EXPERIMENTING ON SETTLING VELOCITIES OF NEGATIVELY BUOYANT MICROPLASTICS

Liliya Khatmullina, Shirshov Institute of Oceanology Russian Academy of Science, Atlantic branch (ABIORAS), Russia
Igor Isachenko, ABIORAS, Russia
Elena Esiukova, ABIORAS, Russia
Irina Chibarenko, ABIORAS, Russia

liliakhatmullina@gmail.com

Presence of small plastic particles (< 5 mm), defined as microplastics, in the ocean and, especially, in coastal areas became evident in the last decade. From physical point of view, this fact indicates emergence of a new type of particles in the ocean. In contrast to the abundance of studies concerning sources, actual distribution and ecological effects of those particles, there are almost no detailed descriptions of physical mechanisms determining their distribution and behavior in the water column. Settling velocities of microplastics were measured in a series of experiments conducted in a 1-meter high glass tank filled with distilled water, in accordance with the typical methodology used in sedimentology. At first approximation, we supposed that the semi-empirical formulations developed for the natural sediments would be applicable to the microplastics. Results of preliminary experiments on microplastics of simple shapes justified this hypothesis. The majority of the implemented equations of settling velocity fitted well with the experimental data. Next step would be to test these formulations on the marine microplastic particles with greater variability in shapes. ***The research is supported by the Russian Science Foundation, project number 15-17-10020.***

Key words: microplastics, settling velocity

I. INTRODUCTION

Microplastics, defined as small plastic particles (< 5 mm), have been raised in popularity among the scientific society in recent years. Predominant amount of studies concentrate on the sources of microplastics, their actual concentration in the marine environment and ecotoxicity [see reviews by 1, 2, 3], while only a few describe marine microplastics from the physical point of view [4, 5, 6]. Better understanding of vertical transport of microplastics is fundamental for improving estimates of their concentration, size distribution, and dispersal in the world ocean [4, 5, 6, 7].

Once a plastic particle gets into the ocean, it either stays at the surface or subsurface (if it is less dense than the water), or it sinks (if it is denser than water and if it has overcame the surface tension). Reisser et al. [6] have experimentally revealed different relationships between the shape, size and rising velocity of buoyant microplastics. As for the plastics with negative buoyancy, we should firstly notice that, since any plastic particle in the ocean represents a substrate for the bacteria and algae growth, density of initially buoyant particle

could subsequently increase due to the biofilm formation and fouling so that it becomes dense enough to sink below the sea surface [7, 8]. To our knowledge, the fate of these fouled microplastics is unknown, as well as the settling process of initially non-buoyant particles in general.

Our study is focused on the sedimentation process of non-buoyant artificially made microplastics, which was studied through the series of laboratory experiments. We aim to test the applicability of existing semi-empirical formulations of settling velocity by comparing the theoretical predictions with the experimental data.

Theoretical assumptions

Particle sedimentation has been a subject of numerous investigations. Noticeable success was achieved in formulating the natural grains settling velocity [e.g. 9, 10, 11, 12, 13], which is regarded as one of the basic terms in sedimentology [14]. Terminal settling velocity (w_s) of a particle falling in the fluid implies the movement without acceleration and thus the balance between the gravitational force and hydrodynamical drag,

$$\frac{1}{2}C_d S w_s^2 = (\rho_s - \rho) g V, \tag{1}$$

where C_d denotes drag coefficient; ρ and ρ_s – fluid and particle density, respectively; g – acceleration due to gravity; S and V – the cross-sectional area and volume of the particle.
Giving the following equation of settling velocity for a spherical particle of diameter d,

$$w_s^2 = \frac{4}{3C_d}\Delta g d, \tag{2}$$

where $\Delta = \rho_s/\rho - 1$.

Well-known analytical solution for the settling velocity and drag coefficient of a perfect sphere was proposed by Stokes (1851) on the assumption of laminar flow,

$$w_s = \frac{1}{18}\frac{\Delta g d^2}{\nu}, \tag{3}$$

$$C_d = \frac{24}{\mathrm{Re}}, \tag{4}$$

where ν is kinematic viscosity of the fluid, and Re denotes Reynolds number, calculated as $\mathrm{Re} = w_s d/\nu$ and lower than 1 for laminar flow.

However, turning to the natural particles and microplastics, it becomes challenging to acquire a fully analytical settling velocity equation due to the not constrained regime and complexity of particle forms. Spread of Reynolds values yielded by the small natural or microplastic particles implies that inside the selected size group (0.5-5 mm in particular) the settling process of particles represents laminar, transitional and turbulent regimes, which as such are distinct from physical point of view and could not be described by one general equation. The possible measure to combine these distinct behaviors in one formula is to use the function of two asymptotic solutions (for high and low Re), which was suggested by different researchers (enumerated in ref. [12]). Relationships of the settling velocity for the particles of different shapes are usually based on those for spheres with implementation of various coefficients accounting for the effects of the shape [11, 13, 16].

Dietrich [17] did a comprehensive work concerning the settling velocity of sediment particles based on an extensive dataset. His approach had no explicit relation to the regime of the flow and was basically an attempt to fit the data (with broad range of Re numbers and shapes) to the 4-order polynomial function in terms of dimensionless velocity (W_*) and dimensionless diameter (D_*) calculated as follows,

$$W_* = \frac{\rho w_s^3}{(\rho_s - \rho) g \nu}\ , \tag{5}$$

$$D_* = \frac{(\rho_s - \rho) g D_n^3}{\rho \nu^2}, \tag{6}$$

where D_n is nominal diameter of the particle, equal to the diameter of a sphere of the same volume as the particle [14].

His approximations account for the roundness and shape of a particle, expressed by Powers [18] roundness coefficient (*P*) and Corey [19] shape factor (*csf*), calculated as follows,

$$csf = \frac{c}{\sqrt{ab}}\ , \tag{7}$$

where *a*, *b*, and *c* are the longest, intermediate, and shortest axes of the particle, respectively.

Dietrich's formulas (for exact equations and coefficients see [17]) could be regarded as a continuous representation of the data set he employed, thus they were consequently used by other researchers for calibration of their semi-empirical equations and data comparison [11, 13, 20].

Cheng [9] generalized previously proposed semi-empirical equations in a simplified relation between the drag coefficient and the Reynolds number, using the aforementioned "two-asymptotes approach",

$$C_d = \left[(\frac{A}{\mathrm{Re}})^{1/m} + B^{1/m} \right]^m, \tag{8}$$

where *A*, *B*, and *m* denote semi-empirical coefficients, which account for the shape and are calibrated using the experimental data. For example, a spherical particle yields *A*=24, *B*=0.4, *m*=2 [11]. Therefore, in the laminar flow regime (Re<1) $C_d \rightarrow 24/\mathrm{Re}$ in accordance with (4), and in the developed turbulent flow regime (Re>10^5) C_d approaches a constant value, *B*.

Using (8), terminal settling velocity is expressed as follows,

$$w_s = \frac{\nu}{d} \left[\sqrt{\frac{1}{4} (\frac{A}{B})^{2/n} + (\frac{4}{3} \frac{d_*^3}{B})^{1/n}} - \frac{1}{2} (\frac{A}{B})^{1/n} \right]^n . \tag{9}$$

Zhiyao et al. [12 and references therein] expressed settling velocity equations from 19 studies in the notation which is idem or close to the Cheng's formulation (8) to compare the resulting *A*, *B*, and *m* coefficients and to propose their own expression. Our experimental microplastics settling velocities were compared to some of those predictions. Camenen [11] *enhanced (8)* and, instead of constant coefficients, proposed a set of additional equations for *A*, *B*, and *m*, which implement *csf* shape factor and *P* roundness. Ahrens [10] suggested another asymptotic approach (10), which was calibrated on natural sand grains and thus could account for the effects of angularity, without, however, defining the relation to the shape explicitly,

$$w_s = \frac{C_1 \Delta g d^2}{\nu} + C_t \sqrt{\Delta g d}\ , \tag{10}$$

where the first and second terms are associated with laminar and turbulent flow regimes, respectively. The coefficients C_1 and C_t were calibrated [10] using the data on natural sediments settling.

II. MATERIALS AND METHODS

To date, there is no common definition of microplastic size [1, 3]. In this study, we focused on the plastic particles within the 0.5-5mm size range, upper limit is set in accordance with majority of publications [2, 3, 21], and the lower limit is conditioned by the employed experimenting facility. It is noteworthy that the settling characteristics of smaller particles could be fundamentally different owing to their Reynolds numbers.

Sample collections

Microplastic particles were prepared by hand from the Polycaprolactone (PCL) plastic (measured density ρ_s = 1.131±0.005 g cm^{-3}), which softens during heating (up to 60˚C) and stiffens in room temperature, and thus allows to produce granules of different shapes and sizes. We concentrated on two groups of artificially made plastic particles, which have not been influenced by any natural factors (e.g. sun radiation, bio-fouling, etc.): a) quasi-spherical particles fabricated from PCL; b) cylinder-shaped granules with equal height and diameter cut from circular rods of PCL (further referred as PCL cylinders). We aimed to make the rods with equal diameter along their length ranging from 0.5 to 5 mm. The obtained rods were cut into segments in such a way that the length of resulting cylinder would equal its diameter, giving length to diameter ratio of around unity.

Experimental facility and procedure

Our method of settling velocity measurement is consistent with the experiments described by Ref. [22, 23, 24]. We used a glass column 10 mm thick, of square section (180x180) mm^2 (inner size), and height of 110 cm. Marking lines were painted over the tank 11 cm from the top, and 12 cm from the bottom, giving 87 cm of working distance. The depth of the upper marking line was determined in such a way that the particles would already achieve the terminal settling velocity (no acceleration) by the time of crossing the mark.

The tank was filled with distilled water. Air temperature outside the tank and temperature of water near the surface and at the bottom of the tank was measured before and after each series of drop experiments to confirm the relative thermal stability (±0.5˚C) and thus the absence of significant convection in the water during the experiment.

Obvious inaccuracy in respect to the fabrication process was anticipated, thus, the diameter and length of each PCL cylinder and diameters of quasi-spherical particles were measured beforehand. Particles were placed approximately 1 cm below the surface of water (so that particles are not restrained by surface tension) in the center of the tank by forceps, and then let go of without inducing any additional acceleration. Finally, the time a particle took to cover the working distance (87 cm) was measured with a stopwatch and terminal settling velocity was calculated as a ratio of working distance to the duration of fall. Experimental results were presented in terms of dimensionless settling velocity and dimensionless diameter according to (5, 6) and compared to the number of semi-empirical predictions. Goodness of fit was estimated by calculating average value of the relative error defined as

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{W_s(\text{predicted})_i}{W_s(\text{experimental})_i} - 1\right|, \tag{11}$$

where N denotes the number of individual measurements.

III. SPHERICAL PARTICLES

Spherical particles were relatively well-studied in a vast number of works [9, 11, 16 and references therein]. Obviously, physical processes related to the settling of plastic spheres could not be fundamentally different from those of spheres of any other material. Thus, the main aim of drop experiment with PCL spheres was to test the experimental facility by comparing obtained data with the already existing datasets. However, owing to the peculiarities of fabrication process (particles were made by hand), it was unattainable to produce ideal spheres of different diameters from PCL, but rather ellipsoids, which could be seen from the resulting histogram of *csf* (Fig.1). The values of *csf* (0.8-1) were close to but not equal unity, which is characteristic of ideal spheres. Nevertheless, we assume that this discrepancy is minor and the intended test on our PCL spherical particles is credible.

Dietrich's [17] curves could be regarded as a compact representation of a wide set of experimental data, analyzed in his study. Thus, by comparing our data to the Dietrich's approximation for smooth spheres, we analyze their consistency to the previously obtained dataset and consequently the validity of employed experimental procedure (Fig.1).

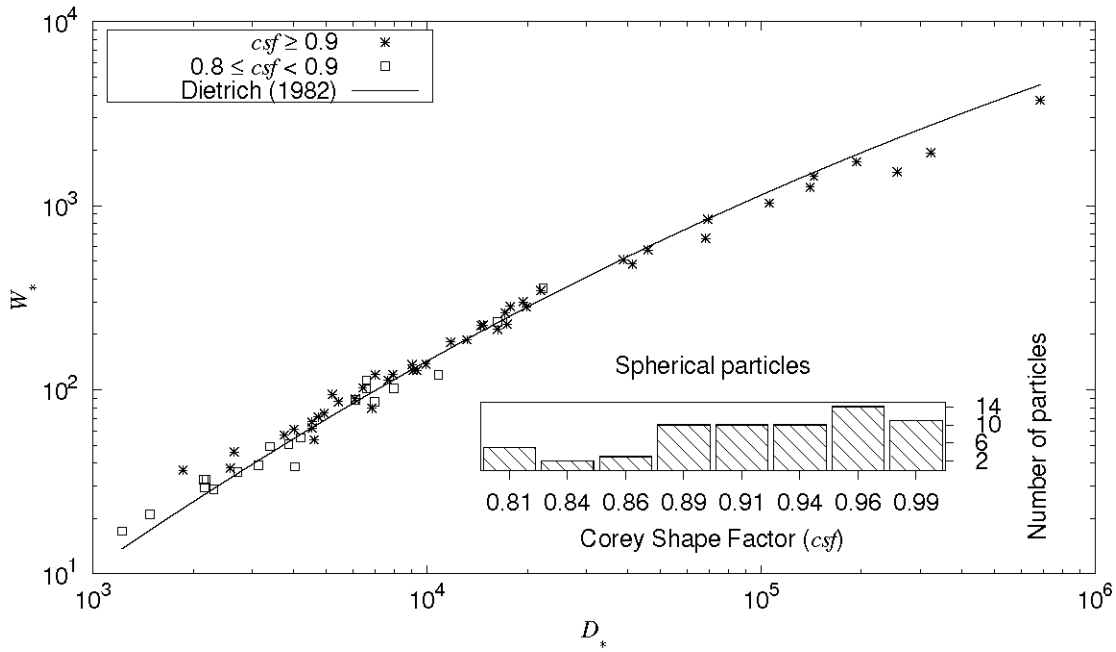

Figure 1. Spherical PCL particles. Corey shape factor (csf) and settling velocity of particles within two csf-groups (labels according to the legend). Curve is a fourth order polynomial fit for smooth spheres, given by Dietrich [17].

Figure 1 shows that although smaller particles are commonly less equal to spheres (*csf*=0.8-0.9), they correspond well with the approximation curve. For bigger particles, which on the contrary have relatively better concurrence to the spherical form, this minor divergence from sphericity results in slightly lower settling velocities compared to those of perfect spheres. Overall, experimental data is in good agreement with Dietrich's results, with

relative error of 4% (Table 1). This is in accordance with [11, 13, 24], who stated that the effect of shape is more pronounced for bigger particles/higher Reynolds numbers.

IV. CYLINDER-SHAPED PARTICLES

Drop experiments with PCL cylinders revealed the presence of various secondary movements during the particle sedimentation in water. Surface curvature of non-spherical particles induces flow separation, resulting in higher drag coefficient, and makes the settling more instable with occurrence of rotation, oscillation and tumbling. As noticed by [17] and [24], this effect becomes more pronounced with higher Reynolds numbers. Additionally, during falling PCL cylinders did not always follow a rectilinear trajectory. Hazzab et al. [17] suggested that the frequency of these secondary movements is attenuated with the increase in Reynolds numbers. In this study, we did not analyze such characteristics quantitatively, which, however, would be of interest in the future estimations on real marine microplastics.

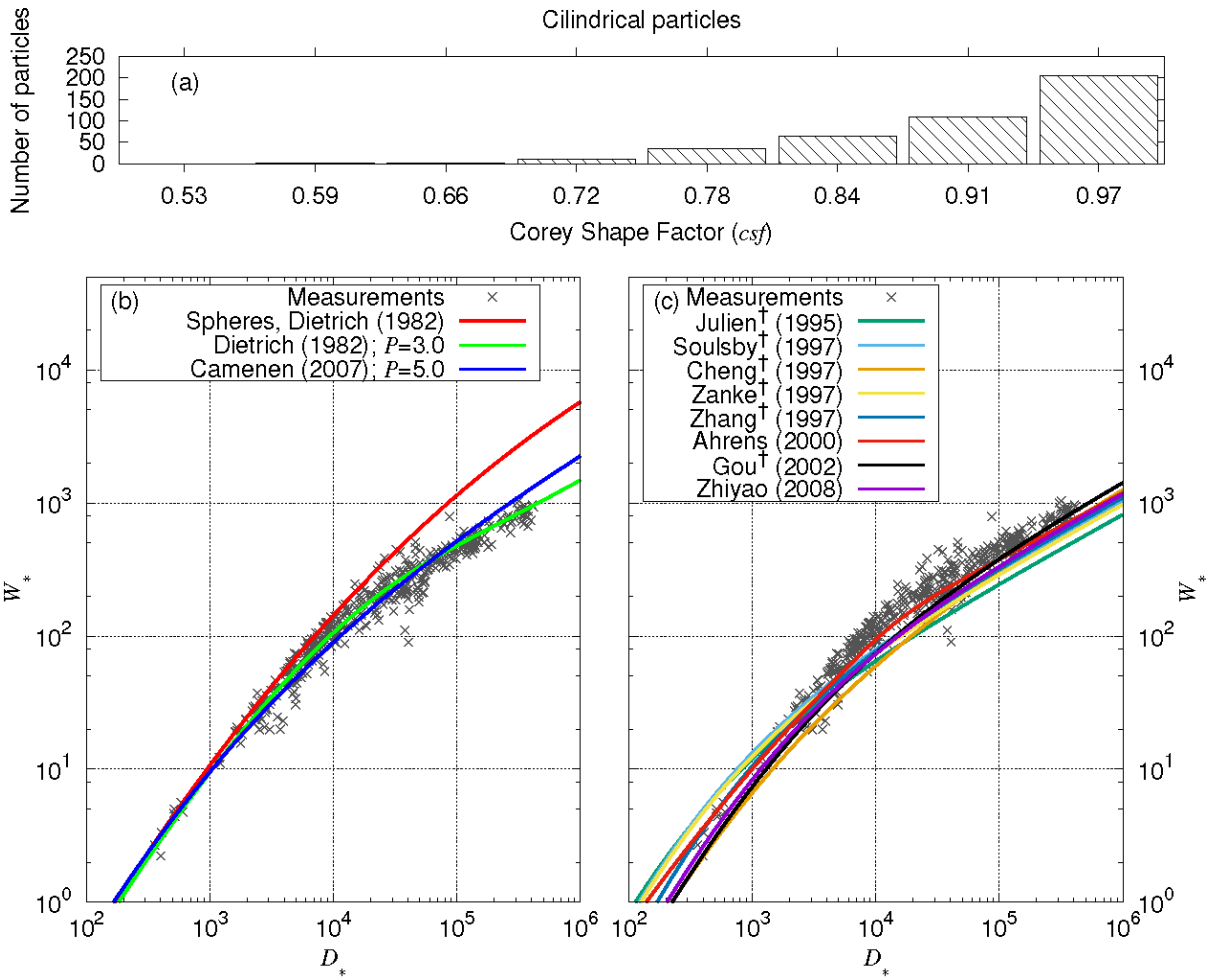

Figure 2. Cylinder-shaped particles. a) csf histogram, and b), c) experimental and predicted settling velocities in dimensionless terms (5, 6). Grey crosses represent experimental data; lines show the semi-empirical curves from different publications, colored according to the legend. † marks publications, which were used in comparative analysis by Zhiyao et al. [12] and are not directly referenced in this paper

We compared the experimental data with several existing semi-empirical formulations calibrated by using data on settling velocities of angular or naturally shaped grains (Fig.2, Table 1). Figure 2b represents two equations: Dietrich's [17] approximation of naturally shaped grains, and Camenen [11] formulation, – that explicitly account for particle shape and roundness. Both approaches involve defining *csf*, which formula (7) is comprehensible (for PCL cylinders we used mode value of *csf*=0.97, Fig. 2 a), and Powers roundness, *P*, which estimation is on the contrary rather subjective and could vary by around ±1 between two observers of the same grain [17]. Herein, we have not assigned the *P* value to our particles, but have selected the best curve fitting to our data points by changing *P* with 0.5 step as it is unrealistic to estimate *P* with higher resolution. Interestingly, the resulting *P* value was different for Camenen (5.0) and Dietrich (3.0) predictions, despite the fact that Camenen inter alia calibrated his formula on the Dietrich's approximation (and so it was more logical if roundness would be identical). Therefore, although these two formulas have in general better predicting ability as they directly consider the effect of shape and fit the data well (relative error of 7.4 and 5.3% for Camenen and Dietrich, respectively), the lack of clarity in roundness estimation reduces the applicability of these approaches. This problem becomes more striking considering the microplastic particles, which forms could be far from those of naturally occurring grains, thus indicating a need of development of a more quantitative approach to define roundness. Ref. [13, 20] also noticed the complexity of Dietrich's formulas and the fact that Powers roundness factor is rarely measured in practice.

Dietrich approximation of smooth spheres is also plotted in Fig.2b. It could be seen that starting from $D_* \approx 10^4$, which corresponds to Re≈100, settling velocity of angular particles noticeably diverges from that of spheres in agreement with the aforementioned statement that angularity of shape increases the drag and reduces the settling velocity for the same D_* as a sphere.

Table 1. Accuracy of fit of several existing formulations against the experimental data. Average value of the relative error, *E*, was calculated by (11). † marks publications, which were used in comparative analysis by Zhiyao et al. [12] and are not directly referenced in this paper

Experimental dataset	**Authors**	**E**
Spherical particles	Dietrich, 1982	4.0%
Cylinder-shaped particles	Dietrich (1982); csf=0.97, P=3.0	5.3%
	Camenen (2007); csf=0.97, P=5.0	7.4%
	Julien† (1995)	15.2%
	Soulsby† (1997)	10.4%
	Cheng† (1997)	14.5%

	Zanke† (1997)	11.7%
	Zhang† (1997)	10.2%
	Ahrens (2000)	6.6%
	Gou† (2002)	9.5%
	Zhiyao et. al (2008)	10.6%

In general, all the other settling velocity predictions used for comparison in this study (Fig.2c), in which shape is incorporated in general by a set of coefficients that does not vary, were able to predict the experimental values with a good degree of consistency (within 15.2% of relative error). Among them Ahrens curve represented the best fit to data (6.6% error, Table 1).

The studied group of particles is characterized by a quite simple form, which is comparable to that of the natural sediments. However, as reported by some authors [25], significant part of the microplastics found in sediments is devoted to fibers. Settling process of such specific particles will be the scope of future investigations.

V. CONCLUSIONS

In this study, we made an attempt to incorporate microplastics as a distinct type of particles in the already existing body of sedimentology research. Terminal settling velocity, being a major sedimentology term, provides not only a fundamental view of the matter sinking in the ocean, but in our case can consequently shed light on the problem of microplastic transportation/distribution in the ocean and uncover why and how ocean sediments act as a sink of microplastics [1, 25]. In a set of experiments on the microplastic particles of simple forms, we have successfully tested the experimental facility and procedure, which could be further used in the experiments on real marine microplastic.

Measured settling velocity was in accordance with available experimental data. When applied to microplastics, the existing semi-empirical models predict the settling velocity of spherical and cylinder-shaped particles in the intermediate range of Reynolds numbers quite accurately. We pointed the potential of Dietrich and Camenen approximations to estimate the settling velocity of microplastics of different shapes with a good degree of consistency on a stipulation that the roundness coefficient would be calculated in a quantitative way.

In accordance with several existing studies, shape of a particle was found to be the major term in determining the character of settling and corresponding settling velocity, which is of great concern for marine microplastics which represents variety of forms.

VI. REFERENCES

[1] A.L. Andrady, “Microplastics in the marine environment,” *Marine pollution bulletin*, vol. 62, pp. 1596-1605, 2011.

[2] M. Cole, P. Lindeque, C. Halsband, C., and T.S. Galloway, “Microplastics as contaminants in the marine environment: a review,” *Marine pollution bulletin*, vol. 62, pp. 2588-2597, 2011.

[3] R. Thompson, “Microplastics in the Marine Environment: Sources, Consequences and Solutions” in *Marine Anthropogenic Litter* , M. Bergmann, L. Gutow, and M. Klages, Eds. Springer International Publishing, pp. 197-212, 2015.

[4] T. Kukulka, G. Proskurowski, S. Morét-Ferguson, D.W. Meyer, and K.L. Law, “The effect of wind mixing on the vertical distribution of buoyant plastic debris,” *Geophysical Research Letters*, vol. 39, 2012.

[5] A. Isobe, K. Kubo, Y. Tamura, E. Nakashima, and N. Fujii, “Selective transport of microplastics and mesoplastics by drifting in coastal waters,” *Marine pollution bulletin*, vol. 89, pp. 324-330, 2014.

[6] J. Reisser, B. Slat, K. Noble, K. du Plessis, M. Epp, M. Proietti, et al., “The vertical distribution of buoyant plastics at sea,” *Biogeosciences Discuss.*, vol. 11, pp. 16207-16226, 2014.

[7] K.L. Law, S.E. Morét-Ferguson, D.S. Goodwin, E.R., Zettler, E. DeForce, et al., “Distribution of surface plastic debris in the eastern Pacific Ocean from an 11-year data set,” *Environmental science and technology*, vol. 48(9), pp. 4732-4738, 2014.

[8] F.M. Fazey and P.G. Ryan, “Biofouling on buoyant marine plastics: An experimental study into the effect of size on surface longevity,” *Environmental Pollution*, vol. 210, pp. 354-360, 2016.

[9] N.S. Cheng, “Simplified settling velocity formula for sediment particle,” *Journal of Hydraulic Engineering*, vol.123(2), pp. 149–152, 1997.

[10] J. P. Ahrens, “A fall-velocity equation,” *Journal of Waterway, Port, Coastal, and Ocean Engineering*, vol. 126(2), pp. 99–102, 2000.

[11] B. Camenen, “Simple and general formula for the settling velocity of particles,” *Journal of Hydraulic Engineering*, vol.133, pp. 229-233, 2007.

[12] S. Zhiyao, W. Tingting, X. Fumin, and L. Ruijie, “A simple formula for predicting settling velocity of sediment particles,” *Water Science and Engineering*, vol. 1(1), pp. 37-43, 2008.

[13] J.A. Jimenez, and O.S. Madsen, “A simple formula to estimate settling velocity of natural sediments,” *Journal of Waterway, Port, Coastal, and Ocean Engineering*, vol. 129(2), pp. 70–78, 2003.

[14] Interagency Committee, “Some fundamentals of particle size analysis: A study of methods used in measurement and analysis of sediment loads in streams,” Subcommittee on Sedimentation, Interagency Committee on Water Resources, St. Anthony Falls Hydraulic Laboratory, Minneapolis, Rep. No. 12, 1957.

[15] G. Stokes, “On the effect of internal friction of fluids on the motion of pendulums,” *Trans. Cambridge Philos. Soc.*, vol. 9, pp. 8–106, 1851.

[16] A. Terfous, A. Hazzab, and A. Ghenaim, “Predicting the drag coefficient and settling velocity of spherical particles,” *Powder technology*, vol. 239, pp. 12-20, 2013.

[17] W.E. Dietrich, “Settling velocity of natural particles,” *Water resources research*, vol. *18*(6), pp.1615-1626, 1982.

[18] M.C. Powers, “A new roundness scale for sedimentary particles,” *Journal on Sedimentary Petrolology*, vol. 32, pp. 117-119, 1953.

[19] A. Corey, “Influence of the shape on the fall velocity of sand grains,” Master’s thesis, Colorado A&M College, Fort Collins, Colo, 1949.

[20] W. Wu, and S.S. Wang, “Formulas for sediment porosity and settling velocity,” *Journal of Hydraulic Engineering*, vol. 132(8), pp. 858-862, 2006.

[21] C. Arthur, J. Baker, H. Bamford, “NOAA Technical Memorandum NOS-OR&R30,” *Proceedings of the international research workshop on the occurrence, effects and fate of microplastic marine debris*, September 2008.

[22] R.J. Gibbs, M.D. Matthews, and D.A. Link, “The relationship between sphere size and settling velocity,” *Journal of Sedimentary Research*, vol. *41*(1), pp. 7-18, 1971

[23] J. R. L. Allen, “Physical sedimentology”, Allen & Unwin, 1985

[24] A. Hazzab, A. Terfous, and A. Ghenaim, “Measurement and modeling of the settling velocity of isometric particles”, *Powder Technology*, vol. 184(1), pp. 105-113, 2008

[25] L.C. Woodall, A. Sanchez-Vidal, M. Canals, G.L. Paterson, R. Coppock, et al., “The deep sea is a major sink for microplastic debris,” *Royal Society open science*, vol. 1(4), 2014.