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Fine soil particle aggregation in ultra-fine bubble irrigated paddy fields

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ABSTRACT

The flotation method of ultra-fine bubbles (UFB) aims to address pollution and has been used for combating the undesirable water conditions of contaminated soils. Hence, water containing UFB is gaining increasing attention for potential agricultural applications. Although certain hypotheses have been proposed, such as the collection of ions in water through the electrical characteristic of UFB, no clear experimental data have been provided. We found that improvement in turbidity may cause the adsorption of fine soil particles in the water by the UFB, thereby improving the quality of the water. The data from the paddy field showed that a decrease in turbidity (below 2 nephelometric turbidity units) occurred over a short period of time (3 days). UFB concentration is directly related to turbidity with a coefficient of determination of 0.93. This phenomenon was also observed through the distribution of bubbles and soil particles, where the average particle size increased because of the aggregation of soil particles and the decrease in turbidity in the paddy field, indicating that UFB collect soil particles and thereby improve water quality. Therefore, UFB are highly effective in cleaning rice field water and will be a preferred method for purifying the environment in the future.

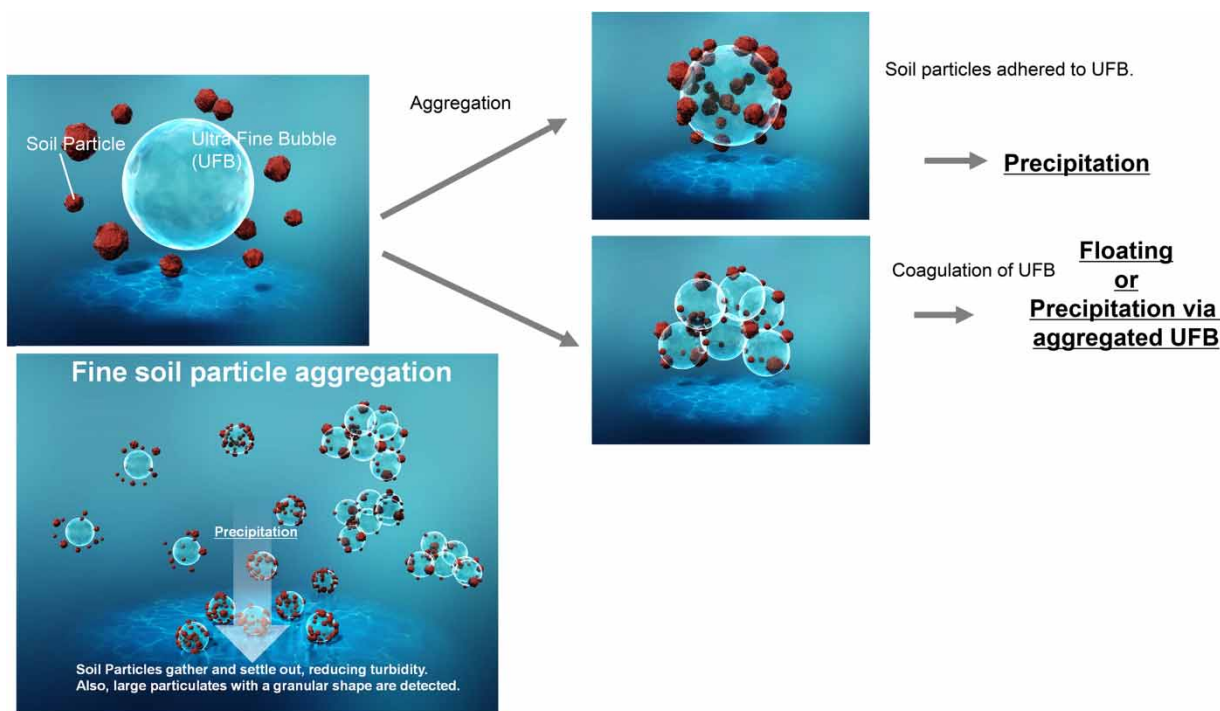
Key words: aggregation, fine soil particles, paddy field, pollution, rice growth, ultra-fine bubbles

HIGHLIGHTS

- Ultra-fine bubbles (UFB) improve water quality for agricultural applications.
- Temporal variation of concentration of bubbles and fine soil particles and turbidity is considered.
- Turbidity decreases with time, and UFB concentration is strongly correlated with turbidity.
- UFB size increased because of aggregation of fine soil particles.
- Decrease in turbidity in the paddy field indicated that UFB attract fine soil particles.

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GRAPHICAL ABSTRACT



INTRODUCTION

Submicron-scale bubbles suspended in water are referred to as ultra-fine bubbles (UFB) by ISO 20298-1 (fine bubble technology). Research on the use of UFB is ongoing, particularly for environmental and agricultural applications. Additionally, interest in UFB–water is growing as it can be prepared using only water and air, making it an inexpensive alternative that can be prepared anywhere. Several stability hypotheses exist for the properties of UFB in water. Regarding the relationship between UFB and impurities, stability theory posits that impurities stabilize UFB (Yasui *et al.* 2016). We have also shown that UFB can stay in pure water for a long time, depending on the conditions (Ueda *et al.* 2013). Stability hypotheses suggest that UFB stability results from microscopic particles attached to the bubbles. An increase in the number of UFB may also decrease the electrical conductivity of the water as water molecules and ions may collect around the bubbles, thereby altering the properties of the water.

Regarding the agricultural use of water with UFB, we have examined the environmental response of food crop species, such as soybean (Iijima *et al.* 2020), cereal, and leguminous species (Iijima *et al.* 2022a), using deionized water. In experiments, UFB promoted high growth under oligotrophic conditions in hydroponic soybean cultivation. UFB also exhibited the same effect on growth when polyethylene glycol was added as desiccation stress (Iijima *et al.* 2022b). Thus, we infer that UFB promote plant growth under environmental stress. However, it is currently unknown whether UFB affect crop growth in soil cultivation. We are currently conducting field demonstrations in rice crop trials and are monitoring the parameter characteristics.

UFB have been effectively used in radical generation (Liu *et al.* 2021), flocculation, sedimentation (Xiao *et al.* 2018), and flotation beneficiation. Recent research has focused on improving the flotation effect of UFB (Chang *et al.* 2020; Li *et al.* 2022; Wang *et al.* 2022). The current application of UFB in rice cultivation suggests the possibility of genetic changes in the cells as well as the contribution of chemical effects such as radical activity. In addition, limited studies have discussed direct soil and water changes and provided accurate data. Therefore, we focused on the direct effects of UFB on soil and water. UFB are also being used for cleaning oil sands and are expected to be used as an environmentally friendly technology (Thuy Bui *et al.* 2022). By clarifying the relationship between soil particulates and UFB through field trials, the significance of these applied studies would become more apparent.

In this study, we examined the presence of UFB and their changes over time in several paddy fields. By simulating the soil in the laboratory, we also examined changes in the UFB and particulate mixture using Japanese Industrial Standard (JIS) test powder 1 (Type 11 Kanto loam). We believe that this is a novel approach in that we are attempting to elucidate the process from an engineering-related perspective, in addition to the growth of bot rice plants and soil microorganisms.

METHODS

This study compared the characteristics of UFB–water in paddy fields using irrigation water and groundwater samples. Two water samples were prepared for the experiments: one was collected in an experimental field for rice cultivation at the University of Shiga Prefecture (Figure 1; Field 1, 2500 Yasaka-cho, Hikone, Shiga 522-8533, Japan), and the other was collected from a paddy field owned by Wing Amagi (Figure 2; Field 2, 3196 Yanaga, Asakura, Fukuoka 838-0031, Japan). UFB–water was generated as air UFB using a pressurized shear-type generator (Eatech-2W (Field 1) and Eatech-1W (Field 2), Corp Eatech) in both cases. In Field 1, we cultivated rice crops with a one-factor (control and UFB irrigation) randomized design with four replicate fields, and in Field 2, we cultivated rice crops considering one factor (control and UFB irrigation) without any replicated fields.

In Field 1, rice was cultivated using irrigation water from Lake Biwa. Water samples were collected on July 23, 2021. The water samples were directly collected from the field supply outlet (control) from the normal paddy field and UFB paddy field (see Figure 1). The paddy field water samples were collected far from the supply outlet (paddy field (normal and UFB)). In Field 2, rice was cultivated by pumping groundwater (Figure 2). Water samples were collected again on September 23, 2021, and sample points were collected at similar locations as in Field 1.

The parent materials of the soils in Field 1 and Field 2 were granite and volcanic ash, respectively. The soil type was Gleyic Fluvisol in Field 1 and Haplic Fluvisol in Field 2 (WRB 2015). According to soil type, the dominant soil clays were smectite, vermiculite, fine mica, and chlorite in Field 1 and allophane and imogolite in Field 2. For granite, there were micro-colloids of 450 nm or less, as well as larger soil particles, which might interact with UFB (Vilks & Bachinski 1996). Volcanic ash particles exist as particle aggregates and settle in the soil (Brown *et al.* 2012). Therefore, the fine soil particles were thought to be fewer than those of granite.

The collected water samples were individually sealed and refrigerated in 80-mL polyethylene terephthalate (PET) bottles, which were opened and measured at the moment each sample was obtained. The sample bottles were kept for up to 7 days (Field 1) and 10 days (Field 2). The number density of air bubbles and fine soil particles was measured using a nanoparticle tracking analyzer (Nanosight LM10, Malvern Panalytical Ltd), and the average values of three measurements were obtained. Turb[®] 550 (WTW, Xylem Japan) was used to measure turbidity. Electrical conductivity, dissolved oxygen concentration, and pH were also measured to determine whether changes occurred during sample storage. The supernatant of the PET bottle was sampled during all measurements, so that precipitated impurities did not affect the measurements.



Figure 1 | Test field at the University of Shiga Prefecture.

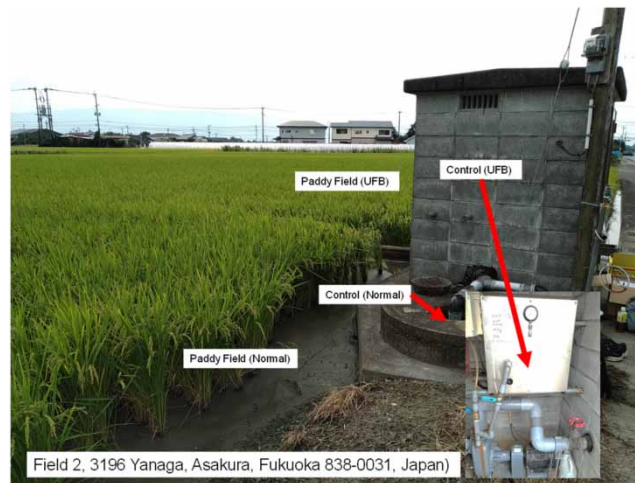


Figure 2 | Test field at Wing Amagi.

RESULTS AND DISCUSSION

We investigated the time variation of the total UFB, fine soil particle concentration, and water turbidity data. We also compared the concentration distribution in each field. For the samples of Field 2, only the distribution was considered to determine the similarity of the trend to that of Field 1, as the soil type of Field 2 was volcanic ash, and almost no change occurred in the turbidity conditions. Type 11 Kanto loam, which was used for the experiments, was also examined for changes in the distribution map.

The data illustrated in **Figure 3** are averages of three measurements for all samples, and the standard deviations are included. The horizontal axis represents the number of days elapsed, and the vertical axis represents the total concentration measured. UFB samples had lower total number concentrations than those of normal samples at Day 0. For up to 3 days, the paddy field (UFB) had a lower concentration, while that of the control (UFB) was higher than that of the control (normal). This reverse trend between the control and paddy field is assumed to be due to the rapid decrease in UFB and fine soil

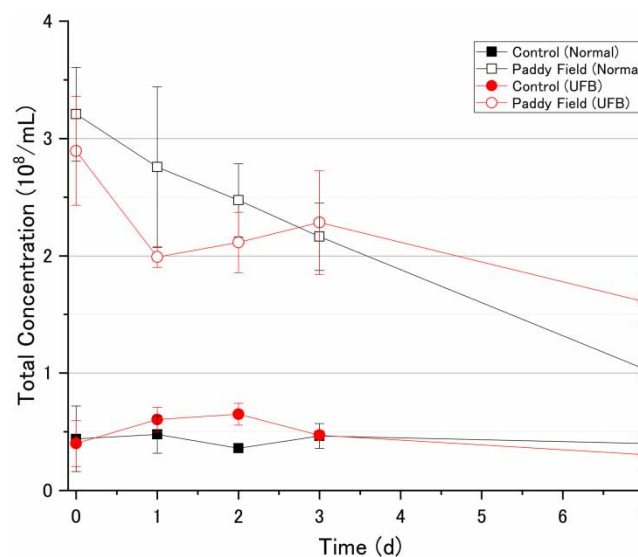


Figure 3 | Daily variation in the total density of air bubbles and fine soil particles (Field 1). ■: Water collected directly from the outlet of the control field, □: Paddy field water from the control field, ●: Water collected directly from the outlet of the ultra-fine bubble (UFB) field, ○: Paddy field water from the UFB field.

particles in the paddy field sample, while UFB in the control sample increased slightly. After 7 days, the trend reversed again, suggesting some interaction between UFB or fine soil particles.

The time variation of turbidity is illustrated in Figure 4. The turbidity trend was not reversed with time, as was the case for the total concentration; the turbidity of the paddy field (UFB) was always lower than that of the normal, whereas the turbidity of the control (UFB) was higher in the normal.

The dissolved oxygen concentration, electrical conductivity, and pH of the refrigerated samples were simultaneously measured over 7 days, but no changes were observed under all conditions. Therefore, we believe that the samples were properly preserved.

The density distribution summarizes the results for each sample for the first, third, and seventh days of generation in Figure 5. Notably, both control samples did not show much change in their distributions, whereas the control (UFB) showed a characteristic peak at approximately 350 nm after 7 days, followed by a rapid decrease in the number density. As for the peak of the distribution, the trend is similar to that of the marginally larger grain size peak shown in the distribution diagram ('LT case') by Wang *et al.* (2021).

In the paddy field case, the first day of generation had a peak at approximately 200–300 nm in the paddy field (UFB). However, the peak almost disappeared after 3 days. In contrast, the paddy field (normal) had a peak at approximately 350 nm, which remained even after 7 days. The same trend was observed at approximately 300–400 nm for the paddy field (UFB). Because the nanoparticle tracking analyzer could not distinguish between UFB and fine soil particles, it was challenging to confirm which of the two peaks was represented. Although coal and glass adsorption has been reported as possible collateral evidence for the adsorption of fine particles and bubbles (Chang *et al.* 2020) (Rosa & Rubio 2018), this study is the first to confirm the actual temporal variation.

The same bubble number density distribution was also found for Field 2 (Figure 6). The distribution illustrated in Figure 6 is for samples obtained 1, 3, and 10 days after generation. The turbidities of the water samples in Field 2 were low after 1 day (control direct: 0.07 nephelometric turbidity unit (NTU), UFB direct: 0.10 NTU); the turbidities were approximately 1/8th of those of their Field 1 counterparts. Additionally, the time variation was within the error range and could not be confirmed. In contrast, the initial total density of the paddy field (UFB) of Field 2 was approximately the same as that of Field 1 ($0.24\text{--}0.33 \times 10^8/\text{mL}$), and the conditions of the paddy water in both fields were approximately the same.

Figure 6 shows that impurities were almost undetectable in control, which remained unchanged after 10 days. However, a peak concentration at approximately 300 nm was noted for the paddy field (normal, day 1), and it remained for 3 days. In contrast, the paddy field (UFB) sample exhibited a high peak at 350 nm after 1 day. Nevertheless, after 3 days, the peak was reduced, and the water became clearer overall. This result indicates that when UFB water is injected into the paddy

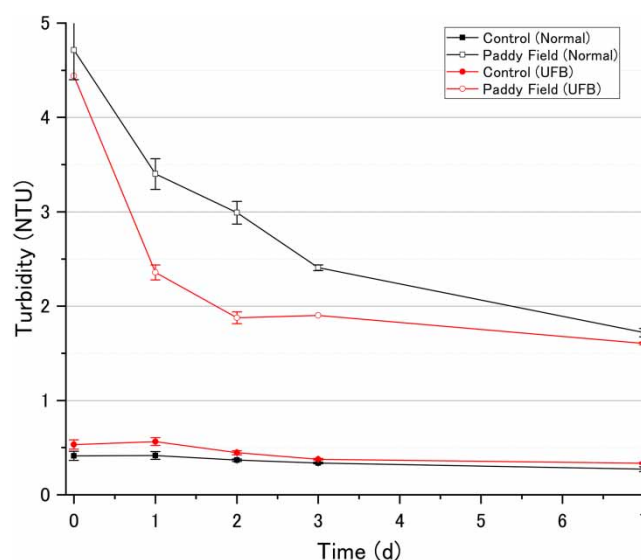


Figure 4 | Daily change in turbidity (Field 1). ■: Water collected directly from the outlet of the control field. □: Paddy field water from the control field. ●: Water collected directly from the outlet of the UFB field. ○: Paddy field water from the UFB field.

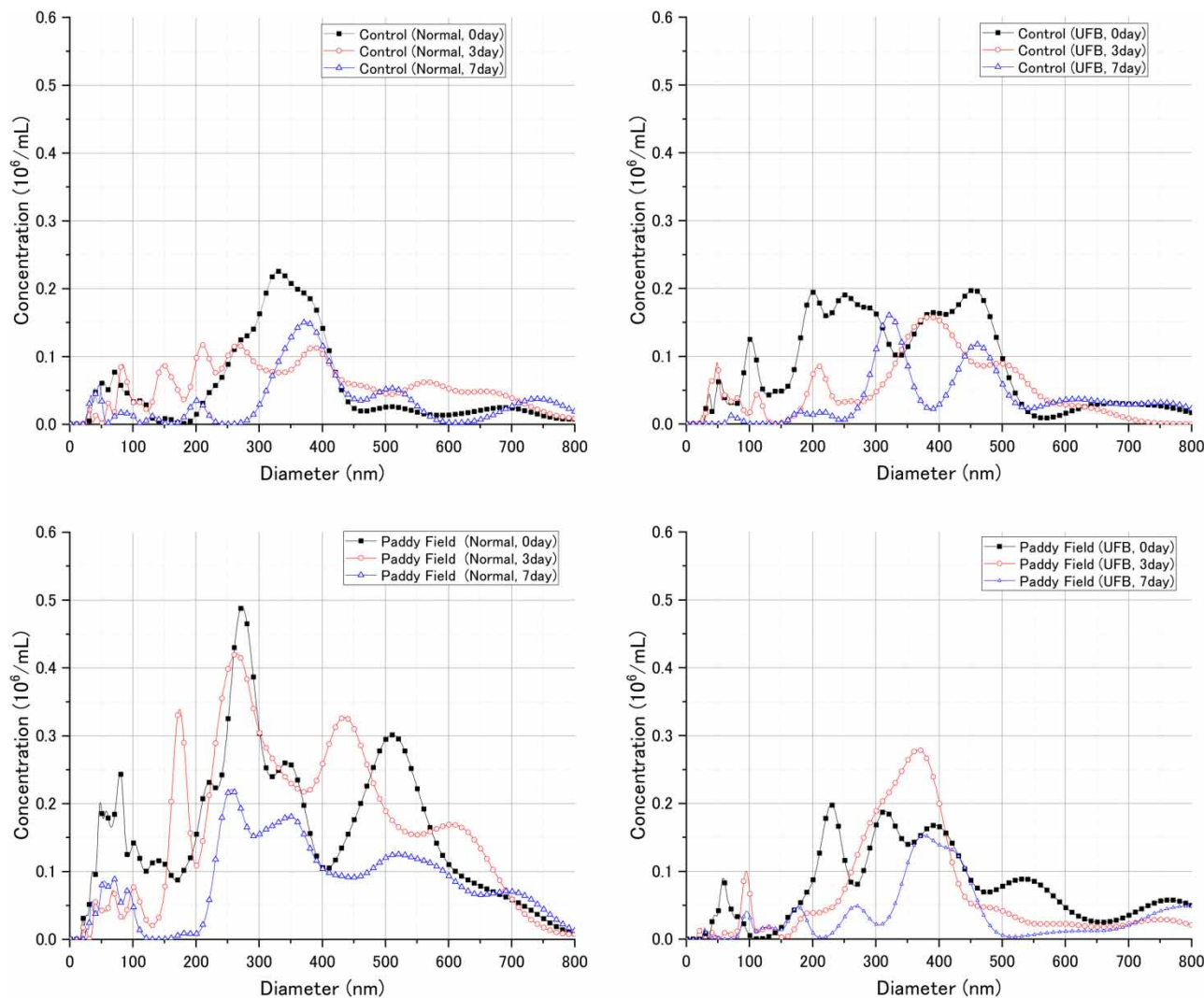


Figure 5 | Density distribution of the number of UFB and fine soil particles (Field 1) (top left: control direct, bottom left: control field, top right: UFB direct, bottom right: UFB field).

field, the impurities in the water and the UFB rapidly condense and are either suspended or precipitated and removed from the water. A similar phenomenon was observed in Field 1.

A laboratory-level test was conducted to evaluate UFB under the same conditions as those present in real-world paddy field water. This test was conducted to confirm the promotion of impurity removal using UFB water in Fields 1 and 2. For the test, UFB water was first generated using a pressurized shear-type UFB generator, and its time variation characteristics were confirmed. The control and UFB water samples were prepared using pure water and JIS test powder 1 (Type 11 Kanto loam); the samples were adjusted to the same turbidity as that of the fine soil particles in Field 1 (approximately 7.0 NTU). For these samples, we verified how the bubble concentration distribution changed from the time immediately after adjustment (day 0) to 1 week later. The variations in the bubble density distributions of the samples with time are illustrated in Figure 7. The figure shows that the purified water had a wider particle size distribution, whereas the UFB water had a uniform peak. The number of distributions decreased over time for both samples, but the decrease was larger for the UFB water; its peak near 250 nm disappeared after 3 days. This result is consistent with the trend observed in Fields 1 and 2. The results suggest that the presence of UFB promotes powder agglomeration. Compared with the case of purified water, the initial turbidity was approximately 7.0 NTU for all samples. In particular, the initial turbidity was 1.89 and 1.76 NTU for the purified water and UFB water, respectively; this decreasing turbidity trend was also observed in the real-world samples. Furthermore, there were

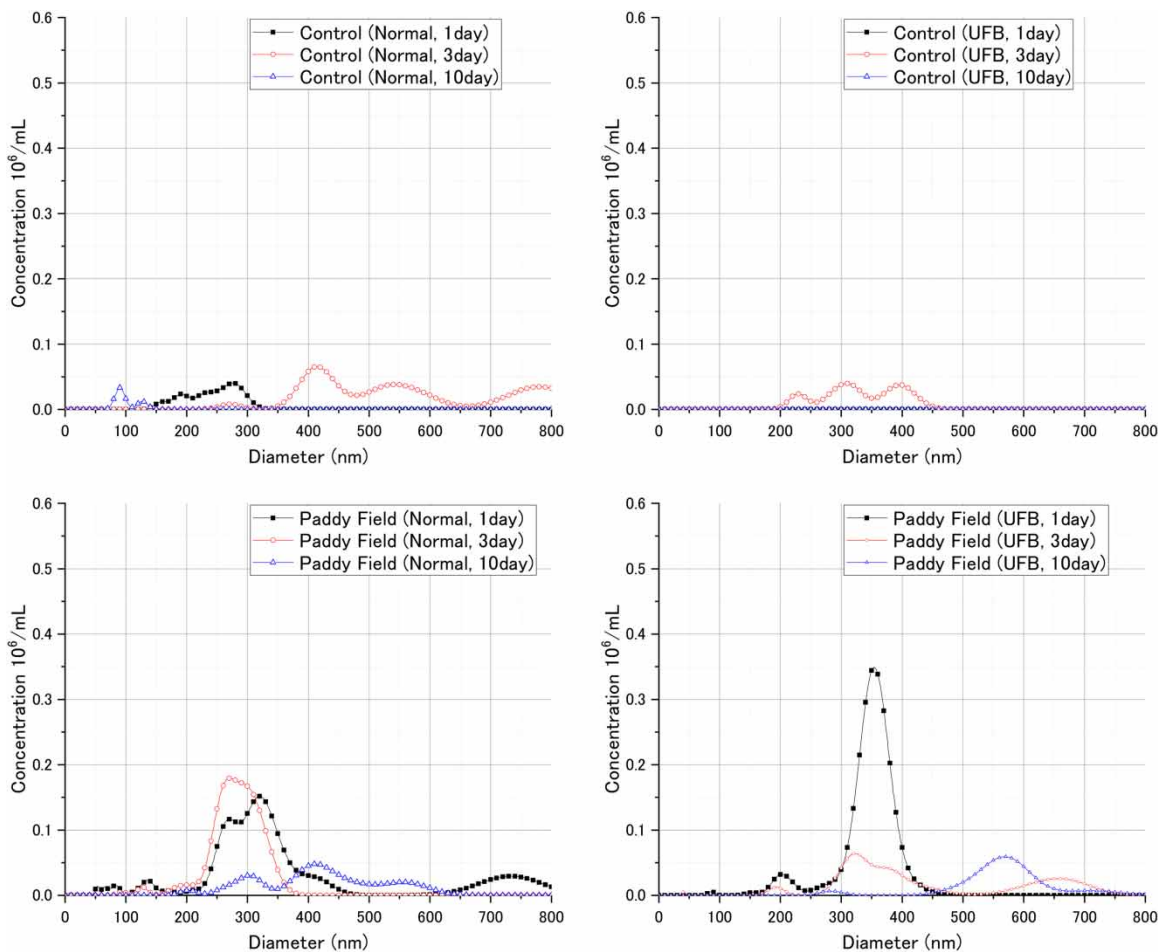


Figure 6 | Density distribution of the number of UFB and fine soil particles (Field 2) (top left: control direct, bottom left: control field, top right: UFB direct, bottom right: UFB field).

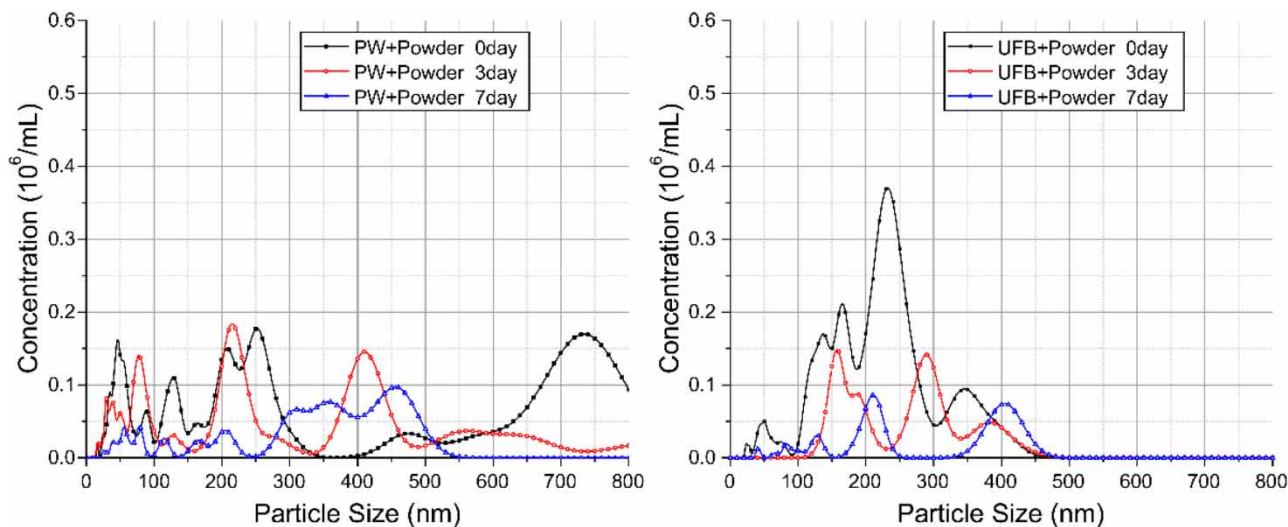


Figure 7 | Density distribution of the number of UFB and fine soil particles (left: purified water with JIS test powder 1, right: UFB water with powder).

no differences in the electrical conductivities, pH, oxidation–reduction potentials, or dissolved oxygen concentrations among the samples. Therefore, no influence from external sources was confirmed.

To confirm the correlation between turbidity and total concentration and distribution, we examined the correlation for each sample of Field 1. The correlation between total concentration and turbidity is illustrated in Figure 8, and the relationship of average diameter and mode particle size with turbidity is illustrated in Figures 9 and 10, respectively. The relationship between the total concentration and turbidity has been described.

Although only judged by the distribution of the particle size, it can be seen that the average particle size of water in the paddy field is approximately 400–500 nm due to UFB contamination, which is about 100 nm larger than normal (Figure 9). However, the mode value is distributed in the same way as the normal (Figure 10). In addition, since the bubble concentration is lower for UFB (Figure 8), we found that the number of bubbles decreased overall, and the average bubble diameter increased as a result of sedimentation or levitation when UFB and fine soil particles were combined. This is also consistent with a decrease in turbidity. We also calculated the fitting value between the number density and turbidity. The coefficient of determination (COD) values were above 0.93.

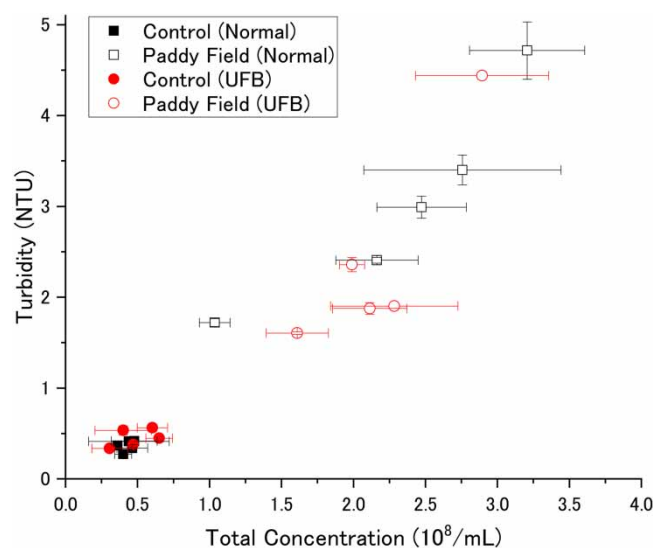


Figure 8 | Correlation between NTU and total concentration in Field 1.

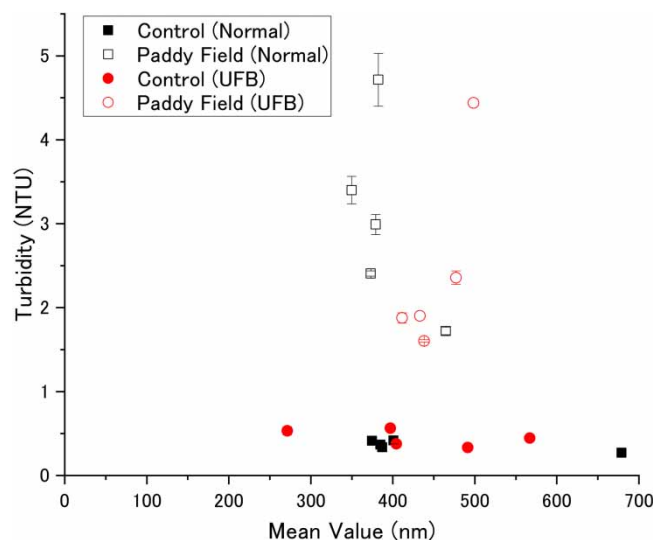


Figure 9 | Correlation between NTU and mean value in Field 1.

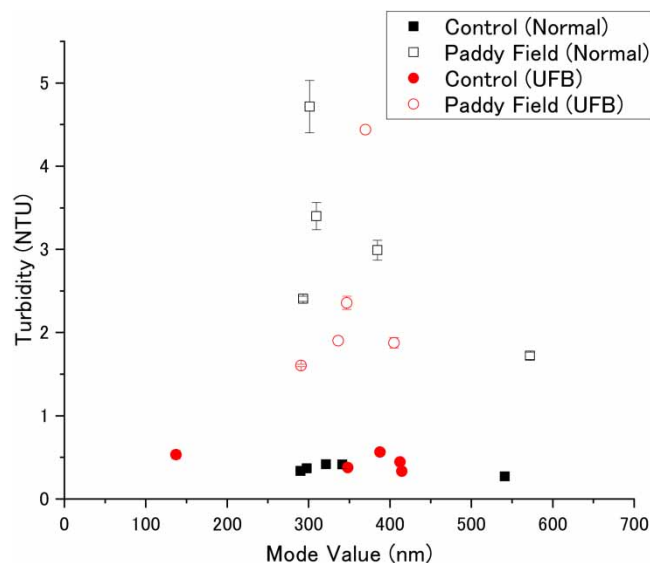


Figure 10 | Correlation between NTU and mode value in Field 1.

In control, the UFB shows some degree of consistency for the mode value, while the mean value varies widely. Since the normal samples (control and paddy field) tend to have a constant value for both mode and mean values, the interaction between particles contained in the raw water and UFB can be seen. Although slight, the UFB concentration is high, which may indicate an increase in turbidity due to UFB turbidity.

Regarding the difference between the mode and mean values in the control sample, we suspect that the bubble size may have been slightly larger because of the fine soil particles adhering to the UFB; in the paddy field, it is clear that the adhered UFB tended to be crushed and reduced, and the turbidity also decreased. Hence, we believe that our hypothesis is plausible. The results of this study are insufficient to suggest any influence from weather conditions. However, we will consider the influence of weather using similar tests in an ongoing study at the University of Shiga Prefecture. We have included a supplementary figure for further information.

CONCLUSIONS

In this study, we examined the coagulation effect of fine soil particles when UFB water was used in rice cultivation. The concentration of bubbles and fine soil particles, turbidity, and variation in density distribution over time were the variables considered. Our observations and experiments established a time-dependence relationship between UFB and fine soil particles in rice-cultivated soil, and we found that a relationship existed between the properties of UFB and water turbidity. In particular, the data in the paddy field showed that turbidity reduction (2 NTU or less) occurred over a short period (3 days). In the distribution of bubbles and fine soil particles, the average UFB size increased because of the aggregation of fine soil particles (control), and the decrease in turbidity in the paddy field indicated that UFB attract fine soil particles and thereby improve the water quality.

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AUTHOR CONTRIBUTIONS

Y. Ueda and M. Iijima conceived the presented idea. Y. Ueda developed the theory and verified the analytical methods. M. Iijima supervised the findings of this work. Y. Izumi, Y. Hirooka, and Y. Watanabe carried out the field experiments. Y. Ueda wrote the manuscript with support from M. Iijima, Y. Hirooka, Y. Watanabe, and Y. Izumi. All authors discussed the results and contributed to the final manuscript.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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