Observation, analysis and modelling in complex fluid media

Settling classes for fine suspended particles

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Available online 8 September 2006

Abstract

The modelling of pollutant transfer in freshwater systems can be refined by considering the heterogeneities of the sedimentary dynamics and of the chemical reactivities of fine suspended particles. One of the first steps is the fractionation of these fine particles into effective settling classes. Although several methods exist, most of them are based on either granulometric considerations and/or arbitrary threshold criteria. This article presents the bases of an experimental method focusing on the direct measurement of the settling velocities without considering the granulometry and/nor any threshold criteria. The experimental work consists in recording the temporal evolution of the vertical distribution of the suspended solid concentration in a settling tank. A mathematical analysis provides the number of particle groups, and the mass contribution and the settling velocity for each This procedure is described and applied for validation, as a first step, to calibrated suspensions. Additional work is needed for a further analysis of the physical constraints involved in the model, as well as for more extensive experimental validation. ***To cite this article: C. Brach-Papa et al., C. R. Mecanique 334 (2006).***

Résumé

Classes de décantation des particules fines en suspension. La modélisation des transferts de polluants dans les hydrosystèmes peut être affinée en considérant l'hétérogénéité de la dynamique sédimentaire et des réactivités chimiques des fines particules solides en suspension. Pour celà, l'une des premières étapes est de fractionner ces fines particules en classes de vitesses de chute effectives. Bien qu'il existe déjà plusieurs méthodes, celles-ci sont plus ou moins basées sur des considérations granulomètriques et/ou des critères de seuils arbitraires. Ce papier présente les bases d'un protocole expérimental dédié à la mesure directe de ces vitesses de chute sans considération granulométrique à priori ni critère seuil. Une analyse mathématique des évolutions temporelles de la distribution verticale de concentration d'une suspension solide en colonne de décantation permet la détermination directe du nombre de groupes et, pour chacun, de sa contribution massique et de sa vitesse de chute. La procédure est décrite et appliquée pour validation, en première étape, à des suspensions calibrées. La démarche devra être approfondie afin de mieux analyser les contraintes inhérentes au modèle, et aussi conduire des validations expérimentales plus complètes. ***Pour citer cet article : C. Brach-Papa et al., C. R. Mecanique 334 (2006).***

Keywords: Fluid mechanics; Polydisperse suspension; Settling velocity; Fine particles

Mots-clés : Mécanique des fluides ; Matières en suspension ; Vitesse de chute ; Particules fines

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1. Introduction

In freshwater systems, the transfer of pollutants (e.g., radionuclides, heavy metals) depends on the behaviour of the sediment particles in terms of dispersion, sorption, deposition, erosion, . . . In these systems, the natural suspended material is heterogeneous. The modelling of transfers of these particles and their associated pollutants can be refined coupling hydro-sedimentary and chemical dynamics using a multi-class approach [1]. The finest particles mainly consist of mineral elements containing aluminosilicates with large specific surfaces (10–100 $m^2 g^{-1}$). They have cohesive properties and a high sorption rate towards organic and inorganic xenobiotics. The largest particles are made of detrical materials containing quartz with low specific surfaces ($<10^{-3}$ $m^2 g^{-1}$), and lower reactivity with pollutants. Consequently, the modelling of the transfer of pollutants mainly needs to consider the chemical and sedimentary behaviours of the fine particles. One of the first steps is to characterize the kinetics of the clearance rate over a calm water column in terms of effective settling velocities for each group of particle. This parameter is important because it includes different characteristics, such as the size, the shape and the density of particles, which affects their transfers and those of their associated pollutants.

Two families of methods exist. On the one hand settling velocities are deduced from the granulometric distributions, using the Stokes formula for example. The most usual method, called the Atterberg procedure [2], uses arbitrary specific sedimentation times to isolate the finest particles (<50 μm). Finally, the use of laser diffraction is also popular: it is based on the measurement of the diffusion/refraction angles for light when a particle crosses the laser beam.

On the other hand, settling velocities can be directly measured in experimental settling columns or/and in situ. In-situ measurements are possible through the use of optical (turbidimeter) and acoustical (Doppler velocimetry) probes or high-resolution cameras directly immerged within the studied medium [3]. Optical or acoustical probes allow monitoring the evolution in space and time of the concentration profiles, which are analysed to evaluate the effective settling velocities. High-resolution cameras provide simultaneous determination of the granulometric distribution, the shape of the particles, and their settling velocities. Nevertheless, the resolution of the cameras is still not sufficient for studying fine sediment [4].

In practice, these in-situ methods do not allow one to collect matter and perform physico-chemical analyses and to determine the associated pollutant loads. At this end of the particle size spectrum, experimental settling columns are more appropriate. Two main protocols are applied: one uses a floating layer resulting from a uniform injection of matter on the surface of the water column while the other one uses an initially homogeneous suspension load over the column. For the first one, settling velocities are obtained from the measurement of the deposited matter quantities as a function of time [5]. For the second one, settling velocities are estimated from the calibration of mathematical models on the evolution of the suspended load [6]. However, the major difficulty remains the consideration of appropriate sampling times.

Related to the latter family, the present article describes the protocol TALISMEN (French acronym for metal transfers at liquid/solid interfaces in natural water). The objectives are to validate a simple experimental protocol for characterizing the main settling classes of a heterogeneous suspension of fine particles without granulometric considerations and/or threshold criteria. The first part of the paper describes the method while the second part reports the results of its application to calibrated suspensions at the end of validation.

2. Experimental device

The experimental device consists of a 350 l glass column (1.5 m × 0.46 m × 0.46 m). The bottom is designed as a two-part sediment collector for the isolation and the extraction of the deposited matter. A mechanical stirrer (Milton Roy Serie VDI, 1 m shaft length, 80 mm propeller diameter) is used to homogenize the suspension before the settling experiments. Turbidity measurements are performed with two optical backscatter sensors (OBS-3, D&A Instruments) immersed at different depths and connected to a data acquisition system (Agilent 34970A) driven by the Benchlink data logger software (Agilent). 1 l water samples at the same depths as the turbidimeters are taken using a peristaltic pump (Masterflex serie 323). They are used for gravimetric and granulometric analyses, which allow the determination of particle sizes ranging from 0.05 to 900 μm using a laser diffraction analyser (Master sizer S, MALVERN).

Initially, the homogeneous suspended solid concentration conditions are obtained by turbulent mixing. When the turbidity signals, obtained at different depths in the tank, stay constant and equal, the mechanical stirrer is stopped. The settling phase begins at this time, t_0. The turbidity values at t_0 are used as references for normalizing the turbidity signals. The settling process is investigated until the turbidity signals have decreased to a relatively constant value. During the settling period, about ten water samples are taken at regular intervals. Gravimetric analyses allows conversion of the turbidity signals from NTU to $kg\,m^{-3}$, while granulometric analyses allow comparisons with the effective settling velocities obtained using the methods described below.

3. Mathematical analysis

The settling groups and their effective settling velocities are deduced from a two-step mathematical analysis of the temporal evolution of the suspended solid concentration recorded during settling. The first step provides the number of groups, their contributions to the total suspended solid concentration and their decay constants at the measurement depth. The second step gives their effective settling velocities, corresponding to the values averaged over the water column height.

3.1. Step 1

As suggested by several authors, the evolution of suspended solid concentration during a settling experiment can be assumed to be a first-order decay process [7]. Consequently, the evolution of the suspended solid concentration can be mathematically expressed as the sum of several first-order exponential decays. With the hypothesis that each term corresponds to a group of settling particles, this property allows one to obtain the number of settling groups, their specific contributions and their effective settling velocities.

$$SS_z^t \approx \sum_i SS_{z,i}^0 \cdot \exp\bigl(-\lambda_{z,i} \cdot (t - t_0)\bigr) \tag{1}$$

where t is time, z is the depth, SS_z^t is the total suspended solid concentration, $SS_{z,i}^0$ is the initial concentration of the group i at t_0 and $\lambda_{z,i}$ is the decay rate of the group i at the depth z.

The recorded signals are normalized by SS^0, the initial total suspended solid concentration obtained at the end of the mixing phase (at t_0):

$$\frac{SS_z^t}{SS^0} \approx C_z^t = \sum_i C_i^0 \cdot \exp\bigl(-\lambda_{z,i} \cdot (t - t_0)\bigr) = \sum_i C_{z,i}^t \tag{2}$$

In this equation, C_z^t is the normalized total concentration and C_i^0 and $C_{z,i}^t$ are respectively the initial and the instantaneous contributions of the group i.

The optimal coefficient set $\{C_i^0; \lambda_{z,i}\}$ is obtained decomposing the signal corresponding to SS_z^t/SS^0 with a mathematical fitting method based on the minimization of the sum of the squared residuals, SSR, between the experimental and adjusted data:

$$SSR = \sum_{t=0}^t \left(\frac{SS_z^t}{SS^0} - C_z^t\right)^2 \tag{3}$$

This decomposition is automated by the use of the 'Solver' function of the Microsoft-Excel software, which is commonly used for similar purpose. For a given number of groups, this function is applied to minimize SSR according to three constraints: (i) the initial contributions cannot be negative, $C_i^0 \geqslant 0$; (ii) the sum of the contributions is equal to one, $\sum C_i^0 = 1$; and (iii) the decay rates are all positive, $\lambda_{z,i} \geqslant 0$. The optimal number of settling particle groups is automatically determined considering that it is reached when SSR is not improved by the use of an additional group. To make sure that an absolute and not a local minimum is obtained, a procedure has been developed to run several times the 'Solver' function with different appropriate sets of initial values.

3.2. Step 2

At this step, the effective settling velocity of each of the groups is deduced from the temporal evolutions of the total contributions integrated on the water height h for each settling group i. These contributions are given by:

$$C_i^t = \frac{1}{h} \cdot \int_{z=0}^{z=h} C_{z,i}^t \cdot \mathrm{d}z \tag{4}$$

Their temporal evolutions are directly linked to the effective settling velocities, We_i^t, by:

$$\frac{\mathrm{d}C_i^t}{\mathrm{d}t} = -\frac{We_i^t}{h} \cdot C_i^t \tag{5}$$

According to Eqs. (2) and (4), one can then write:

$$We_i^t = h \cdot \int_{z=0}^{z=h} \lambda_{z,i} \cdot \exp(-\lambda_{z,i} \cdot (t - t_0)) \cdot \mathrm{d}z \Big/ \int_{z=0}^{z=h} \exp(-\lambda_{z,i} \cdot (t - t_0)) \cdot \mathrm{d}z \tag{6}$$

Eq. (6) can be solved if the vertical distribution of $\lambda_{z,i}$ is known. The experimental values (Fig. 1) show that this vertical distribution can be correctly approximated by a linear interpolation:

$$\lambda_{z,i} = a_i \cdot z + b_i \tag{7}$$

The parameters of these vertical distributions can be deduced after step 1, if the turbidimetric measurements are available for at least two depths in the settling tank. For each settling group, the effective settling velocity depends on time and on the water depth. These effective velocities are finally given by:

$$We_i^t = \frac{h}{t - t_0} - a_i \cdot h^2 \cdot \frac{\exp(-a_i \cdot h \cdot (t - t_0))}{1 - \exp(-a_i \cdot h \cdot (t - t_0))} + b_i \cdot h \tag{8}$$

This relation converges to $We_i^t = -a_i \cdot h^2 + b_i \cdot h$ when $t \to \infty$. Thus, the condition $We_i^t \geqslant 0$ is verified, whatever t, if $a_i < 0$ and $b_i > 0$.

To compare these effective settling velocities to the granulometric information associated with the particles or given by the laser diffraction analysis, it is necessary to relate these velocities to effective diameters. At this end, it is considered that the concentration ranges used during the experiments are low enough to not influence the effective settling velocities by concentration effects. For spherical particles of small size ($d < 100$ μm), the particle Reynolds number (Re), which is the ratio of the inertia and viscosity forces, is smaller than 1. The effective settling velocity of such particles can then be related to an effective diameter by Stokes' law.

$$Re = \frac{We \cdot de}{\nu} < 1 \quad \Rightarrow \quad We = \frac{g \cdot R \cdot de^2}{18 \cdot \nu} \tag{9}$$

For particles larger than about 100 μm ($Re > 1$), the following relation can be used [8]:

$$We = \frac{10 \cdot \nu}{de} \left[\left(1 + \frac{0.01 \cdot R \cdot g \cdot de^3}{\nu^2} \right)^{0.5} - 1 \right] \tag{10}$$

In these relations, g is the gravitational acceleration, de is the effective diameter, $R = (\rho_s - \rho_w)/\rho_w$ is the specific density of the particles, ν is the kinematic viscosity of water. ρ_s, the density of the particles, is that of quartz material: $\rho_s = 2650 \ \mathrm{kg\,m^{-3}}$.

4. Application to calibrated sand suspensions

In order to test the suitability of the approach to correctly identify and characterize the settling groups, the proposed procedure is tested on well-known calibrated sand fractions obtained by dry-sieving 'Fontainebleau sand' with 500, 200, 100 and 50 μm mesh size sieves. The first applications concern these individual sand fractions and the second ones are applied to a stoichiometric massic mixture of these sand fractions.
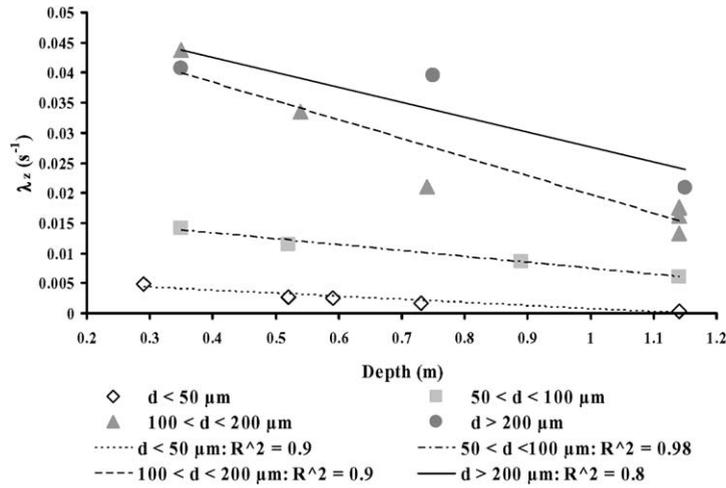
Fig. 1. $\lambda_{z,i}$ distribution for each sand fraction.

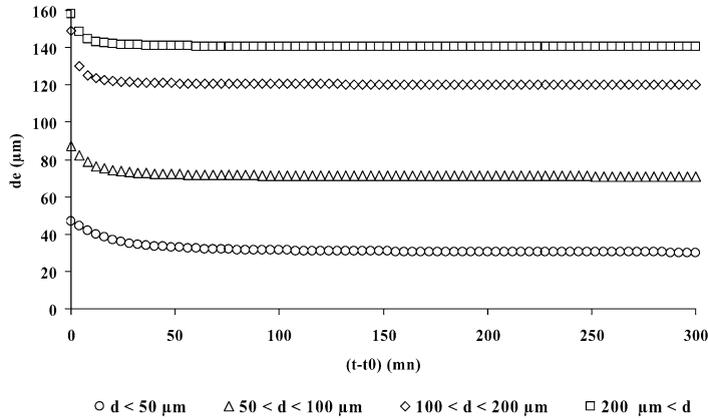Fig. 1. Distribution des $\lambda_{z,i}$ pour chaque fraction de sable.

Fig. 2. Temporal evolution of de for each fraction.

Fig. 2. Evolution temporelle des de pour chaque fraction.

4.1. Individual sand fractions

The experimental protocol is applied to 30 g of each individual sand fraction. The first objective is to validate the hypothesis of linear vertical variations of $\lambda_{z,i}$ and the second one is to test the ability of the protocol to find the granulometric characteristics of these sand fractions.

For the first objective, the turbidity measurements are performed for several depths in the settling tank. The $\lambda_{z,i}$ distributions obtained for each of the sand fractions are presented on Fig. 1. These results show that these distributions are nearly linear, with resulting determination coefficients (R^2) larger than 0.9 except for the coarsest sand fraction ($d > 200$ μm).

Fig. 2 presents the temporal evolutions of the effective diameters deduced from the distributions of $\lambda_{z,i}$. It can be seen than the effective diameters given by the method are well centred on the sand fraction characteristics, except, once again, for the coarsest sand fraction ($d > 200$ μm).

4.2. Stoichiometric massic mixture

For the stoichiometric massic mixture experiments, the water depth is $h = 1.3$ m. The protocol is applied to about 60 g of a mixture made of equal proportions in mass of the four calibrated sand fractions (Table 1).

Table 1
Sand mixture composition
Tableau 1
Composition du mélange de sable

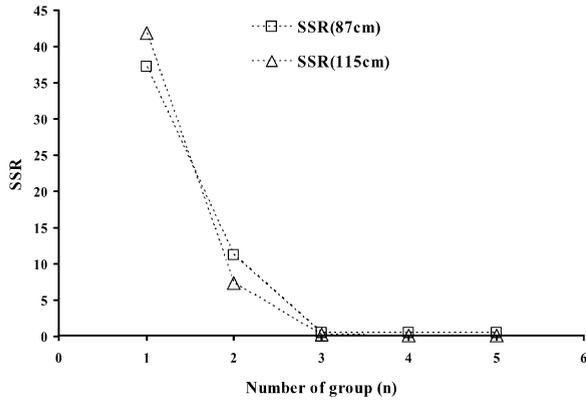|	$200 < d < 500$ (μm)	$100 < d < 200$ (μm)	$50 < d < 100$ (μm)	$d < 50$ (μm)
Masses (g)	15.6	15.9	15.8	16.2

Fig. 3. *SSR* evolutions according to n.

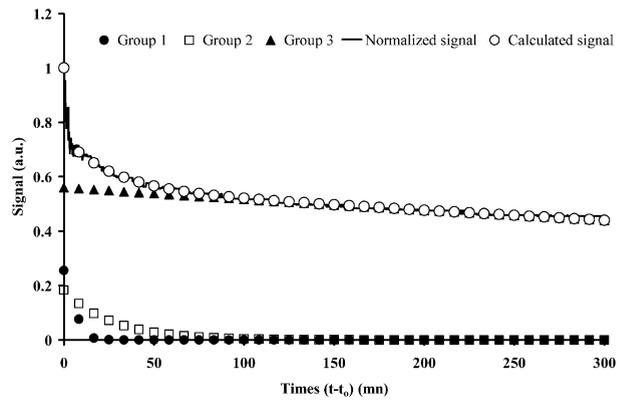Fig. 3. Evolution du *SSR* en fonction de n.

Fig. 4. Fitted signal at $z = 1.15$ m.

Fig. 4. Ajustement du signal à $z = 1.15$ m.

Table 2
Fitted parameters obtained with $n = 3$
Tableau 2
Paramètres d'ajustement pour $n = 3$

	Group 1	Group 2	Group 3
$\lambda_{87,i}$ (s^{-1})	1.74E–02	5.00E–04	2.40E–05
$\lambda_{115,I}$ (s^{-1})	1.09E–02	4.54E–04	9.47E–06
$C_{87,I}^0$ (–)	0.25	0.25	0.50
$C_{115,I}^0$ (–)	0.27	0.20	0.53
Average C_i^0 (–)	0.26	0.22	0.52
a_i (m^{-1} s^{-1})	−2.30E–02	−1.64E–04	−5.19E–05
b_i (s^{-1})	3.73E–02	6.42E–04	6.92E–05

The mathematical adjustment is applied, for a number of groups varying from $n = 1$ to $n = 5$, to the turbidimetric signals monitored during 300 mn with an acquisition frequency of 0.1 Hz at the depths 0.87 and 1.15 m. The evolutions, as a function of n, of the resulting sum of the squared residuals (*SSR*) at these depths are presented in Fig. 3. At each depth, the residues decrease with increasing number of groups up to $n = 3$. Beyond, additional settling groups do not improve the adjustment quality. These results suggest that even if four sieved sand fractions are used to compose the test mixture, this mixture can be decomposed into only three effective settling velocity groups. Fig. 4 presents, for $z = 1.15$ m, the superposition of the experimental signal with the fitted signal and the contributions of the three groups.

The contributions of each of the settling groups, deduced from the median C_i^0 calculated from the average of the values for $z = 1.15$ and 0.87 m, and the parameters of the linear vertical distribution of the $\lambda_{z,i}$ are presented in Table 2.

Firstly, it can be noted that the couples (a_i, b_i) respect the condition to have $We_i^t \geqslant 0$ whatever t (see Eq. (8)). The equivalent settling diameters are deduced with Eq. (10) for the group 1 and with Eq. (9) for the groups 2 and 3. During the settling experiments these diameters evolved from 147 to 85 μm for de_1^t and stay in the neighbourhood of 22 μm for de_2^t and of 5 μm for de_3^t.
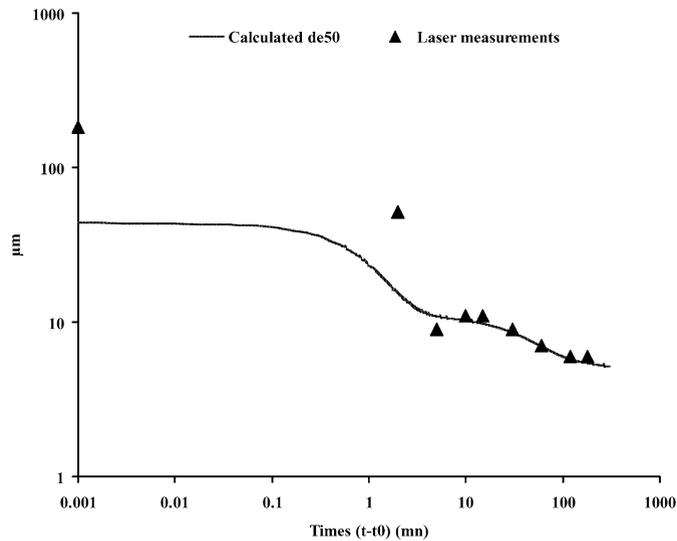
Fig. 5. Computed and measured temporal evolutions of the average diameter at $z = 0.87$ m.

Fig. 5. Evolutions temporelles calculée et mesurée du diamètre moyen à $z = 0.87$ m.

The temporal evolution of these diameters and of their group contributions allows one to deduce the average effective diameters, de_{50z}^{t}, at any time and for any depth within the settling tank using the following formula:

$$de_{50z}^{t} = \frac{\sum_i C_{z,i}^{t} \cdot de_{z,i}^{t}}{\sum_i C_{z,i}^{t}} \tag{11}$$

where $de_{z,i}^{t}$ are deduced from the application of Eq. (9) or (10) (the choice depends on the diameter range) to the effective settling velocities computed with Eq. (8).

Fig. 5 presents, for the depth $z = 0.87$ m, the temporal evolutions of the computed average effective diameters and of the d_{50} measured by laser diffraction from the water volumes sampled at this depth and for different times during the settling experiment.

This comparison shows the ability of the method to correctly represent the evolution of the average diameter except during the first minutes of the settling experimentation when the variations depend on the settling properties of the biggest particles. In the same time, as the general behaviour of the signal is relatively well represented by the mathematical analysis, it can be assumed that these limitations of the method for the biggest particles mainly result from the size of the settling device and the invalidity of the linear hypothesis for the vertical distribution of the $\lambda_{z,i}$ for these particles.

5. Discussion and conclusions

The article presents the bases of a method for the fractionation of a heterogeneous suspension of fine particles into groups of effective settling velocities. This fractionation is needed for the modelling of the transfers in hydrosystems of solid particles and their associated pollutants. The proposed method is based on the monitoring and a mathematical analysis of the vertical and temporal evolutions of turbidity signals. These latter are associated with suspended matter decanting in an experimental settling column. As compared to other approaches, the objectives of the presented method are to prevent from the need of some more or less arbitrary cut off based on the diameters and/or the settling times. The suspension is described in terms of massic contributions and effective settling velocities of the different particle groups. The velocities we obtain depend on time and on the water depth. This feature results from two main points: (i) the capacity of the approach to take into account the continuous heterogeneity characterizing a natural suspension; and (ii) within a given group, the granulometry varies as a function of time and depth, since the largest particles obviously settle faster.

Two kinds of tests are presented starting from four sand samples calibrated by sieving and characterized by laser diffraction granulometry. In the first set of experiments, the proposed methodology is applied to the fractions considered individually. Except for the coarsest sand fraction, the method allows to characterize correctly each of them. As a second test, the procedure is applied to a massic stoichiometric mixture of these four sand samples. Determining three effective settling particle groups, the method tends to confirm that sedimentation experiments are more appropriate than sieving ones to report correctly the settling behavior of a bulk suspension, particularly, when the aim is the modelling of the deposit flux. This point is confirmed by the comparison of the temporal variations of the d_{50} of the bulk suspension measured by laser diffraction during the settling process and given by the method. This shows the ability of the method to correctly assess this parameter except for the first minutes, when the settling process is dominated by the coarsest particles. The poor results associated to the coarsest sand fraction can be mainly explained by the very low residence time of these fractions. To correctly take into account these particles, some improvements have to be performed on the optimisation of the water depth, h, and on the hypothesis on the vertical distribution of their $\lambda_{z,i}$.

Moreover, the procedure seems promising for the characterization of fine materials that are the main vectors of pollutants in hydrosystems. Consequently, its application to real suspensions sampled from the French Rhône River (Marcoule to Arles reach) is presently in progress in order to characterize the settling groups according to the season and the hydraulic conditions. At the same time, the composition of these fractions and their sorption-desorption behavior in relation to pollutants (such as radionuclides or metals) is also studied to complete the parameter set needed for the water quality model developed at IRSN.

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