FLOCCULATION, GRAVITY FLOWS, AND TOTAL ORGANIC CARBON HOTSPOTS IN LAKE: INSIGHTS FROM FLUME EXPERIMENTS

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ABSTRACT

Lakes serve as one of the significant sinks for organic carbon. For lake deposits, it is generally accepted that water depth is a primary control on the spatial distribution of total organic carbon (TOC) accumulation because the deeper part of a lake potentially has a higher organic population to be settled. However, lake TOC distribution is often spatially variable regardless of water depth, and an exact mechanistic explanation of TOC hotspots that do not align with the deepest water depth remaines unsolved. We suggest that flocculation significantly influences the lake TOC distribution pattern. The flocculation of fine sediment is possibly a major contributor to the occurrence of high TOC in deposits, as organic material aggregates with flocculated sediment during settling. The process of flocculation positively correlates with water salinity and thus potentially enriches TOC in deposits under a high saline condition. Furthermore, depending on the extent of flocculation, a lake entering plume type is different, leading to different run-out distances. Therefore, to investigate the role of flocculation associated with the TOC pattern in a lake, we design settling and gravity flow experiments with varying salinity and sediment concentration. Considering the flocculated grain size is seldom recorded in deposits, the current study provides a possible mechanism for why sediments with similar grain size deposited at similar water depths could have varying TOC content. This leads to a better understanding of the spatial distribution of carbon storage and the formation of source rocks associated with hydrocarbon reservoirs.

INTRODUCTION

Organic carbon is common over the Earth's surface and deposited in most terrestrial and aquatic environments (Galvez et al. 2020). Lakes serve as one of the significant sinks for organic carbon, characterized by the accumulation of sediments brought in by various sources and processes (Cole et al. 2007; Anderson et al. 2009; Tranvik et al. 2009; Mendonça et al. 2017). The total organic carbon (TOC) present in lake deposits constitutes a substantial reservoir of carbon, contributing to the global carbon cycle (Mendonça et al. 2017; Toming et al. 2020; Duan et al. 2022). Analyzing accumulated sediments, including TOC content, provides valuable insights into past biological activity and environmental conditions, which helps reconstruct a lake's depositional history, including past changes in climate, vegetation, and human activity (Last and Smol 2001; Zhang et al. 2014; Xu et al. 2023). Organic-rich shale in lake deposits is a significant source rock for hydrocarbon exploration (Xu et al. 2023). The determination of TOC is an essential part of screening and evaluating potential petroleum source rocks since the oil and gas potential of a formation correlates with its carbon content (Jarvie et al. 1991). In essence, understanding the dynamics of organic carbon in lakes is crucial for comprehending the global carbon cycle, reconstructing environmental history, and assessing the potential for oil and gas exploration in sedimentary rocks.

TOC in lake deposits is studied for organic matter source and depositional process (Zeng et al. 2017). Notably, the contributions of autochthonous (originating within the system) and allochthonous (external inputs) sources have direct influences on the spatial distribution of TOC in surface sediments (Anderson et al. 2009; Bechtel and Schubert 2009). Regarding the

autochthonous contrinutions, it is generally accepted that water depth (or water column productivity) correlates with total organic carbon in lake deposits (Muller et al. 1979; Calvert et al. 1991; Tyson 2001). Assuming a uniform distribution of organic molecules in the water column, a taller water column in a deep part of a lake contains more suspended organic material than a shallower part and therefore can accumulate more TOC on the lakebed (e.g., Tyson 2001). Sedimentation rate is also known to be positively related to TOC (Ibach 1982; Tyson 2001), but interestingly, there is a threshold in this relationship, past which increased sedimentation rate becomes negatively related to TOC (Ibach 1982; Tyson 2001). In addition to the autochthonous TOC source, allochthonous source (from the river) also contribute to TOC distribution in lakes, especially where autochthonous productivity is low. Therefore, the highest water depth is not always aligned with TOC hotspots in lakes; instead, there can be spatial variations in TOC distribution associated with source locations (Dean and Gorham 1998; Khim et al. 2005; Bechtel and Schubert 2009; Woszczyk et al. 2011; Wang et al. 2012). However, the exact mechanistic explanation for forming TOC hotspots remains unknown. Here in this study, we investigate the role of the flocculation which leads to local hotspots (Piton et al. 2024) can influence the spatial variability in the TOC distribution.

Clay flocculation is a coagulation process that aggregates suspended clay particles in water, leading to the formation of larger flocs (Pejrup 2003). These clay particles often contain organic matter, and thus the flocculation process plays a crucial role in removing both suspended clay particles and the associated organic material. In addition, clay flocculation has been identified as an engineering treatment for toxic marine algal blooms (Anderson 1997). Algae-clay aggregation leads to sedimentation of the suspended load and results in significant removal

of algae from water columns (Avnimelech et al. 1982; Anderson 1997; Sengco et al. 2001; Ho et al. 2022). Therefore, the flocculation process promotes the removal of particulate organic carbon through the formation of larger flocs that are easier to separate from the water and lead to higher TOC in the lakebed.

This flocculation process is sensitive to salinity (Li et al. 2011; Abolfazli and Strom 2023). Chemical and electrostatic clay particle-particle interactions change their rates of interaction and produce different flocculated grain sizes depending on water chemistry (Stumm and Morgan 1995; Furukawa et al. 2009; Mietta et al. 2009). Increased salinity in a lake is known to enhance flocculation rates (Edzwald et al. 1974) and create large muddy aggregates that settle quickly to the lakebed. However, in high salinity, there is also a threshold in this relationship, past which increased salinity becomes negatively related to the extent of flocculation (Pejrup et al. 2010). A salt content, therefore, is likely to affect clay-organic material aggregation and result in variations of TOC in the deposits.

In addition to the relationship between the salinity setting of the lake and flocculation, the extent of flocculation is also important in determining the river plume type (Strum et al. 1978; Sabatier et al. 2022). The extent of flocculation influences the density of the plume and affects the type of the river plume entering the lake, depending on the density of the plume and the standing body of water. The hypopycnal flow occurs when the river water density is lower than the receiving lake water density, leading to surface floating water and suspended sediments. While the hyperpycnal flow occurs when the river plume is denser than receiving lake water, letting the plume plunge downward due to its negative buoyancy (gravity flow). The organic

distribution would be influenced by these two different types of plumes due to a different run-out distance and sediment accumulation process. Thus flocculation, which contributes to the total organic carbon of the sediments by binding the sediment particles and increasing their stability, plays a significant role in lake deposition environments. The flocculation might drive enhanced sedimentation rate and accumulation of organic material, yet the impact of flocculation in the natural depositional process of organic carbon has not been thoroughly investigated.

Therefore, this study aims to understand how flocculation changes the transport and fate of organic carbon influencing where and how these materials are deposited (Nghiem et al. 2022). In this study, we test the following hypotheses: 1) Clay flocculation aggregated with organic molecules is a cause for high TOC in the lake deposits and 2) change in lake salinity changes the flocculation rate and reduces run-out distance of hyperpycnal underflows. We test the hypotheses by grain settling measurements in settling columns and gravity flow experiments in a flume. A range of salinities and sediment concentrations are applied in the settling column experiment to show the effects of salinity and suspended sediment concentration on mud-algae aggregation (i.e., removal of suspended organic material). In the flume experiment, the effects of salinity and current density on flocculation and run-out distance are tested. This two-dimensional gravity flow experiment with mud-algae aggregation is then used to examine the relationship between the boundary conditions (i.e., lake-water salinity and inflow current density) and TOC in the deposits. Using insights from this research, we discuss the mechanistic interpretation of TOC hotspots in Qinghai Lake, the largest saline lake in China, as an example showing that TOC hotspots are not well aligned with its deepest areas.

METHOD

Settling Velocity

A series of settling column experiments were conducted in 1000-ml beakers. In each run, the beakers were filled with 900 ml of water dyed blue at 0.1g/liter using the Pylaklor Brilliant Blue S-566 powdered dye. Salt was added to increase salinity at 1, 1.25, 1.5, 1.75, 2, 2.25, 2.5, 2.75, 3, 3.25, 3.5, 4, 5, and 7% concentrations by weight (Table S1) in the water columns. Thick white paper covered the bottom, two sides, and backside of the beakers to enhance image quality and allow for brightness calibration (Fig. 1). For preparing solutions, bentonite (D~10 μm) sediment was mixed with fresh water at 2, 3, and 4% bentonite concentrations by weight (henceforth B%) and soaked for a day to make sure that the individual particles dispersed and mixed thoroughly with the water before being added to the measurement beakers (Table S1). 100 ml of the well-mixed bentonite solution at a given concentration was added to each beaker. Note that no salt was mixed in the 100-ml bentonite solutions, but the settling columns had different saline conditions. Each measurement was taken for between 4 hours to 1 day to observe flocculation and grain settling by taking time-lapse images. The time-lapse images were taken at 30-60 second intervals. Once a solution was added to a beaker, a layer of suspended bentonite cloud developed near the water surface and gradually settled to the bottom. We captured the top elevation of the cloud using the time-lapse images.

Dye concentration was used as a proxy for uniformly populated algae, with darker colors corresponding to higher algae concentration and lighter colors to lower algae concentration in the water column. Flocculation is a well-known wastewater treatment process for removing dyes even with a significant concentration, and TOC from suspensions (Yu-Li and Thomas 1995;

Bousher et al. 1997; Papic et al. 2004). Previous wastewater treatment studies have demonstrated that the removal of dye and the decrease in organic material in wastewater behave similarly with both time and coagulant dose (comparable to the salinity in the current study). Therefore, optical clarity by comparing averaged blue values in time-lapse images before and after bentonite grains flocculate and settle could serve as a proxy for the change in TOC in the water column. For quantifying the blueness, averaged blue values (0 to 255) in a 200×200-pixel window at a set location were monitored during the measurements (Fig. 1). Another 200×200-pixel window on the background white paper was used to calibrate any potential changes in lighting during each experiment to maintain the consistency in color measurement (Fig. 1).



Figure 1 Settling measurement setup with three beakers taken from run times (RT) = 0, 20, and 300 minutes in R11 (All run parameters can be found in Table S1). Bentonite suspension settles over time and deposits at the bottom of the beaker. Water is dyed blue using Pylaklor Brilliant Blue S-566 powdered dye from the Pylam Products Company, Inc.

A series of flume experiments investigated the effect of salinity on hyper- and hypopycnal flows through flocculation of suspended sediment. The dimensions of the flume were 100 cm long, 30 cm tall, and 1 cm wide (Fig. 2). The flume was submerged and placed up against the front glass wall inside of a larger tank. The upstream 20 cm has a slope at 45° and the rest is flat. The tank was filled with fresh or saline water at 2, 3, and 4% salinities, and 0.025% blue dye concentration by weight. The tank was reset after each experiment for a designed condition. A single current of 200 grams at 0, 1, 2, 3, 4, and 5% B% in fresh water was introduced at the upstream end in each run. Time-lapse images were taken at 10 second intervals for the initial 10 minutes to document the current and its depositional process, and at 60 second intervals for the last 5 hours of each run to capture the settling of remaining suspended sediment. Video was also taken and used a visual measure of average flow velocity. A color card was placed in the image to correct for any potential changes in lighting conditions during experiments. Since the volume of water in the tank was large, blueness in the tank did not change significantly with a single current compared to those observed in the settling column experiments described above. Instead, the final deposits were thick enough that blue color value could be measured directly in the sediment deposit, as a proxy for TOC. Averaged current velocities over the upstream 40 cm were measured using videos taken during the experiments. The run-out distance was decided using the length of the final deposit only for the hyperpycnal flow runs.

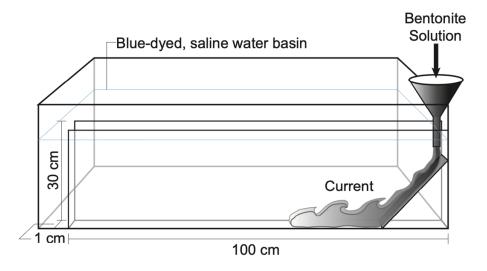


Figure 2 Schematic of the gravidity flow experiment, showing a submerged narrow inner flume and a current generated from the right-end of the basin.

RESULTS

Effect of Salinity and B% on Blueness Changes: Increased Dye Removal by Flocculation with Higher B% and Salinity, and the Threshold Behaviors

Three sets of settling-column experiments are used to indicate the effect of salinity on removing the blue dye from the water and settling it into the deposit (Fig. 3A-3C). Each set uses either a bentonite solution at a bentonite concentration of 2, 3, or 4%. Once a solution is added to the settling column, a suspended sediment cloud forms at the top of the water column and gradually spreads to the bottom. The cloud top also sinks over time but at different rates for different salinities (Fig. 4). In general, low salinity (lighter gray in the plots) induces fast settling velocity in the early stage and the velocity decreases over time, while high salinity (darker gray in the plots) results in slow initial settling but accelerated settling in the later stages. Figure 4, therefore, shows more concave-up shapes (fast to slow transition in the settling velocity) in the time-series data associated with lower salinities, whereas more convex-up shapes (slow to fast transition in the settling velocity) are associated with higher salinities. All settling of suspended

sediment decelerates significantly when the cloud top falls below 30% of the initial elevation, when the process is more likely switch from suspended grain settling to deposit compaction.

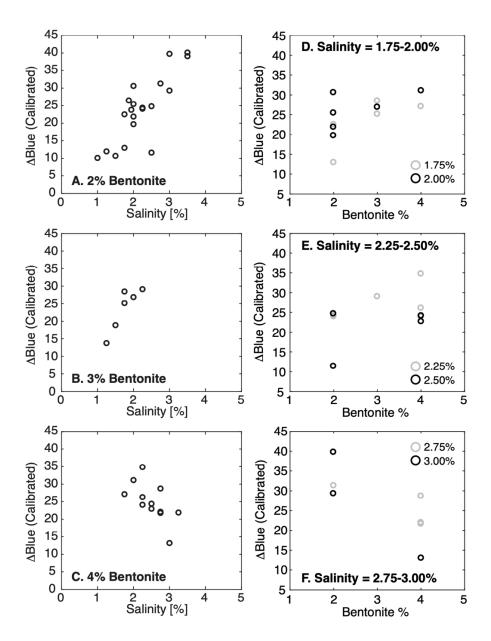


Figure 3 Differences in blue pixel values before and after bentonite suspension settles through the blue-dyed water. (A-C) Δ Blue against salinity under B%s of 2, 3, and 4%, respectively. (D-F) Δ Blue against B% under salinity of 1.75–2, 2.25–2.5, and 2.75–3%, res

When comparing before and after the adding of bentonite to dyed water and settling through the water column, the sets with the bentonite concentrations of 2 and 3% show increases in the blue value differences (Δ Blue) with increasing salinity (Fig. 3A-B). Larger Δ Blue indicates a larger color change from a darker blue to a lighter blue (i.e., more removal of blue dye) in the water. However, using the 4% case reverses the trend; higher salinity reduces Δ Blue (Fig. 3C).

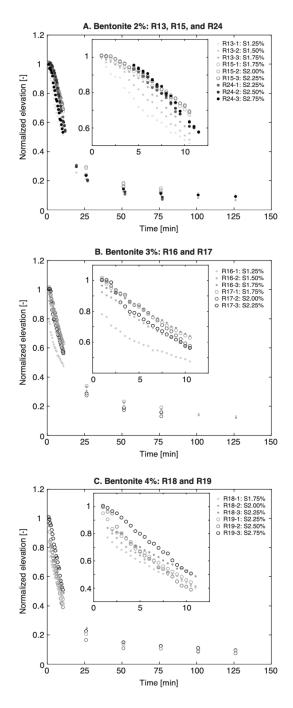


Figure 4 Normalized elevation changes of settling suspended bentonite sediment over time depending on salinity with different Bentonite concentrations at (A) 2%, (B) 3%, and (C) 4%.

Figure 5 shows the elevation changes in the suspended sediment cloud depending on the bentonite concentration. High B% is associated with more concave-up shapes (fast to slow transition in the settling velocity) in the time-series data, whereas low B% is related with more

convex-up shapes (slow to fast transition in the settling velocity) under the same saline condition. Figs 3D-3F show six different salinity conditions against ΔBlue for the bentonite concentrations of 2, 3, and 4%. As B% increases, ΔBlue also increases for the runs with salinity between 1.75% and 2.5% (Figs 3D-3E). However, for salinities of >2.75%, ΔBlue decreases with increasing B% (Fig. 3F) and thus has a reversing trend.

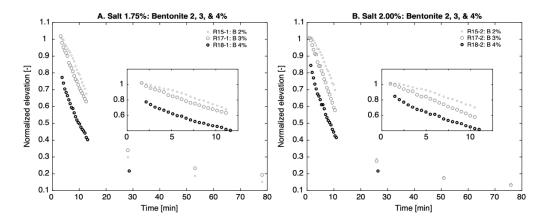


Figure 5 Normalized elevation changes of settling suspended-bentonite sediment over time depending on bentonite concentrations with different salinities at (A) 1.75% and (B) 2%.

Gravity Flow: Shorter Run-out distance of Turbidity Currents by Flocculation with Higher Salinity

Currents in the flume experiments result in either hypo- or hyperpycnal flows depending on the inflowing bentonite concentration and basin salinity (Fig. 6). When the saline water in the basin is denser than the supplied current, a buoyant plume spreads across the water surface. However, over time, sediment gradually aggregates and settles once the flocculated grains become big enough (Fig. 6A). In this case, the settled sediment covers the entire flume length with a nearly uniform thickness (Fig. 6B). For runs with bentonite concentration in suspension that is high enough to overcome the density of the basin water, hyperpycnal underflows are

generated (Fig. 6C). Among these cases, the runs with freshwater in the basin lead to the bottom-hugging currents easily traveling beyond the entire flume length (Fig. 6E). However, unlike the freshwater cases, enhanced flocculation in saline water significantly dampens turbulent mixing and momentum and thus the current velocity decreases quickly. Flocculated coarser grains settle quickly and develop a wedge-shaped deposit that decreases thickness rapidly to the downstream direction, while some remaining finer sediments gradually blanket the earlier, coarser turbidite deposit (Fig. 6D). Therefore, run-out distances of the turbidity currents that flow into saline water are shorter than those in freshwater.

Gravity Flow: Opposite Effects of Salinity and B% on TOC for Turbidity Current vs. Buoyant Plume Cases

Turbidity currents that flow into freshwater show a gradual increase in current velocity with B% (Fig. 6G). Among the 2%-salinity runs, only the currents at 3–5% bentonite concentrations generate hyperpycnal flows and follow a pattern similar to the freshwater cases, showing an increase in current velocity with bentonite concentration. In other words, higher excess density causes faster hyperpycnal flow. This trend is paralleled in the currents at 4–5% bentonite concentrations among the 3%-salinity runs.

For the cases of buoyant plumes, currents usually spread over the top of the water column quicker with lower bentonite concentrations, which is opposite to the cases for hyperpycnal flows. In other words, lighter plumes can slide faster over denser basin water. For hyperpycnal flows at the same bentonite concentration (e.g., 4% bentonite concentrations at 0, 2 and 3% of salinity shown in Fig. 6G), the initial current velocity increases when the basin salinity is at 2%

compared to that in the freshwater, but the initial current velocity significantly decreases for the run with 4% salinity. This also indicates a threshold effect of salinity on the hyperpycnal flow. Run-out distance for hyperpycnal flows increases with higher B% (Fig. 6H). For currents at the same bentonite concentration, those that flow in more saline water show shorter run-out distances (Fig. 6H).

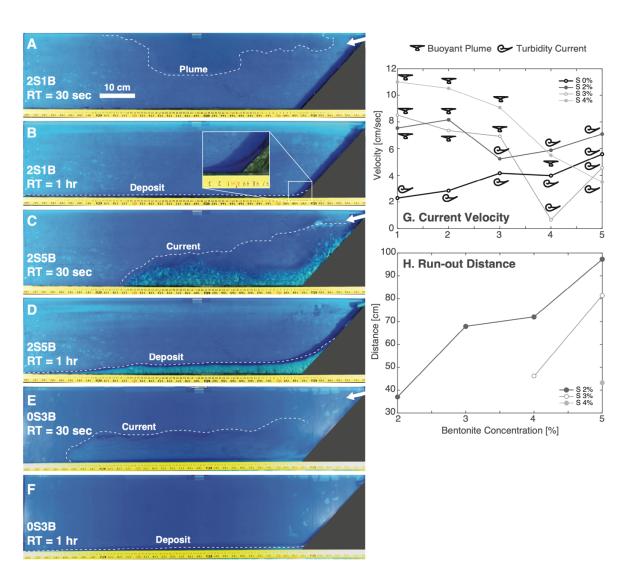


Figure 6 Gravity current experiments. (A) 2S1B at run time (RT) = 30 seconds showing a buoyant plume, (B) the final deposit in 2S1B, (C) 2S5B at RT = 30 seconds showing a turbidity current, (D) the final deposit in 2S5B, (E) 0S3B at run time = 30 seconds showing

Blue values of the final deposit (captured by photographs) are used as proxies of TOC in the deposit (Fig. 7). Higher values represent more organic content in the deposit. For the hyperpycnal-flow runs, currents generated by higher bentonite concentrations reflect deposits with lower blue pixel values and thus less TOC. However, when B% is low so that the current is less dense than the basin saline water, buoyant plumes generated by relatively higher B%s make deposits with higher blue pixel values and thus more TOC. As for the effects of salinity on TOC, the hyperpycnal-current runs into higher salinity (at the same B%) result in lower blue pixel values and thus less TOC in the deposit. Among the runs with buoyant plumes, the trends shown in the measurements (Fig. 7) are not uniform. However, in general, higher salinity produces higher blue values and thus higher TOC is expected. This is also consistent with the settling-column measurements presented in Fig. 3A-3B.

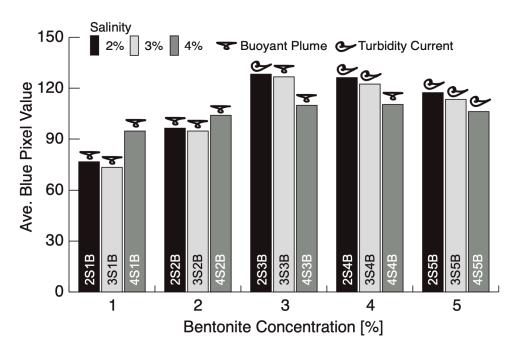


Figure 7 Averaged blue pixel values for runs with 1–5% bentonite concentrations and 2–4% salinity in the basin.

DISCUSSION

Suspension Settling

The current study suggests that during mud grain flocculation, more blue dye (organic particles) can be removed from the water as the grains continuously flocculate and increase in size. Moreover, an increase in salinity promotes mud grain aggregation in the experiments. Previous studies (e.g., Furukawa et al. 2009) documented that clay particles suspended in solutions of higher ionic strength have thinner electrical double layers, and this exposes the surface edges that are more positively charged. The exposed edges aid in grain aggregation with more negatively charged basal planes in other clay particles. Flocculation intensifies with higher salinity chemically in this way, and thus in the experiments. Algal capture could also increase with salinity because the clay particles are absorbed onto more algae surfaces in the water and settled together to the bottom under higher saline conditions, like the experimental observation shown in Fig. 3A-B. However, this applies only when B\% < \-3\% in the current experimental measurements. As B% exceeds \sim 3%, the trend between the salinity and Δ Blue reverses (Fig. 3C). This threshold behavior can be explained as follows: More sediment (i.e., high B%) causes rapid aggregation of clay particles (Tran et al. 2018). Previous experiments have demonstrated that clay particles flocculate with each other and increase in size until they reach an equilibrium size dictated by either the shear induced in the settling process (Hill et al. 1998) or by the turbulent mixing (Burban et al. 1989; Serra et al. 1997; Verney et al. 2011; Strom and Keyvani 2016; Tran et al. 2018). High sediment concentration in the water (B%>~3% for the current experiments) leads flocculating grains to reach the equilibrium size quickly so that the flocculated grains settle without aggregating more with suspended organic particles effectively. As salinity increases, flocculation occurs even more quickly and the grains also reach their

equilibrium sizes in a much shorter period (Edzwald et al. 1974). This process restricts the time for the sediment to capture more organic molecules within the upper water column because coarser grains settle faster and cause the reverse trend (i.e., ΔBlue decreases with salinity). A threshold behavior associated with bentonite concentration (at the salinity ~2.5%) (Fig. 3D-F) can be also explained in a similar way: More densely concentrated bentonite solution can settle quickly and have faster flocculation, which prevents the aggregation of sediment and organic particles.

During a river flood that drains into a lake, the increased amount of fine sediment available to flocculate and aggregate algae would tend to remove biota from the lake water, thereby creating higher TOC deposits in the lakebed. However, above the threshold conditions (B%>~3% and salinity >~2.5% in this experiment), sediment concentration and salinity would be inversely related to TOC in the deposit. It is, therefore, expected that beyond our experimental conditions, for example, very high sediment concentration (B%>~5%) even at a low saline condition, might show a negative correlation with TOC in the deposit because the faster flocculation reduces the time available for clay-algal aggregation. This suggests a rapid mud depositional event may not create favorable conditions for high TOC concentration in the deposit.

Gravity Flow

For the runs with 2% salinity in the basin, hyperpycnal currents flow at a slightly higher velocity compared to those in freshwater (Fig. 6G). Flocculated larger grains under saline conditions in general extract momentum from the current and reduce the velocity. However,

flocculation under this low saline condition has the positive effect of increasing initial flow velocity as coarser grains are generated during early sediment dispersal into the basin and rather add momentum to the density current while collapsing into the deeper basin. Even in this condition, the flocculation continues and eventually slows the current so that the final deposit has a shorter run-out distance compared to those in freshwater. Hyperpycnal currents generated in 3-4% salinity show slower current velocities compared with those in freshwater due to turbulent damping and momentum extraction by rapid flocculation (Fig. 6G), which is also consistent with previous studies (e.g., Packman and Jerolmack 2004). Higher salinity causes faster aggregation of clay particles in the turbidity current, leading to rapid floc settling and proximal deposition of a thicker turbidite. For the buoyant plume cases, lighter currents above denser basin water spread faster. It usually took a few minutes for the plumes to start settling as particles are aggregated mostly because their sediment concentrations are low. The initial current speed is therefore not strongly influenced by the flocculation, but rather depends more on the basin salinity. According to McDonell et al. (2024), who studied mud settling velocity in relation to turbulence and salinity in natural estuarine environments, salinity has little effect on flocculation under low turbulence levels (shear dissipation rate, G < 5 Hz). Similarly, the buoyant plumes in this experiment, experiencing low turbulence levels, are likely minimally influenced by salinity in terms of flocculation. In contrast, turbidity currents under higher turbulence levels can effectively regulate grain settling velocity through the flocculation process, leading to shorter run-out distances and more proximal deposition.

For the hyperpycnal flow cases in this experimental study, reaction time between the basin water – including blue dye (proxy for algae) – and the clay particles in the current is a key

to determining TOC in the deposit. Among experiments with the same bentonite concentrations, higher salinity enhanced flocculation and led to faster depositing sediment and deposits with less TOC (Fig. 7). For example, comparing 2S4B with 3S4B the blue value is higher in 2S4B than 3S4B (Fig. 7). For the hypopycnal flow cases, higher bentonite concentrations caused more available clay particles to aggregate with algae in the water, resulting in higher TOC in the deposit (e.g., comparing between 3S1B, 3S2B, and 3S3B in Fig. 7). The buoyant plumes slowly moved along the surface without strong turbulence to mix the basin water with the plume and had a slower rate of flocculation. In this case, salinity helps to initiate the flocculation process and to aggregate more organic particles with clay so that there is increased TOC in the deposit (e.g., comparing between 3S2B and 4S2B, and between 3S1B and 4S1B in Fig. 7). The 3S3B and 4S3B runs also show hypopycnal flows. Their measured blue values show an opposite trend compared to that observed in most other runs. We would expect that there is a threshold after which increased salinity no longer aids the increase in TOC of the deposit (also shown in Fig. 3F).

The 2S3B experiment produced the highest TOC in the final deposit and resulted from the following conditions: 1) the maximum possible salinity that can yet generate a hyperpycnal flow causing turbulent mixing of saline water and clay particles, promoting flocculation, and 2) the minimum possible sediment concentration that can flow slowly enough to mix longer with the ambient water to capture more algae.

Implications for a Natural Case: TOC hotspots in Qinghai Lake, China

Qinghai Lake, the largest saline lake in China, receives its discharge mainly from the Buha River during the monsoon season (Fig. 8). The lake has a deep depositional center with a nearly uniform 25–30 m depth that extends northwest to southeast. First, based on a previous study of the water depth and TOC distribution (Tyson 2001), TOC is expected to be highest in the deep basin center and reduced in shallower water depths. However, the deep basin centers in Qinghai Lake do not overlap well with areas with high TOC ($> \sim 3\%$). Second, regarding the relationship between sedimentation rate and TOC, the sedimentation rate measured in the southeastern lake (32–77 cm/ky: Ji et al. 2005) exceeds the proposed threshold, above which further increases in sedimentation rate lead to a negative correlation with TOC. The northwestern lake area close to the Buha River, i.e., the main sediment supply in the lake is expected to have even higher sedimentation rates, well above the threshold, compared to the southeastern area where the sedimentation rate was measured. Therefore, the deposits of the northwestern area should have less TOC than those in the southeastern lake. Nevertheless, field measurements show that the TOC hotspots ($> \sim 3.5\%$) are more concentrated in the northwestern parts of the basin (Fig. 8). The previously suggested first-order controls, therefore, did not fully explain the locations of TOC hotspots in Qinghai Lake.

It is challenging to interpret the exact controls on the TOC distribution in natural lake systems. However, based on the insight gained from the current experiments and field observations from Qinghai Lake, we suggest the following large-scale causes for the distribution of TOC in the lake deposits. Before discussing further, here we note a few points to consider when applying the experimental results to Qinghai Lake: The coagulation rates of phytoplankton with clays may be lower than those in our experiments (timescale of ~mins—hours) because the

suspended clay of the Buha River is mainly composed of kaolinite (Lanzhou Institute of Geology CAS, 1979). Kaolinite has a lower cation exchange capacity and is less prone to aggregation than bentonites with significant fractions of Montmorillonite (Keyvani and Strom 2014). Moreover, the salinity in Qinghai Lake has typically been <16 mg/mL (1.6%) throughout the past several centuries (Lanzhou Institute of Geology CAS 1979). The thresholds observed in the experiments cannot be directly applied to natural systems; further investigation into the specific thresholds for salinity and clay concentration in natural environments is required to improve the applicability of the insights from the current study. Nevertheless, this research provides a valuable guideline for interpreting the dynamic processes influencing the distribution of TOC in natural lakes.

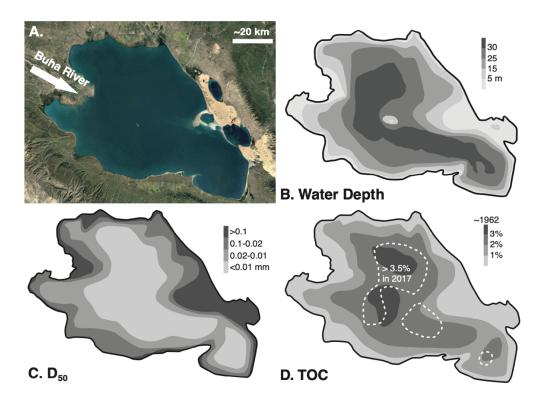


Figure 8 (A) Qinghai Lake, (B) water depth, (C) grain size on the lakebed, and (D) spatial distribution of TOC in 1962 and the dotted contour lines for TOC > 3.5% in 2017.

Flood-induced hypopycnal plumes occur in Qinghai Lake especially during monsoon season (Figs S1 and S2). A part of the plume may plunge into the proximal deeper lake as hyperpycnal underflow, while other parts of the plume follow surficial, wind-driven currents and settle gradually with time. According to wind field data (Chen et al. 2021), Qinghai Lake can be divided into western and eastern regions with prevailing wind from the north and northwest, and from the south and southeast, respectively. Strong wind mixing likely distributes the plumes across the lake. Assuming uniform distribution of suspended sediment by strong surface mixing, settling of clay particles from the littoral to the low-energy profundal zones at the deeper lake center (the latter is usually a favorable site for organic matter) should provide further time for algae-clay aggregation and result in high TOC in the deposit. Figure 8D shows organic-rich areas with TOC >2% that overlap well with the profundal zones. However, organic-rich hotspots with TOC >~3% are do not overlap with the profundal zones. Rather these hotspots are concentrated in the northwestern area near the Buha River outlet. We propose that plunging turbidity currents from the Buha River, with run-out distances shortened due to flocculation, account for this mismatch. Lamb et al. (2020) suggested that mud flocculated in rivers has a nearly constant settling velocity of ~0.34 mm/s independent of the grain size. This is about 4 times faster than would be expected for non-flocculated grains that have a grain size of 0.01 mm, similar in size to the mud found in the profundal zones of Qinghai Lake (Fig. 8C). If the mud were flocculated, one might expect a reduction in the mud transport distance by a factor of four relative to an unflocculated state. Considering the suggested settling velocity for flocculated mud of ~0.34 mm/s for flocs in freshwater, the saline condition in the lake may further support the proximally located TOC hotspots in Qinghai Lake (even though the low salinity may not be a significant factor in shortening mud transport distances as the salinity in Qinghai Lake has been below 16

mg/mL (1.6%) throughout the past several centuries). Additionally, the effect of organic matter itself warrants consideration. Abolfazli et al. (2024) reported that biomatter can enhance both the flocculation rate and the size of equilibrium flocs. This suggests that natural systems with higher organic matter content may experience amplified flocculation, potentially influencing the location of TOC hotspots in ways that differ from estimates based solely on hydrodynamic processes.

Unfortunately, in situ monitoring of flocculation processes and the resulting deposits in natural systems is challenging at the lakebed due to the fragile nature of flocculated particles. Additionally, floc size is rarely preserved in sedimentary records, as only the original clay particle sizes prior to flocculation are typically recorded in the deposits (Fox et al., 2004). This makes it difficult to interpret sediment-transport processes that lead to high TOC content based on particle-size distribution in the deposit. However, insights from the current experimental study offer a plausible explanation for why sediments with similar grain sizes, deposited at comparable water depths, can exhibit varying TOC content.

CONCLUSIONS

High sediment concentration of inflow and salinity in the receiving basin (e.g., lake) cause rapid aggregation of clay particles, binding organic matter in the aggregates, and thus higher TOC in the deposits. However, these trends reverse when sediment concentration and salinity increase beyond threshold values (in this experiment, bentonite concentration at ~3% and salinity at ~2.5%). Beyond the thresholds, it is likely that flocs reach their equilibrium sizes quickly, leaving a relatively shorter amount of time for clay-algal aggregation.

Hyperpycnal currents under saline conditions have slowing velocities due to turbulent damping by rapid flocculation. Thereby producing shorter run-out distances compared to those in freshwater. For the buoyant plumes, the lighter currents spread quickly over the denser basin water, and the initial current speed is not strongly influenced by the flocculation. In the gravity flow experiments, the condition to maximize time and magnitude for mixing of the current with the ambient water to capture more algae was achieved when 1) the highest possible salinity in the basin water that still allows the incoming flow to remain hyperpycnal, and 2) the lowest possible sediment concentration that can reduce the current speed, allowing longer time for interaction between the current and organic particles in the ambient water; both were met.

The TOC hotspots observed in Qinghai Lake do not overlap well into the profundal zones. Instead, these hotspots are located close to the mouth of Buha River. We propose that this mismatch can be explained by increased mud flocculation that enhances algal deposition and settling in the lake leading to shorter run-out distances of the plunging turbidity currents from the river. This flocculation effect can also be enhanced by elevated salinity in the lake, further supporting more proximal located TOC hotspots in Qinghai Lake.

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Supplementary Material

Table S1. Experimental parameters.

Γable S1. Experimental parameters.								
Run	Beaker	Salt % in	Bentonite %	Initial water				
Number	Number	weight	in weight	volume (ml)				
10 10	1 2	2 3.5	2	1000 1000				
10	3	7	2	1000				
11	1	1	2	1000				
11	2	2	2	1000				
11	3	3	2	1000				
12	1	2.5	2	1000				
12	2	3	2	1000				
12	3	3.5	2	1000				
13	1	1.25	2	1000				
13	2	1.5	2	1000				
13	3	1.75	2	1000				
14	1	1.87	2	1000				
14	2	1.94	2	1000				
14	3	2	2	1000				
15	1	1.75	2	1000				
15	2	2	2	1000				
15	3	2.25	2	1000				
16	1	1.25	3	1000				
16	2	1.5	3	1000				
16	3	1.75	3	1000				
17	1	1.75	3	1000				
17	2	2	3	1000				
17	3	2.25	3	1000				
18	1	1.75	4	1000				
18	2	2	4	1000				
18	3	2.25	4	1000				
19	1	2.25	4	1000				
19	2	2.5	4	1000				
19	3	2.75	4	1000				
20	1	2	2	300				
20	2	2	2	600				
20	3	2	2	900				
21	1	2	2	300				
21	2	2	2	600				
21	3	2	2	900				
22	1	2.25	4	1000				
22	2	2.5	4	1000				
22	3	2.75	4	1000				

23	1	2.75	4	1000
23	2	3	4	1000
23	3	3.25	4	1000
24	1	2.25	2	1000
24	2	2.5	2	1000
24	3	2.75	2	1000
29	1	2	2	1000
29	2	3.5	2	1000
29	3	7	2	1000
30	1	3	2	1000
30	2	4	2	1000
30	3	5	2	1000
31	1	2	1	1000
31	2	2	2	1000
31	3	2	4	1000

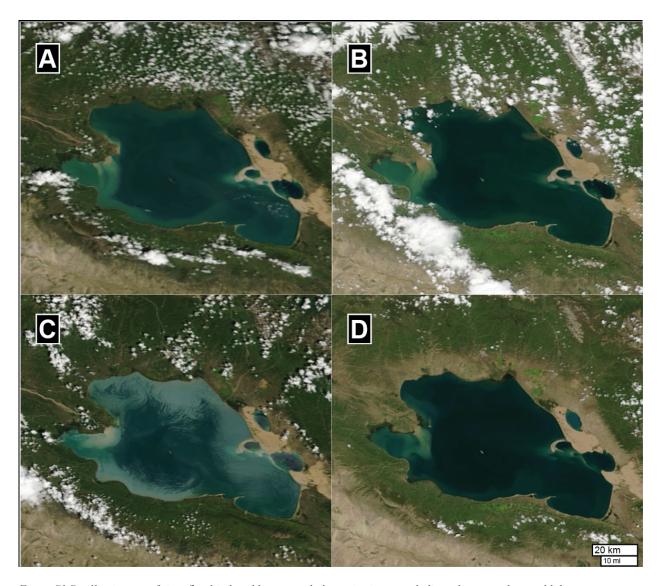


Figure S1 Satellite images of river flood-induced hypopycnal plumes in river mouth, littoral zone, and central lake areas (worldview.earthdata.nasa.gov). Satellite images of Qinghai Lake on (A) 2012-Aug-23, (B) 2015-Jul-05, (C) 2018-Aug-10, and (D) 2021-Jul-12. Surface wind data of Qinghai Lake on 2015-Jul-10 (earth.nullschool.net), indicating the prevailing wind direction of Qinghai Lake was SE (140°), which may trigger a hypopycnal plume move across the lake.

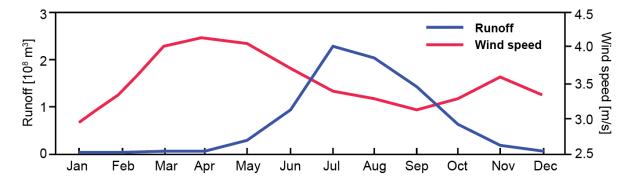


Figure S2. Monthly variations of runoff and wind speed of Qinghai Lake level (Modified after Cui et al., 2017).