



Rheological properties of marine organic aggregates: importance for vertical flux, turbulence and microzones

Rheology
Marine organic aggregates
Vertical flux
Microzones
Turbulence

Rhéologie
Agrégats organiques marins
Flux vertical
Microzones
Turbulence

Ian R. JENKINSON ^a, Bopi A. BIDDANDA ^b, Carol M. TURLEY ^c, Paulo C. ABREU ^b, Ulf RIEBESELL ^b and Victor S. SMETACEK ^b

^a Agence de Conseil et de Recherche Océanographiques, Lavergne, 19320 La Roche Canillac, France

^b Alfred-Wegener-Institut für Polar- und Meeres-Forschung, am Handelshafen 12, D-2850 Bremerhaven, Germany

^c Plymouth Marine Laboratory, Citadel Hill, Plymouth PL1 2PB, England.

ABSTRACT

For marine environments where aggregates are composed of $\geq 50\%$ organic matter, various parameters relevant to aggregation are quantified and compared with those in other aggregation regimes. Some possible environmental effects of the rheological properties of marine organic aggregates are reviewed. The interaction between shear forces in the water and aggregate rheology is likely to determine: firstly, the sinking speed of aggregates and hence vertical organic flux rates; secondly, the volume of microzones, with implications for equilibria and kinetics of chemical reactions in the pelagic milieu; and thirdly, the degree of turbulence damping between aggregates, due to absorption of energy through viscous dissipation inside viscoelastic aggregates during their deformation. Whether organic sub-micrometre particles increase the viscosity of bulk seawater at a given lengthscale (such as that of turbulence) will depend on their shape and the degree to which their rheological and surface-active properties allow adhesion or intertanglement to form matrices. Some preliminary results are given of the rheological properties of some laboratory-produced marine organic aggregates.

Oceanologica Acta, 1991. Proceedings of the International Colloquium on the environment of epicontinental seas, Lille, 20-22 March, 1990, vol. sp. n° 11, 101-107.

RÉSUMÉ

Propriétés rhéologiques des agrégats organiques marins : l'importance pour les flux verticaux, les turbulences et les microzones

Pour les milieux marins où les agrégats se constituent à plus de 50 % de matière organique, nous comparons certains paramètres qui concernent le régime agrégatif avec ceux d'autres milieux. Nous donnons un aperçu des effets possibles sur leur milieu des propriétés rhéologiques des agrégats organiques marins. L'interaction entre le cisaillement *in situ* et la rhéologie des agrégats pourrait déterminer : premièrement la vitesse de descente des agrégats, et ainsi le flux vertical organique ; deuxièmement, le volume des microzones, et ainsi l'équilibre et la dynamique des réactions chimiques en milieu pélagique ; troisièmement, la diminution de la turbulence entre les agrégats due à la dissipation visqueuse dans

les agrégats viscoélastiques durant leur déformation. Le degré auquel les particules organiques sous-micrométriques pourraient augmenter la viscosité de l'eau de mer brute à une échelle donnée de longueur (comme celle de la turbulence), serait déterminé par leur tendance à donner des matrices par adhésion et par entremêlement. La forme ainsi que les propriétés rhéologiques et tensioactives de ces particules détermineront cette tendance. Nous présentons quelques résultats préliminaires des propriétés rhéologiques des agrégats organiques marins produits en laboratoire.

Oceanologica Acta, 1991. Actes du Colloque international sur l'environnement des mers épicontinentales, Lille, 20-22 mars 1990, vol. sp. n° 11, 101-107.

INTRODUCTION: COMPARISON OF AGGREGATION REGIMES IN DIFFERENT ENVIRONMENTS

To any seafarer, oceanographer or scuba diver, it is obvious that the mid-Pacific Ocean, the Norwegian fjords, much of the Mediterranean and parts of the North Sea at times are part of "Le Grand Bleu". The Elbe Estuary, however, never is. It is also obvious to the visitor that the sediment-coloured waters of most estuaries with alluvial sediments are the colour they are because intense bottom-generated turbulence opposes settling out by continually sweeping sediment back into suspension. It is in these waters, that a huge amount of research has been done on aggregation and settling processes, largely because of the concern generated by coastal erosion and the silting up of shipping channels. Similarly, much work has been done on aggregation in another turbulent system, that of the activated sewage sludge plant. Research on aggregation in blue and green waters has proceeded more slowly.

As the present contribution concerns organic aggregates, we feel it useful to define some of the more evident qualitative differences and similarities among aggregation regimes in the sea and in sludge plants. These are expressed in Table 1.

As suggested in Table 1, low-turbulence green and blue waters generally differ from high-turbulence sediment-coloured waters in the following parameters: colour; origin of mixing energy (surface or bottom); rate of turbulent energy dissipation; total suspended solids; organic or inorganic matter dominating in flocs. They do not appear to differ systematically, however, in: stratification; the relationship between floc size and the excess density of flocs (*i.e.* floc density minus water density). As mentioned in Table 1 (note 2), where sediment finer than sand is in short supply, as off the north coasts of Brittany (Shepard, 1963), the water naturally remains poor in suspended sediment and thus stays blue or green.

Additionally, it seems that green water may represent either blue water with sufficient phytoplankton to turn it green, or occasionally water moderately rich in sediment also with sufficient phytoplankton to turn it green. Since aggregates in blue and green waters are primarily >> 50 % organic, while those in sediment-coloured waters are primarily >> 50 % inorganic, the aggregates in this sediment-rich, green water are likely to be primarily

organic. For these reasons, it appears that most coastal waters can be assigned to one of each of the following three regimes: low-turbulence, surface-mixed blue and green waters; high-turbulence, bottom-mixed blue and green waters; sediment-coloured waters (practically always high-turbulence, and generally bottom-mixed). Mixed water columns also exist, where the bottom water is principally bottom-mixed and the surface water surface-mixed, with changes in water colour and probably in depth-related variation aggregate composition.

POLYMERS AND THE FORMATION OF ORGANIC AGGREGATES IN THE SEA

There is increasing evidence that the viscosity of bulk water is negatively related to shear rate both in some phytoplankton cultures (Jenkinson, 1986; Ramus and Kenney, 1989) and in some parts of the sea (Jenkinson, 1989). Phytoplankton abundance in the sea has also been found positively related to viscosity (Jenkinson and Biddanda, submitted) as well as to the abundance of both organic aggregates (Alldredge and Silver, 1988; Riebesell, 1991 *a*) and sub-micrometre particles, which appear to be mostly non-living and polymeric (Koike *et al.*, 1990). Bacteria, gelatinous zooplankton and, in coastal waters, epibenthos may also contribute thickening materials to seawater. Shear rate in the sea is related principally to the degree of turbulence, and since sheltered coastal waters at times show low turbulence and high biological activity, it is here that the greatest effects of biopolymer-associated thickening may be felt.

As well as being thickened by dispersed polymers, highly productive waters frequently contain organic aggregates of size greater than ≈ 30 µm, in volume fractions of up to 0.7 % (Alldredge and Silver, 1988). In calm, unturbulent Adriatic waters aggregates, sometimes as large as 1 m or more (Stachowitzsch, 1984; Stachowitzsch *et al.*, 1990), can represent a biomass of 10 g dry weight m⁻³ (Herndl and Peduzzi, 1988). While most of this flocculated organic material originates principally from phytoplankton, organic aggregates may be stabilized and even further enlarged, by interparticle bacterial bridges (Biddanda, 1985; 1986), and by the "sticky" mucus secreted directly by phytoplankton (Chang, 1984; Smetacek and Pollehne, 1986). The aggregates are subsequently disrupted by protozoa grazing

on the bacteria (Biddanda and Pomeroy, 1988; Lochte, 1991) as well as by external forces (Eisma and Kalf, 1987; Alldredge and Silver, 1988; Lubbers *et al.*, 1988; Alldredge *et al.*, 1990; Jenkinson, 1990; Turley, 1991).

In a study in which aggregates were found to occupy a volume fraction ranging from 2×10^{-7} to 0.01 excess viscosity, although strongly correlated with chlorophyll level, was not significantly correlated with aggregate volume fraction. This suggests that dispersed rather than

aggregated polymers were the primary source of the measured enhancement in seawater viscosity.

An important question thus arises: if organic aggregates do not significantly increase measured viscosity, can they nevertheless modify turbulence, and if so how can this modification be modelled? We shall suggest that the rheological properties of suspended aggregates may significantly alter, firstly the turbulence spectrum and, secondly the sinking rates of aggregated organic material.

Table 1

Approximate ranges of parameters in three different mixing regimes commonly studied. Numbers in square brackets refer to references given below.

Étendue approximative de paramètres dans trois régimes de turbulence fréquemment étudiés.

Parameter	"Green and blue" waters (low-turbulence)	"Sediment-coloured" waters	Activated sludge plants
Colour	Blue (dominated by colour of pure water) or green (dominated by photopigments) Occasionally brown/yellow (dominated by Gelbstoff in coastal runoff) Rarely other colours (in exceptional algal blooms)	Generally from yellow to brown. Colour dominated by inorganic constituents of suspended sediment.	Generally greyish. Dominated by colour of sludge, including constituent heterotrophs
Dominant origin of mixing energy	Generally surface input (wind, surface waves), less commonly convection (due to surface cooling) or internal origins (internal waves)	Generally bottom mixing (currents, or swell entering shallow water)	(Mechanical mixing)
Mechanical (mainly turbulent) energy dissipation rate, ϵ ($\text{W m}^{-3} = \text{kg m}^{-1} \text{s}^{-3}$)	From $\leq 7 \times 10^{-10}$ in some intermediate waters and oceanic thermoclines, to 10^{-4} or 10^{-3} in surface waters during moderate winds (much more in storms), but as low as $\leq 3 \times 10^{-7}$ in very calm weather [2, 4, 5]	In mid depth: from $\approx 7 \times 10^{-5}$, "the lowest that can be imagined" in estuaries at slack water and at mid depth, to 0.1 or 1 (high turbulence) at a water depth of 90% total depth, for mean current speed of 1.5 m s^{-1} (but up to 3×10^{-5} in the $\approx 5\text{-}\mu\text{m}$ -thick viscous sublayer, just adjacent to the bed) [6] <i>See note 1</i>	Probably higher than general values for "sediment-coloured" waters
Stratification	Stratified to well mixed	Stratified to well mixed	Always (?) well mixed
Total suspended solids g m^{-3}	$\approx 10^{-2}$ to 2 g m^{-3} [6]	1 to 10^5 g m^{-3} [6]	700 to 4800 g m^{-3} [3]
Proportion of organic matter in flocs	Generally $\geq 50\%$ [1]	Generally $\leq 50\%$ [1]	$\geq 50\%$ [3]
Excess density of flocs (based on values determined from size and sinking rates only).	For all marine conditions, from both quiescent, offshore waters, to turbulent estuaries, values lie within a factor of 5 above or below the size-related relationship assumed by McCave (1984), that is $\approx 10^3 \text{ kg m}^{-3}$ for flocs of diameter $1 \mu\text{m}$ to $\approx 2 \text{ kg m}^{-3}$ for 1-mm flocs [6]		

Notes: 1) Values of turbulence used in the laboratory by workers interested in estuarine sediment transport processes lie between viscous sublayer and bulk values for estuarine turbulence: for instance, 2 to 10^3 W m^{-3} (Dietlin, 1982) or 1.2 to 10^3 W m^{-3} , calculated from values given for bed stress (Krone, 1962).

2) Other less common waters exist such as high-turbulence blue or green waters. These are characterized by bottom-dominated mixing and by a turbulent appearance reminiscent of estuarine conditions. Because little fine sediment is available, however, total suspended inorganic matter remains low. Such waters occur, for example, off the north coast of Brittany. Because of high ambient shear rates, rheological properties seem likely to be dominated by the newtonian component of viscosity, η_W .

3) Where turbulence intensity was given in reference by rms shear rate, q , dissipation per unit volume, ϵ , has been calculated from the formula, $\epsilon = 7.5 \eta q^2$, where η is dynamic viscosity, here taken as 10^{-3} Pa s . Similarly, ϵ expressed per unit mass has been converted using a density of 10^3 kg m^{-3} .

*References used in the Table: [1] Eisma and Kalf (1987); [2] Farmer *et al.* (1987); [3] Li and Gancarczyk (1986); [4] Soloviev *et al.* (1988); [5] Van Leer and Rooth (1975); [6] Van Leussen (1988).*

Table 2

Physical properties of laboratory-produced aggregates.

Propriétés physiques des agrégats produits en laboratoire.

Aggregate origin		Equivalent diameter (mm)		Excess density (kg m ⁻³)		Yield stress (Pa)		Critical shear (dimensionless)	
Aged seawater	Mean	0.80	(24)	14	(24)	28	(21)	2.7	(21)
	s.d.	0.21		6		28		1.2	
Aged culture of <i>Skeletonema</i>	Mean	1.21	(35)	6	(35)	27	(33)	1.6	(33)
	s.d.	0.71		2		42		1.1	
Aged plankton tow	Mean	1.16	(26)	8	(26)	15	(24)	0.9	(24)
	s.d.	0.54		5		13		0.7	

Values in parentheses denote the number of particles subject to each measurement. Equivalent diameter represents particle volume in terms of the diameter of a sphere of the same volume. Excess density was determined using Stoke's law from measurement of sinking rate. Yield stress is the stress sustained by an aggregate before it yields. Critical shear is the amount of shear sustained by an aggregate before it yields.

REGULATION OF SIZE IN ORGANIC AGGREGATES

The size of marine organic aggregates is regulated by constructive and disruptive processes. The constructive ones include physical flocculation mechanisms, surface adhesion and biological cementing (McCave, 1984; Biddanda, 1986; Eisma and Kalf, 1987; Alldredge and Silver, 1988; Riebesell, 1991 b). Disruption occurs when the forces exerted on the aggregate, both by shearing in the surrounding water and by drag resistance due to sinking through the water, combine to exceed the strength of the aggregate for long enough to pull it apart (Alldredge *et al.*, 1990; Jenkinson, 1990; Turley, 1991). This is consistent with observations that the largest aggregates occur during episodes of low turbulence (Stachowitzsch, 1984; Alldredge and Gotschalk, 1989; Riebesell, 1991 a).

RHEOLOGICAL PROPERTIES OF ORGANIC AGGREGATES

It has been demonstrated theoretically and in experimental systems that aggregates composed of similar primary particles show elasticity, at least for small deformations (Buscall *et al.*, 1988). Casual (unpublished) observation indicates that marine organic aggregates are also flexible and elastic.

Working with natural, fragile aggregates 3 to 22 mm in diameter, Alldredge *et al.* (1990) found that they broke up when mean turbulent shear forces (calculated from shear dissipation rates given) ranged from 1 to 10 mPa. These forces, which may be taken as approximately equal to the shear strength of the aggregates, were negatively related to aggregate size. In the same study, gelatinous appendicularian houses, as well as aggregates of detrital debris and of sewage flocculation bacteria were stronger than 100 mPa.

Using a rheometer fitted with concentric cylinders (a Couette system), we measured the rheological properties

of aggregates produced in the laboratory from aged seawater, from a diatom culture and from an aged plankton tow (Turley *et al.*, 1988; Jenkinson *et al.*, 1988). Preliminary results are shown in Table 2. They indicate that the mean yield stress (physical strength) of the aggregates varied from 15 Pa for the aggregates formed from an aged plankton tow to about 27 Pa for those formed from aged seawater or aged *Skeletonema* cultures. The critical shear of our laboratory aggregates, that is the mean amount of shear they suffered before yielding, was 0.9 (aged plankton tow), 1.6 (*Skeletonema*) and 2.7 (aged seawater). The aged-seawater aggregates were thus the most "stretchy", and the plankton-tow aggregates the most brittle. That our aggregates stretched before yielding allows an estimate of complex modulus (yield stress/critical shear) to be determined. Based on mean values, this is 18, 17 and 10 Pa respectively, for the plankton-tow, *Skeletonema*-culture and aged-seawater aggregates. Yield consisted of fracture in some laboratory aggregates but of plastic flow in others. We are preparing a more detailed presentation of these results and of the methods used. As in Alldredge *et al.*'s aggregates, strength in our laboratory aggregates was inversely related to size.

That the measured yield stress of our laboratory-made aggregates was much greater than in those investigated *in situ* by Alldredge *et al.* (1990) may be partly because our aggregates were smaller than those of Alldredge *et al.*, and partly because laboratory aggregates are more compact than those formed *in situ* (Logan and Wilkinson, 1990). However the deformation forces to which the aggregates were subject are not strictly comparable, as they were turbulent in the study of Alldredge *et al.*, but produced by a steady imposed shear rate in ours. Furthermore breaking strain was calculated by Alldredge *et al.* as that needed to break the aggregates once, and was thus probably the strength of a line of weakness, while in our study the material of the aggregate was almost certainly sheared more homogeneously, and was also compressed in the measurement gap.

INFLUENCE OF ORGANIC AGGREGATE RHEOLOGY ON TURBULENCE

Polymeric thickening dispersed homogeneously has been shown theoretically to damp turbulence by increasing the size of the smallest dissipation structures, Kolmogorov eddies (Jenkinson, 1986). When aggregates are present two further mechanisms of turbulence reduction are proposed.

First mechanism

As long as aggregates remain strong enough to withstand the surrounding shearing forces, any deformation in the aggregate will be elastic and thus non-permanent. In this way aggregates represent turbulence-free zones in a turbulent sea, for as long as the aggregate lasts. Lazier and Mann (1989) have shown theoretically that elimination of turbulent diffusion can markedly enhance microzones. This is confirmed by the production, in some phytoplankton-produced aggregates, of microzones of increased pH, which promote fixation and accumulation of manganese (Lubbers *et al.*, 1988). In other aggregates O₂-deficient microzones are produced (Alldredge and Cohen, 1987). Paerl *et al.* (1987) have suggested that N₂-fixation by anaerobic bacteria in pelagic systems is mediated by the availability inside particles of such O₂-deficient microzones.

Second mechanism

If turbulent energy were perfectly transmitted or reflected by suspended aggregates, the cascade of turbulence would not be affected. Since aggregates are viscous as well as elastic, however, they tend to wobble in the turbulent field, and such viscoelastic deformation absorbs energy. Turbulent energy will therefore be correspondingly reduced in the surrounding fluid.

In a liquid without elastic properties, a relationship is implicit between turbulent dispersion of material and turbulent dissipation of energy. The presence of viscoelastic aggregates is likely to decouple this relationship (Jenkinson, 1990).

INFLUENCE OF ORGANIC AGGREGATE RHEOLOGY ON VERTICAL ORGANIC FLUX

Vertical flux of organic matter is largely mediated by organic aggregates (Fowler and Knauer, 1986; Alldredge and Silver, 1988). The sinking speed through still water is determined primarily by aggregate size and excess density, although shape is important at Reynold's numbers > 1. As the size, and possibly the shape of aggregates are partially determined by their rheological properties (Jenkinson, 1990; Turley, 1991), data on aggregate rheology are required for modelling flux.

ORGANIC SUB-MICROMETRE PARTICLES

In addition to the organic aggregates ≥ 30 µm to 1 m so far discussed, Koike *et al.* (1990), using a low-disruption, high-resolution particle counter, found up to ≈ 10⁷ organic particles ml⁻¹ in surface waters of the northern Pacific Ocean. These particles are of equivalent diameter between 0.38 and 1 µm and if they are compact in shape their calculated volume fraction does not exceed ≈ 0.000 1 %. Their destruction by brief sonication indicates that they are fragile; electron microscopy of water samples containing these sub-micrometre particles suggests that most of them are non-living, macromolecular aggregates; and their ability to pass through filters of pore size about one quarter of their measured equivalent diameter indicates that they are flexible and perhaps of string-like shape, (Koike *et al.*, 1990; Toggweiler, 1990).

The size of sub-micrometre particles suggests that they must have a low vertical sinking or rising speed. As pointed out by Koike *et al.* (1990), they thus cannot contribute significantly to vertical flux unless they associate with each other or with larger particles. Irrespective of whether they constitute "picoaggregates" or even single macromolecules > 10⁵ daltons, as suggested by Toggweiler (1990), if they include components sufficiently long to entangle with or adhere to adjacent particles, they could loosely aggregate to form more extensive zones of 3-dimensional gel matrix. Bulk rheological properties would then be modified up to the length-scales of such zones of gel. Exopolymers derived from cultured microalgae have already been found by Ramus and Kenney (1989) to constitute "very complex macromolecular aggregations", which can further flocculate to form a gel matrix.

CONCLUSIONS

The rheological properties of aggregates partially determine, firstly their size, shape and sinking speed, and secondly extraction of turbulent energy from interaggregate water. In addition, their volume fraction, their shape and their surface properties will determine the degree to which they may adhere or intertwine to form three-dimensional matrices.

Data on the rheological properties of marine organic aggregates, as well as on the nature, shape and surface properties of organic sub-micrometre particles are required for modelling both vertical organic flux and small-scale turbulence in the sea. These data would be particularly welcome for coastal waters, which appear generally to include those most rich in aggregates.

In addition to pure rheological data, information is needed about the flow of water in small (perhaps 1 to 100 cm³), unbounded zones *in situ*. Such data would provide further insight into rheological and other heterogeneity in seawater, and might shed more light on turbulent processes.

Acknowledgements

Ian Jenkinson and Carol Turley gratefully acknowledge the hospitality and help received whilst working at the Alfred-Wegener-Institut, Bremerhaven. They were financed by Alfred-Wegener-Institut Gastwissenschaftler Fellowships.

We are grateful to an anonymous referee for detailed help and suggestions. This work is contribution n° 339 from the Alfred-Wegener-Institut and n° 14 from the Biogeochemical Ocean Flux Study (BOFS).

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