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Study on the impact of particle size distribution on turbidity in water

Weipeng He, Jun Nan*

Skate Key Laboratory of Urban Water Resource and Environment, School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, PR China
Tel. +86 451 86084169; Fax: +86 451 86283001; email: nanjun_hit@163.com

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ABSTRACT

To better understand the impact of particle size distribution on water turbidity, a series of flocculation tests were carried out at different slow stirring speeds to produce the experimental data needed. Results were obtained using an online turbidimeter, coupled with an online particle counter, and all the measurements were performed at regular intervals. It was found that within the turbidity range of 0–40 NTU, particle amount increased linearly with increasing turbidity, whereas for the range of 40–100 NTU, particle amount remained almost unchanged and the correlation between particle amount and turbidity became poor, possibly due to the fact that particles in different size ranges contributed differently to the change of turbidity. The results also demonstrated that particles larger than 5 μm were greatly related to water turbidity, but for particles smaller than 5 μm, a poor correlation between the number of particles and turbidity was found. Therefore, the number of particles smaller than 5 μm should be particularly measured and controlled during water treatment processes to produce maximum removal of particles from water. As expected, the particle counter can be used as a useful supplement to the turbidimeter, in order to effectively evaluate water treatment efficiency and ensure water safety.

Keywords: Flocculation; Particle size distribution; Turbidity; Small particle; Particle counter

1. Introduction

Colloidal material in water is a major carrier of pollutants, such as toxic metals, microorganisms, and anthropogenic organic compounds. Therefore, it is necessary to ensure maximum removal of colloids and other small particles from water [1–3]. Typical solid/liquid separation processes begin with flocculation, followed by sedimentation and/or filtration [4]. Ideally, all of the small particles should be incorporated into relatively large aggregates or flocs that may settle rapidly, and/or produce permeable filter cakes, giving clear supernatants or filtrates [5]. Since turbidity is greatly related to solid/liquid separation and water quality, the effectiveness of treatment processes is generally measured by the turbidity of treated water at almost all the water treatment works (WTWs) [6].

Three common types of turbidimeters (i.e. bench top, portable, and online instruments) are employed today. Modern turbidity measurements are based on the technique of nephelometry, which measures the amount of light scattered at right angles to an incident light beam by particles present in water [3]. Factors affecting the turbidity measurement include particle

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*Corresponding author.

size, color, configuration, surface nature, etc. [2,7]. Simply stated, turbidity is the measure of relative clarity of water [8]. In general, the lower the turbidity level, the higher the water quality, indicating a lower risk that people may develop gastrointestinal diseases [8–10]. However, it is important to note that there is no direct relation between the number of particles and the turbidity of a sample [3]. As reported by Srivastava [11], a very low turbidity value does not completely ensure that particles are absent. This suggests that turbidity can only provide rather limited information on the separation processes.

Essentially, flocculation is a process to encourage destabilized small particles to form larger aggregates, whereas sedimentation and filtration processes can only remove particles in specific size ranges. Therefore, the performance of solid/liquid separation processes may be directly measured in terms of particle size distribution (PSD) [12–14]. Online particle counters can quantify the actual particle number and the distribution with size, and therefore the instruments are routinely used for real-time PSD characterization and treatment efficiency determination at WTWs. When compared to turbidity, particle counting may provide more detailed information on the characteristics of suspended particles in water, and especially, be more sensitive to small particles [9,15–17]. McCoy and Olson [18] investigated the relationship among turbidity, particle number, and bacteriological quality within water distribution lines, and concluded that no predictable relationship existed between bacteriological quality and water turbidity. Fortunately, particle counting can be used for examining the removal of pathogenic entities from drinking water, thus guaranteeing maximum public health protection [11]. The most suitable application for particle counters is as a diagnostic tool for the identification of operating conditions which reduce or minimize particle number in treated water. To ensure water safety, more and more WTWs begin to use particle size measurements in conjunction with turbidity measurements to monitor and control water treatment operation [9].

In the present study, a series of jar-test flocculation and sedimentation experiments were carried out under different slow stirring conditions, to investigate the impact of PSD on water turbidity. During the overall process of each test, an online turbidimeter was used to determine water turbidity, and an online particle counter was applied to measure the number and size distribution of particles in water. All these measurements were performed at regular intervals. Firstly, the PSD evolution with time during three typical processes of flocculation was examined. Secondly, the relationship between particle amount and water turbidity was discussed via correlation analysis. Furthermore, similar discussion was conducted on the relationship between particle number in each size range and water turbidity. Finally, the number of particles smaller than 5 μm was proposed to characterize the removal efficiency of smaller particles from water. The study has showed that the combined use of the new index, residual turbidity, and particle amount may provide valuable guidance on the maximum removal of suspended particles, and the optimization of water treatment processes, thus ensuring water safety.

2. Materials and methods

2.1. Suspension and coagulant

Kaolin clay (Tianjin, China) suspension was used as testing water sample. The stock suspension was prepared similarly to that of Yukselen and Gregory [19]. The particles had an average size of about 3 μm (Fig. 1), determined by a 2200 PCX Particle Counter (HACH, USA). For flocculation tests, the stock

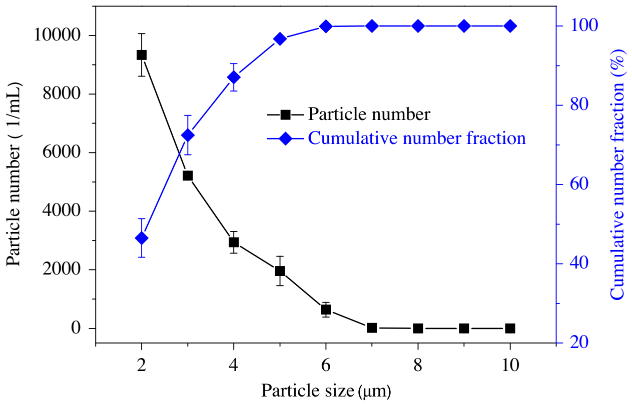

Fig. 1. PSD in testing water sample before coagulant addition. The size and number were determined by a PCX 2200 Particle Counter (HACH, USA). Error bars represent the standard deviation for each point.

solution was diluted in the tap water of Harbin, China, giving a final clay concentration of 100 mg/L. Harbin tap water has medium total hardness (ca. 160 mg/L as $CaCO_3$) and alkalinity (ca. 115 mg/L as $CaCO_3$) and a pH of around 7.8. To avoid the disturbance of divalent metal ions such as Ca^{2+} and Mg^{2+} in tap water, a small amount of humic acid (Shanghai, China) was added into the testing sample [20]. The final suspension containing 100 mg/L kaolin and 2 mg/L humic acid had a turbidity of about 100 NTU, determined by a MicroTOL online turbidimeter (HF, USA). All the tests were conducted at room temperature (22 ± 1°C) and replicated 2–3 times.

Polyaluminum chloride (PACl) was selected as the coagulant to form colloid aggregates. Stock PACl solutions of 1% w/w were prepared by dissolving the reagent in deionized water (5 g dissolved in deionized water to 500 mL) and, for the flocculation tests, directly pipetted in the testing water without further dilution.

2.2. Characteristics of the stirred tank

The flocculating reactor used was a rectangular stirred tank (homemade) with a bottom length $D = 280$ mm and a liquid height $H = 230$ mm, and filled with 18 L of testing water sample (see Section 2.1) as working fluid. For agitation, a R1342-type impeller (IKA, Germany) with a diameter $d = 50$ mm was used and the center of the impeller was positioned at $C = H/3$ from the tank bottom. This mixing system was successfully used in some of our previous studies, e.g. in [2].

2.3. Apparatus

A MicroTOL online turbidimeter (HF, USA), coupled with a 2200 PCX Particle Counter (HACH, USA), was used in a modified version of the standard flocculation-test procedure (Fig. 2), to measure PSD dynamics and water turbidity. The PSD was described by particle amount, and the number of particles within different size ranges. Here, the size ranges corresponded to the channels (i.e. CH1–CH5) in the particle counter (Table 1), and the particle amount was defined as the sum of particle numbers in all channels.

For dynamic monitoring, water sample (at 5 cm below the liquid surface) from the flocculating reactor was circulated at a flow rate of 100 mL/min through 5 mm internal diameter transparent plastic tubing by means of a peristaltic pump. The pump was located after the turbidimeter and the particle counter to avoid the effects of possible floc breakage in the pinch portion of the pump. Readings for the PSD and turbidity were taken every 1 s and the results were stored in a computer for subsequent spreadsheet analysis.

2.4. Procedure

Optimal PACl dosage was firstly determined by performing a series of flocculation tests with incremental increases in the coagulant dose (between 1 and 10 mg/L as Al). For each test, after a certain amount of PACl coagulant dosed in the reactor, the testing suspension (100 mg/L kaolin and 2 mg/L humic acid in Harbin tap water) was mixed rapidly at 400 rpm for 30 s, followed by a slow stirring phase at 100 rpm for 20 min. Then the turbidity of supernatant (at 5 cm below the liquid surface) was measured, after a 20-min sedimentation (without mixing), by a MicroTOL online turbidimeter (HF, USA). As

Table 1
Particle size ranges used in this study. The size ranges were expressed as various channels (i.e. CH1–CH5) in the particle counter

Channel	CH1	CH2	CH3	CH4	CH5
Size range (μm)	2–3	3–5	5–15	15–25	>25

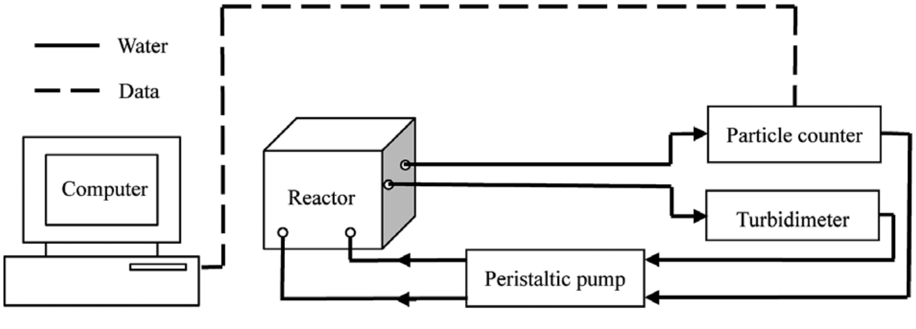

Fig. 2. Schematic of experimental setup.

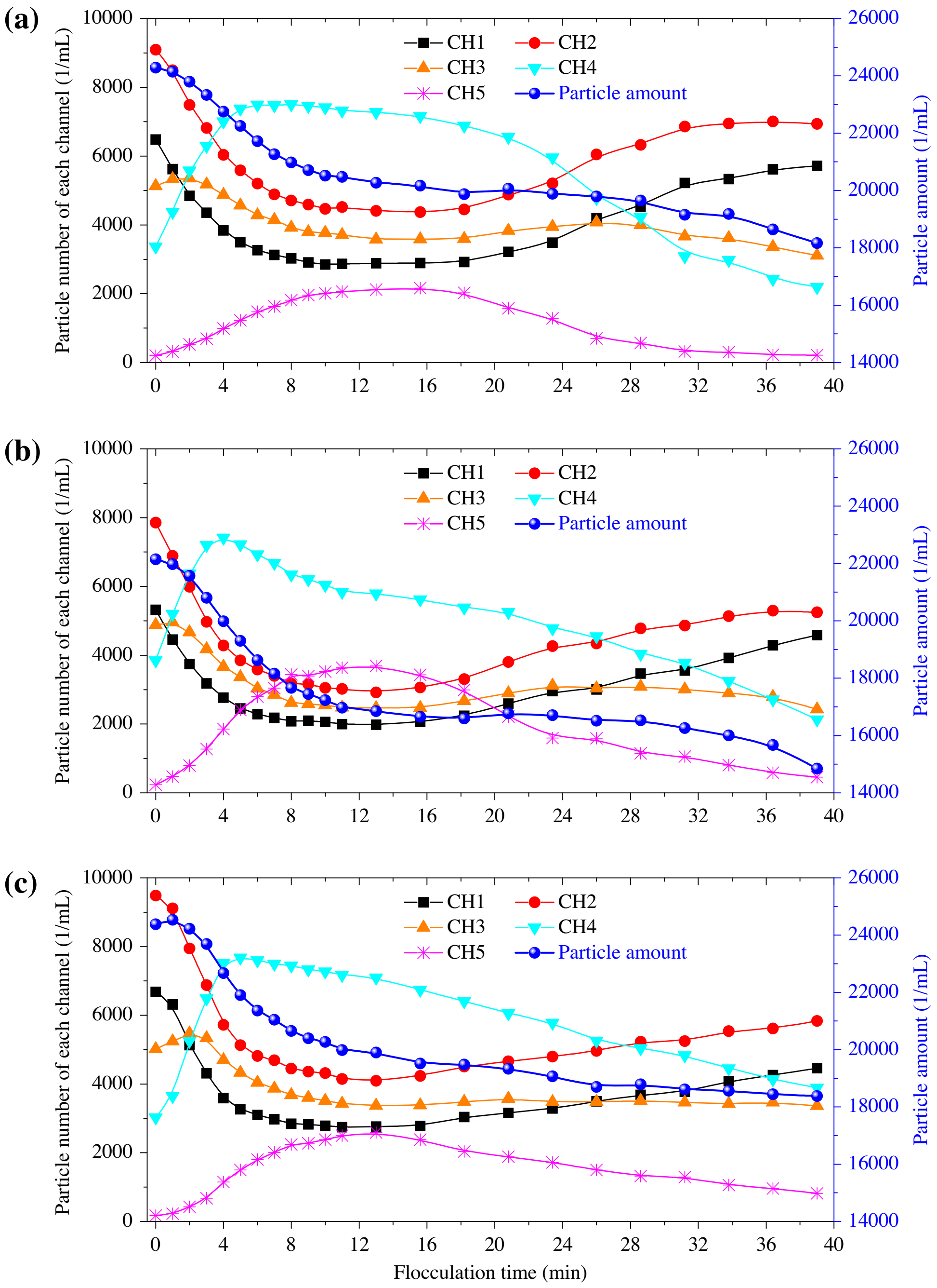

Fig. 3. Variation in PSD with flocculation time under different slow stirring speeds (*N*): (a) $N = 50$ rpm; (b) $N = 90$ rpm; (c) $N = 150$ rpm. (PACl dosage, 2.2 mg/L as Al; rapid stirring, 400 rpm for 30 s; slow stirring time, 39 min.)

described by Duan and Gregory [8], the amount of coagulant giving the minimum turbidity is the optimal dosage. In this study, turbidity removal rate reached a peak value of about 90% at the PACl dosage of 2.2 mg/L as Al, which was selected as the optimal dosage and then used for all other flocculation tests mentioned below.

For dynamic tests, after allowing 1 min for steady-state turbidity and PSD to be established, PACl solution was dosed and the suspension was rapidly mixed at 400 rpm for 30 s, followed by a slow stirring phase at six different stirring speeds (50, 70, 90, 110, 130, and 150 rpm) for 39 min, and a 20-min sedimentation.

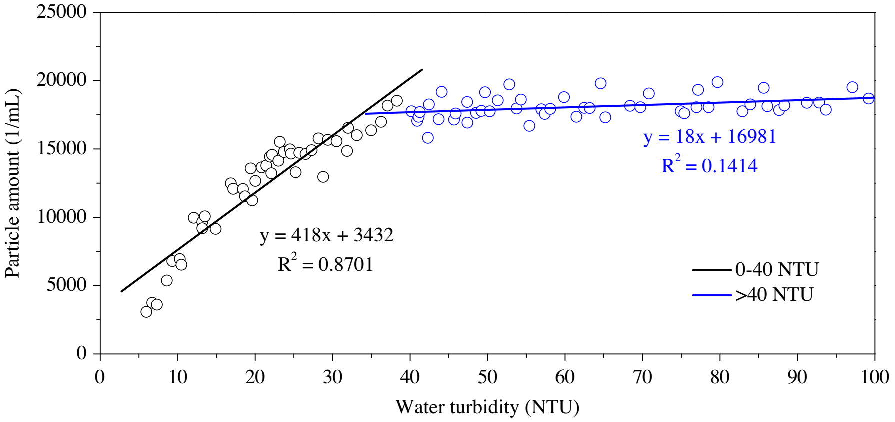

Fig. 4. Correlation between particle amount and water turbidity, measured during three flocculation-test processes (including flocculation and sedimentation phases) with slow stirring speeds of 50, 90, and 150 rpm. (PACl dosage, 2.2 mg/L as Al; rapid stirring, 400 rpm for 30 s; slow stirring time, 39 min.)

3. Results and discussion

3.1. Evolution of PSD during flocculation

Flocculation is considered as a process to change the PSD dynamics, i.e. to turn a large number of small particles into a smaller number of larger ones [3,12,13]. Extensive studies have focused on the PSD evolution during flocculation, monitored using particle size analyzers (e.g. Malvern Mastersizer 2000) [6], image analysis [21–23], or turbidity fluctuation methods [3,20], but there has been little previous work using the online particle counting technique. Here, a 2200 PCX Particle Counter (HACH, USA) was used to monitor the change in PSD with flocculation time, and the results for slow stirring speeds (*N*) of 50, 90, and 150 rpm are shown in Fig. 3.

The PSD evolution, measured by the particle counter, clearly exhibited the continuous floc growth. At $N = 50$ rpm, the number of particles in CH1–CH3 decreased sharply and the number of particles in CH4–CH5 increased dramatically in the early stage of flocculation (Fig. 3a), indicating that small particles aggregated rapidly to form larger ones. As flocculation continued, the formed larger particles caused a lower particle collision rate [20] and therefore further growth was restricted. After a certain time, particle number in each channel reached a steady-state value, but the time needed appeared to be different, i.e. steady state was attained faster for CH1 and CH2 than for CH3 and CH4, followed by CH5. This suggested that large particles did not aggregate directly from primary particles. Instead, they were formed

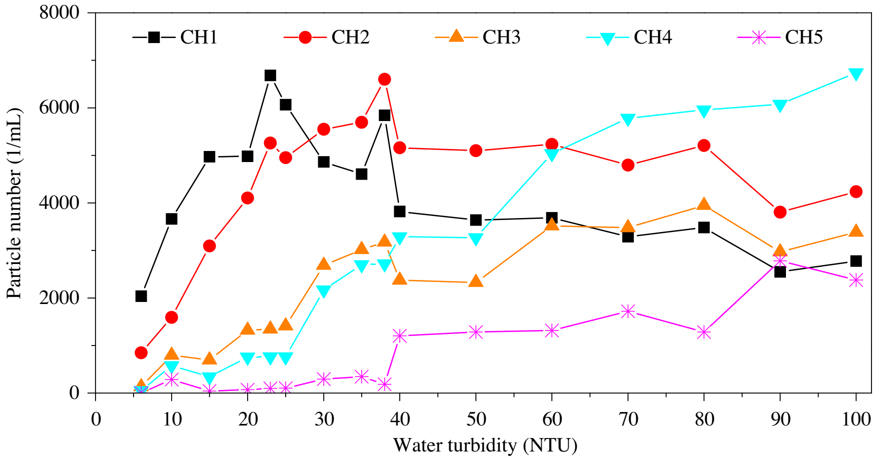

Fig. 5. Relationship between particle number in each channel of the particle counter and water turbidity. (Same conditions as for Fig. 4.)

with flocculation time through the binding of small aggregates and primary particles. A similar finding was reported by Stone and Krishnappan [23], who stated that small particle clusters (micro-flocs) are the formational units of larger flocs during flocculation. After about 22 min, the number of particles in CH1–CH2 began to increase and the number of particles in CH3–CH5 began to decrease (Fig. 3a), possibly due to floc breakage [20] and/or settling [24], resulting from relatively long duration of slow stirring.

The variation trend in PSD with flocculation time at slow stirring speeds of 90 (Fig. 3b) and 150 rpm (Fig. 3c) was similar to that shown in Fig. 3a, but more significant changes in particle numbers in CH1–CH5 were observed. Serra et al. [13] found that each device has a shear rate for which there is a transition from aggregation dominated conditions to breakage dominated conditions. They also claimed that although there were sufficient collisions to make larger particles at higher shear, these particles were not stable due to breakage. Therefore, it was possible that more dramatic breakage caused more significant PSD changes with increasing shear.

Additionally, the variation curves of particle amount (i.e. the sum of particle numbers in CH1–CH5) showed that particle amount decreased during the overall process of flocculation whatever the shear (Fig. 3). They also demonstrated that a smallest number of largest flocs were formed at 90 rpm, followed by slow stirring speeds of 50 and 150 rpm.

3.2. Impact of PSD on water turbidity

Particle size, configuration, color, and refractive index determine the spatial distribution of the scattered light intensity around the particle, thus affecting the turbidity measurements [3]. Generally, particles formed by flocculation are not uniform in size, but rather can vary over a wide range; whereas only the particles in specific size ranges can be removed by subsequent separation processes, such as sedimentation and filtration [4,12,14]. This means that significant changes in PSD occur during the overall processes of flocculation and sedimentation. Also, water turbidity changed remarkably. Therefore, the PSD and corresponding turbidity determined during three flocculation-test processes (including flocculation and sedimentation phases) with slow stirring speeds of 50, 90, and 150 rpm were used here (see Section 2.4 for experimental conditions), to better understand the relationship between PSD and water turbidity.

Fig. 4 shows the relationship between particle amount and water turbidity. Within the turbidity range of 0–40 NTU, the particle amount significantly increased with increasing water turbidity, and there was a good correlation between particle amount and water turbidity (correlation coefficient, $R^2 = 0.8701$). A similar conclusion was reached by McCoy and Olson [18], who stated that turbidity and particle numbers were directly proportional. However, within the range of 40–100 NTU, a poor correlation was found between particle amount and water turbidity, and the R^2 was merely 0.1414 (Fig. 4); in this case, particle amount almost unchanged with increasing turbidity, and even a same particle amount sometimes produced different values of turbidity. This was likely attributed to the fact that the floc particles in different size ranges contributed differently to the change of turbidity [2], and this phenomenon would be discussed later.

The relationship between particle number in each channel of the particle counter, and water turbidity is shown in Fig. 5. When water turbidity was lower than 25 NTU, small particles dominated, and particle number in CH1 was the largest, followed by CH2. For water turbidity values between 30 and 60 NTU, CH2 began to have the largest number of particles, followed by CH1, and the number of particles in CH3–CH5 increased with increasing turbidity. For water turbidity values of 60 NTU or above, larger particles sharply increased and particle number in CH4 exceeded particle number in CH2. Interestingly, the number of particles in CH1–CH2 slightly changed when water turbidity increased from 40 to 100 NTU. All the results shown in Fig. 5 indicated that similar PSDs existed in water of different qualities. Also, water turbidity was mostly affected by larger particles (here, i.e. particles in CH3–CH5).

As mentioned in Section 1, all modern turbidimeters generally utilize the nephelometric measurement principles, suggesting that when light passes through a fluid containing suspended particles, the light beam interacts with the particles, and the particles absorb the light energy and re-radiate light in all directions. According to a recent review paper written by Gregory [3], the spatial distribution of the scattered light intensity around the particle was greatly affected by particle size and refractive index, i.e. for particles much smaller than the wavelength of the incident light, typically expressed in nanometers (nm), they scatter light of approximately equal intensity in all directions; whereas larger particles form a spectral pattern that results in greater light scattering in the forward direction (away from the incident light) than in the other directions. In the last case, the scattering pattern and intensity of the light beam transmitted through the sample can also be affected by the particles absorbing certain wavelengths of the transmitted light. This may be used to explain the results from the curves shown in Fig. 5. Within the turbidity range of 0–40 NTU, small particles (i.e. particles in CH1–CH2) dominated, so they might independently scatter the incident light. In addition, the number of small particles increased with increasing turbidity (Fig. 5), and increasing particle numbers caused stronger intensity of the scattered light, thus producing larger values of turbidity. That was why a good correlation between particle amount and water turbidity was observed for water turbidity values of 0–40 NTU (Fig. 4). Within the turbidity range of 40–100 NTU, larger particles (i.e. particles in CH3–CH5) became dominant over smaller particles, and the number of these larger particles increased with increasing turbidity (Fig. 5). These larger particles greatly affected the scattering pattern and intensity of the light beam transmitted through the sample. Nevertheless, there was a poor correlation between particle amount and water turbidity for water turbidity values of 40–100 NTU (Fig. 4), possibly due to the insignificant change in the number of small particles (Fig. 5). This phenomenon motivated us to further investigate the impact of small/large particles on water turbidity.

Fig. 6 shows the relationship between the number of particles smaller than 5 μm ($N_{d<5}$), corresponding to smaller particles (i.e. particles in CH1–CH2), and water turbidity. Within the turbidity range of 0–40 NTU, the $N_{d<5}$ increased with increasing turbidity, but for the range of 40–100 NTU, the $N_{d<5}$ decreased with increasing turbidity. In addition, the values of R^2 were both smaller than 0.6500, indicating a poor correlation between the $N_{d<5}$ and water turbidity. These results demonstrated that the turbidity exhibited more complex behavior for smaller particles. Therefore, the number of these smaller particles should be carefully monitored at WIWs (see Section 3.3).

The relationship between the number of particles larger than 5 μm ($N_{d>5}$), corresponding to larger particles (i.e. particles in CH3–CH5), and water turbidity is shown in Fig. 7. The $N_{d>5}$ increased linearly with increasing turbidity in the overall range of turbidity examined here, and the value of R^2 was 0.8678 for the turbidity range of 0–40 NTU and 0.8332 for the range of 40–100 NTU. This suggested that water turbidity was greatly related to larger particles, which was in excellent accordance with our previous study [2].

Figs. 6 and 7 indicated that water turbidity was more sensitive to larger particles than smaller particles. In other words, a lower turbidity value appeared to represent the smaller number of larger particles, but did not provide useful information on the number of smaller particles [11]. It also seemed that the turbidity of water would be lowered by remarkably reducing the number of larger particles, which was not true of smaller particles. Therefore, the number of smaller particles (size smaller than 5 μm) should be particularly measured and controlled during the process of water treatment.

3.3. Practical implications in the engineering aspects

To effectively evaluate water treatment efficiency and ensure water safety, the number of particles smaller than 5 μm ($N_{d<5}$) was proposed and then applied in conjunction with water turbidity. Flocculation tests were conducted under six different slow stirring speeds (N) of 50, 70, 90, 110, 130, and 150 rpm, respectively. The $N_{d<5}$, residual particle amount, and residual turbidity measured after a 20-min sedimentation are shown in Fig. 8.

When the slow stirring speed (N) increased from 50 to 90 rpm, the $N_{d<5}$, residual particle amount, and residual turbidity all decreased, producing better

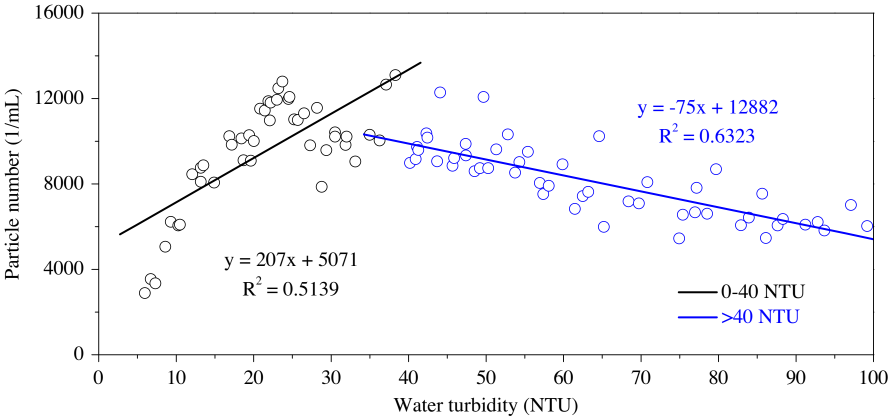

Fig. 6. Correlation between the number of particles smaller than 5 μm ($N_{d<5}$) and water turbidity. (Same conditions as for Fig. 4.)

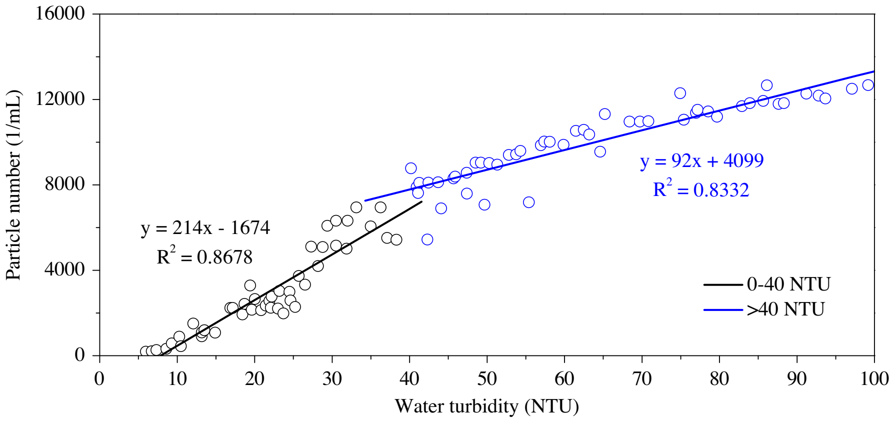

Fig. 7. Correlation between the number of particles larger than 5 μm ($N_{d>5}$) and water turbidity. (Same conditions as for Fig. 4.)

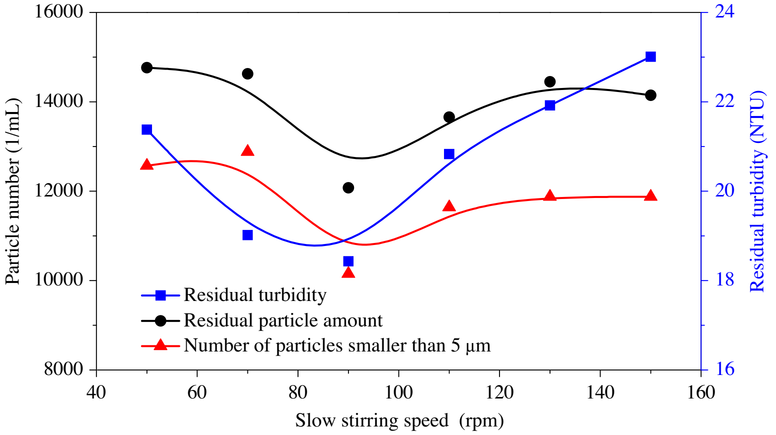

Fig. 8. Impact of slow stirring speed on the results of flocculation tests. (PACl dosage, 2.2 mg/L as Al; rapid stirring, 400 rpm for 30 s; slow stirring time, 39 min.)

water quality (Fig. 8). At $N = 90$ rpm, all the three indexes reached the minimum values, producing the best quality of settled water. For higher slow stirring speeds ($N > 90$ rpm), all the three indexes decreased and the treatment efficiency became poorer (Fig. 8). These were consistent with the study of Serra et al. [13], who defined three shear ranges, i.e. low shear, where the aggregation dominated over breakage and aggregate growth rate increased in proportion to the shear; intermediate shear, where flocculation rates were maximized and breakage minimized; and large shear, where breakage dominated aggregation, although sufficient collisions occurred to encourage the rate of aggregation. Under lower shear (e.g. 50 and 70 rpm), the increased shear produced a higher rate of floc growth due to the increased particle collisions [20]. At 90 rpm, it seemed that the maximized flocculation rate caused that the PSD would be shifted toward larger sizes. As suggested by Boller and Blaser [25], larger particles generally have higher removal efficiencies than smaller particles of similar density, thus producing the best flocculation efficiency. At 110 rpm or above, significant breakage occurred to break larger aggregates into smaller pieces, and therefore, the quality of settled water became poor due to the irreversibility of PACl-floc breakage [20].

4. Conclusions

The main conclusions of this study were listed as follows:

1. Temporal changes in PSD, determined using an online particle counting technique, clearly exhibited the continuous floc growth. During flocculation, large particles did not aggregate directly from

primary particles, but were formed over time through the binding of small aggregates and primary particles. At higher shear, more dramatic breakage caused more significant PSD changes with increasing shear.

2. Particle amount increased linearly with increasing water turbidity within the turbidity range of 0–40 NTU. However, for the range of 40–100 NTU, particle amount removed almost unchanged and a poor correlation was found between the two indexes. This was likely attributed to the fact that particles in different size ranges contributed differently to the change of turbidity.
3. Similar PSDs existed in water of different qualities. Also, water turbidity appeared to be greatly related to larger particles due to turbidity measurement principles.
4. Water turbidity was mostly affected by the number of particles larger than 5 μm ($N_{d>5}$), and the $N_{d>5}$ increased linearly with increasing turbidity in the overall range of turbidity (0–100 NTU), suggesting that turbidity seemed to represent the number of larger particles. However, for particles smaller than 5 μm, the turbidity exhibited more complex behavior, and there was a poor correlation between the number of these particles ($N_{d<5}$) and water turbidity. Therefore, the $N_{d<5}$ should be particularly measured and controlled during water treatment processes.
5. The combined use of turbidity and particle counting techniques can effectively evaluate water treatment efficiency and ensure water safety at water treatment works.

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