.24×10^{-10}
$N_i(\text{cm}^{-3})$	10^6	3×10^3	0.6	10^4	30	0.03
$N_{2-4}(\text{cm}^{-3})$		6×10^{-5}			2×10^{-5}	

* Assuming 100% efficiency

N_i is the number concentration of particles of diameter d_i

$N_{2 \text{ to } 4}$ is the number concentration of 'snow' particles in the size range 2 to 4 mm

Subscripts for loss rates L are B = Brownian, S = shear, G = differential sedimentation, (2 to 4) mm and 50 μm

Frequencies $< 3.17 \times 10^{-8} \text{ cm}^{-3} \text{s}^{-1}$ represent less than one collision per year

current. The calculated loss rate for Brownian (L_B) and shear (L_S) assume a monodisperse suspension ($d_i = d_j$), while the loss rates due to shear L_S (2 to 4) and differential sedimentation L_G (50) and L_G (2 to 4) presume interaction of particle d_i with the larger particle specified in the subscript

The most important feature of Table 3 is that many of the collision frequencies are $< 3.17 \times 10^{-8} \text{ cm}^{-3} \text{s}^{-1}$, or less than one collision per year per cm^3 . There is no significant scavenging of 50- μm particles (having a settling rate of $\sim 15 \text{ m day}^{-1}$) by larger ('snow') particles, or aggregation with similar-sized particles before they have settled out of the water column. Differential settling loss rates come out less than those for the turbulent shear rate L_S despite the fact that the plot of the kernels (Fig. 3) shows that differential settling should dominate. This is a consequence of the fact that L_S is calculated for a monodisperse suspension and has $[n(v_i)]^2$ while L_G (50) has $n(v_i)n(v_j)$ where $n(v_j)$ is not very large. This reinforces the point about effective loss rates being greatest for near neighbours, at least by gravitational settling. At mid-water depths where concentrations are low there is inefficient scavenging of small particles by large ones—whether by settling or turbulent agitation. The principal mechanism of removal of 0.5- μm particles there involves 'snow' particles (2 to 4 mm here) in shear- and settling-controlled collisions but at such low frequencies that the collision rate involves only 5% of the small particles in a year. By the time something happens to fine particles at mid-water depth they have moved elsewhere. Physical aggregation in the nepheloid layer with appreciable frequency is again dominated by shear for 5- μm particles and Brownian motion for submicron sizes. Shear both aggregates similar-sized particles and leads to scavenging of small particles by 'snow' more effectively than differential settling. This presumably also applies to shear-controlled aggregation in the surface mixed layer though shear rates there (thus aggregation rates) may be an order of magnitude higher. Numbers of particles near the surface may also be higher than those assumed, again resulting in increased collision frequency.

The enhanced coagulation rates for small particles with large particles in nepheloid layers suggests that this will be a highly significant mechanism in surface waters too, where turbulence levels expressed as a shear rate may be an order of magnitude greater. In addition, the

concentrations of marine snow may be two orders of magnitude greater than the figure given here [i.e., $5 \times 10^{-3} \text{ cm}^{-3}$ from SHANKS and TRENT (1980) and $\sim 10^{-3} \text{ cm}^{-3}$ from HONJO *et al* (1984)]. Thus, in surface water the smallest particles are aggregated by Brownian motion. Such aggregates are lost by colliding with one another in motion controlled by differential settling, but they are scavenged even more rapidly by large particles with turbulent shear controlling the relative motion. The potential importance of large scavenging particles decreases the importance of FRIEDLANDER's (1965) notion of subranges because coagulation may be dominated by particles that are very dissimilar in size.

The efficiency of particle capture has here been assumed to be around 10%. However, bacterial and algal exuded polymers may promote flocculation by increasing efficiency and by increased floc strength. Recent work has demonstrated that 'marine snow' is the site of extremely active bacterial and algal production (CARON *et al*, 1982; KNAUER *et al*, 1982). It seems likely that this will result in 'snow' being a more efficient scavenger of small particles as it settles. Such enhancement of efficiency may not be restricted to 'marine snow' as bacteria attach to many surfaces, particularly in low-nutrient media (JANNASCH and PRITCHARD, 1972, FLETCHER, 1979). Perhaps 10% efficiency is a lower limit.

The above discussion has considered physical factors involved in aggregation. It must be tempered by the fact outlined in the previous section that some filter-feeding organisms can process the upper water column in a day. Brownian aggregation is likely to put small particles into sizes 1 to 8 μm , where they stand a good chance of being filtered by larger zooplankton if they have not already been collected by microzooplankton. More data on filtration rates and population densities of suspension feeders, particularly below the surface layers, will be essential in elucidating deep-sea particle dynamics. Animals may dominate at depth as it seems probable that they do in the surface.

SIZE DISTRIBUTIONS OF AEROSOLS AND MARINE PARTICLES

Flat oceanic and atmospheric spectra

Flat volume distributions ($\beta = 3$) are a feature of both marine particulate matter and aerosols. In the latter case it is known as the Junge distribution or the Junge subrange. It is of more than passing interest that both in the atmosphere and the oceans there is a tendency to have equal volumes of material in logarithmically increasing size grades. In the atmosphere this holds over the range 0.1 to 100 μm (JUNGE, 1969) and in the ocean from 1 to >100 μm (SHELDON *et al.*, 1972), possibly out to 1000 μm if scaling with the atmosphere is similar. Why should this be so, and are there any other general lessons the marine sciences can draw from the situations in the atmosphere? What are the exceptions to the pattern and what do they tell us?

The role of component subdistributions

In the ocean the data of LAMBERT *et al.* (1981) indicate that the suspended material is made up of a variety of components, each with a log-normal distribution perhaps, and that they add up to a Junge distribution over part of the range. In the ocean, as in the atmosphere, when close to the source (surface or bottom), size distributions are not flat but show various peaks characteristic of the source. The peaks may be related to particular components (e.g., foraminifera, clay, and silt particles in the ocean; fly ash or salt particles in the atmosphere). They may also be related to aggregation, for example, formation of a peak by collisions due to Brownian motion. Oceanic and atmospheric particles are similar in this respect, thus JUNGE's

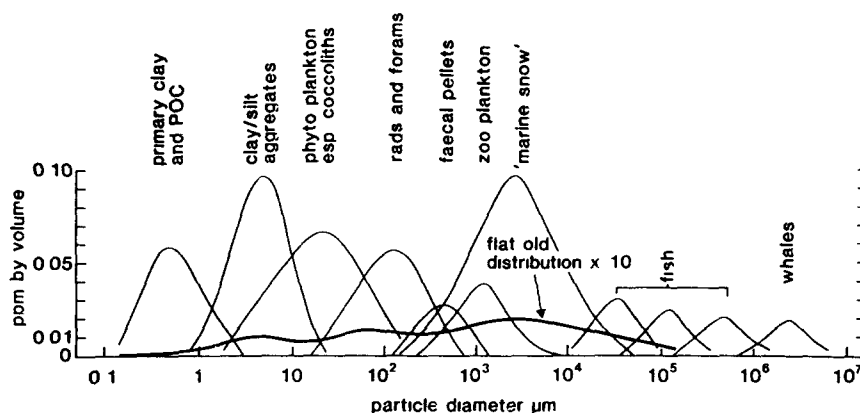


Fig 5 Hypothetical wide-spectrum flat size distribution by volume with suggested component distributions that make it up

(1969) suggestion that mixing of different aerosol components from differing sources produces a statistical combination yielding $\beta = 3$ may also apply to the oceans. I suggest that in a polydisperse system the component peaks cause local regions (in particle volume space) of increased coagulation rate that tend to flatten out the bumps, and that in a polydisperse system shear dominates the coagulation aided by differential settling. The initial condition leading to a flat distribution is thus production of subequal amounts (differing by no more perhaps than one or two orders of magnitude) of material to yield the different initial peaks (Fig 5). The approach to $\beta = 3$ is by no means perfect and values from 2.7 to 3.5 are common, still with some bumps in the volume distributions. I suggest that the coagulation rates in a poly-disperse system will be fast enough to flatten most of the bumps arising from the source, and that the reported fits to the Junge distribution are not all that good (as is evident when spectra are plotted as particle volume instead of particle number). Several numerical experiments to examine the suggestion are possible.

Quasi-stationary evolution of distributions

Both for the atmosphere and for the ocean it has been suggested that the Junge distribution represents asymptotic behaviour, the evolution of spectra to a stable form as exemplified by self-preserving or quasi-stationary distributions. For the ocean the latter case is argued by HUNT (1980) yet he used shear rates and concentrations typical of coastal waters (where the Junge distribution does not obtain) to maintain that the observed flat oceanic distributions are shear controlled. In the atmospheric case JUNGE (1969) disputed the applicability of self-preserving distributions anywhere, and argued that quasi-stationary distributions may only apply to the finest or largest particles, not those in the middle of the distribution where $\beta = 3$. The criteria for applicability of quasi-stationary distribution are that particle gain and loss are about in balance and that the rate of these processes is greater than the rate of change of oceanic (atmospheric) conditions, i.e., approximately stationary conditions. In the atmosphere only the Brownian region and the largest settling particles are thought to have rates of particle flux through the distribution that are fast enough. In the ocean the Brownian range ($< 1 \mu\text{m}$) particles may coagulate fast enough shortly after their erosional supply but would not last for long in equilibrium. This may be why in one nepheloid layer HARRIS (1977) found a slope

approaching that for Brownian coagulation whereas elsewhere in low-concentration conditions BAKER *et al.* (1979) and LAMBERT *et al.* (1981) did not. Even the small part of the range considered by LERMAN *et al.* (1977) (2.26 to 12 μm) is unlikely to be stable over the required period of at least 6 months. In that time one would expect deposition and some fluctuation in nepheloid layers, possibly including erosion, detachment of mixed layers, and supply from above (McCAVE, 1983). Certainly the oceans are not stable over the very long periods required for coagulation at mid-water depths. Thus it appears that in the oceans the rates of processes in the Junge subrange, $d \approx 1$ to 100 μm , are too slow relative to the rate of change of oceanic conditions to produce flat distributions alone.

Rates and places for peak flattening

The two most effective physical mechanisms recognized for changing size distributions are aggregation of small particles by Brownian motion and collection of smaller particles by turbulent agitation with larger particles. The rates of the processes depend on number concentration and on turbulent shear rate. The overall distribution of small particles sensed by nephelometers (BISCAYE and EITREIM, 1977) and of large particles sensed by new optical systems (HONJO *et al.*, 1984) comprises maxima in surface and bottom regions with a broad minimum in between. The importance of organisms in production of faecal pellets as a means of shifting particles from one end of the spectrum to the other (and from top to bottom of the ocean) in one operation must be emphasized. The upper ocean is most turbulent and it must be there that most particle production and modification of initial size distributions occurs. Increased shear in zones of rapid temperature change and large Brunt-Vaisala frequency (EVANS, 1982) should result in a further smoothing of the size distribution of material entering mid-water from above. Lateral supply of material to mid-water in the open ocean is likely to be a slow process, thus part of the fine material in suspension will be old (order of years) and should not show pronounced peaks. Larger components of the size distribution must have come from above on a time scale of a week to months. Some of the mid-water small particles must also have come from above however, as EITREIM *et al.* (1976) and BISCAYE and EITREIM (1977) showed that under regions of high surface primary productivity the mid-water contains a spatial maximum of fine particles. The largest, fastest settling particles would probably yield distributions that were peaked in relation to time and place (time of plankton bloom, under productive areas), but these have not yet been measured. The <100- μm range usually measured probably contains many relatively old particles (order of a year to many years) and has thus had time to evolve towards flatness.

Ocean-atmosphere comparisons

We should beware of presuming that because there are regions of $\beta = 3$ in both ocean and atmosphere they are therefore equivalent in terms of particle dynamics. The oceanic region of flat volume distributions is well away from surface and bottom sources whereas that in the atmosphere is in lower regions much closer to source in terms of both space and time. JUNGE (1969) pointed out that aerosols in clean tropospheric air masses have $\beta \sim 4$, thus the region is not a good analogue for the mid-water situation. The higher value of the exponent in oceanic mid-water, indicative of relatively more large particles in the distribution than in tropospheric distributions, is perhaps due to the fact that the ocean has a source of particles from above (faecal pellets and 'marine snow' falling and breaking up), which provides a spectrum of sizes with relatively more in the coarse end.

Thus the atmosphere, which is more vigorously and rapidly mixed, has particle size distributions that evolve from peaked near the source, through $\beta = 3$ in the lower atmosphere, to $\beta = 4$ in clear tropospheric air. The ocean by contrast moves and mixes much more slowly so that large volumes of surface and bottom water can be dominated by (multiple) peaked spectra. Only far away from individual sources and a long time, probably more than a year, after introduction of particles do ocean spectra become flat, and because of supply from above they do not generally attain a $\beta \sim 4$ form. The closest they get in mid-Pacific water is $\beta \sim 3.5$ (BAKER *et al.*, 1979). The multiplicity of oceanic particle types provides the necessary range of distributions, and the biological activity in surface waters that produces large particles of both 'snow' and faecal pellet types as well as skeletal remains is responsible for ensuring that most regions of the spectrum are represented by source peaks. Thus small particles do not have to work their way all the way up the spectrum to produce a flat distribution. By aggregation and with breakup they help fill in the valleys between the peaks. Geologists should consider that such small, often lithogenic particles, which ultimately constitute the geological record, are only a very small fraction of surface ocean suspended matter, <1% off West Africa according to FOLGER (1970). Thus processes of pelagic sedimentation of lithogenic matter may be viewed as a side effect of excretion and disposal of other waste products of the ocean's biological systems. Such biological intervention in the production of the coarse end of particle spectra (from 10 μm to several cm) is perhaps simply the transition to the situation envisaged by SHELDON *et al.* (1972), where the spectrum continues with equal volumes of biological material in logarithmic size grades out to whale-sized particles.

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