

Modelling impact of precipitation of magnesium using lime on removal of select wastewater contaminants

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ABSTRACT

This article is focused on developing mathematical models to describe the impact of $\text{Mg}(\text{OH})_2$ precipitation on select particulate contaminants removals from wastewater. Modeling was achieved first through separating the impact of $\text{Mg}(\text{OH})_2$ precipitate on contaminants removals from impacts of other precipitates and removal mechanisms. Fortunately, the results confirmed that $\text{Mg}(\text{OH})_2$ precipitation occurred above the pH required to form the other major precipitates; namely calcium carbonate and calcium phosphate. Furthermore, the results confirmed that the $\text{Mg}(\text{OH})_2$ impact was a function of the quantity of $\text{Mg}(\text{OH})_2$ precipitate regardless of whether the precipitate was formed through increasing the pH to precipitate available Mg^{2+} , or through adding more Mg^{2+} , or both. Therefore, it was possible to use the quantity of $\text{Mg}(\text{OH})_2$ precipitate to combine and represent the impacts of both, pH and initial Mg^{2+} level. The available data representing removals of TSS, total COD (TCOD), turbidity, color and total phosphorus from domestic and slaughterhouse wastewaters were normalized against the quantities of Mg^{2+} precipitated and logarithmic models adequately ($R^2 = 0.7$ to 0.9) described contaminants reductions from their values remaining prior to $\text{Mg}(\text{OH})_2$ precipitation. The logarithmic models and experimental data confirmed that most of the treatment benefits associated with $\text{Mg}(\text{OH})_2$ precipitation were realized through precipitating relatively small quantities of Mg^{2+} .

Keywords: Lime treatment; Mathematical models; Precipitation of magnesium hydroxide; Seawater brine; Wastewater treatment

1. Introduction

Efficient removal of particulate contaminants from wastewater is a major treatment objective that can be aided by coagulation, flocculation and precipitation and achieved using a variety of techniques, such as sedimentation, filtration and centrifugation. This study is focused on assessing the impact of precipitation of Mg^{2+} as $\text{Mg}(\text{OH})_2$ using lime on removal of selected contaminants from domestic and slaughterhouse wastewaters. Magnesium is an important mineral in rocks and in seawater and researchers used seawater, sea salt brines and other additives as cheap sources of magnesium to treat a variety

of domestic and industrial wastewaters [1–11]. One of the limitations of using seawater as a source of Mg^{2+} is increased effluent salinity. Shanableh [8] used waste brine from an Australian solar table salt production facility as a source of Mg^{2+} for wastewater treatment and reported that the concentrated brine did not significantly increase effluent salinity because of reduced sodium chloride content and increased concentration of Mg^{2+} in the brine. Desalination brines are widely available in the United Arab Emirates and the region and their disposal poses major environmental challenges. Therefore, recovering resources from desalination brines, such as magnesium, may be an attractive option for use in select wastewater treatment applications.

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Treatment brought about by $Mg(OH)_2$ precipitation is generally attributed to the favorable characteristics of the precipitate, which carries a positive charge [11] and has a relatively high particle surface area due to its fluffy, petal-like or amorphous shape [12]. Furthermore, evidence suggests that $Mg(OH)_2$ contributes to organic matter removal through adsorption or co-precipitation [13,14]. Precipitation of Mg^{2+} requires raising the pH above 10, which is typically achieved using lime. Lime is typically used for raising the pH, softening, enhancing, clarification, precipitation of phosphorous, disinfection and removal of color and heavy metals. Calcium from lime reacts with alkalinity and phosphates in wastewater resulting in calcium carbonate and calcium phosphate precipitates.

The experimental results presented in this article were originally intended to study the effectiveness of treatment of domestic and slaughterhouse wastewaters using lime and sea salt brine. Although the impact of precipitation of Mg^{2+} as $Mg(OH)_2$ on removal of particulate contaminants has been extensively evaluated, no known mathematical expressions have been proposed to describe the impact of Mg^{2+} precipitation. Earlier work on domestic wastewater [8] and current work on slaughterhouse wastewater revealed normalized and clear data trends describing removals of particulate contaminants vs. quantity of Mg^{2+} precipitate (Mg_{ppt}). Therefore, the aim of this study was to develop mathematical expressions to describe the impact of precipitation of Mg^{2+} using lime on removal of select contaminants from domestic and slaughterhouse wastewaters.

2. Materials and methods

Jar test experiments were conducted on both the domestic [8] and slaughterhouse wastewaters to precipitate various quantities of Mg^{2+} using lime. External Mg^{2+} was added using concentrated sea salt brine with approximately 90,000 mg/L Mg^{2+} . The brine used to treat the wastewater was either obtained from solar salt production facility in Australia [8] or produced in the laboratory through successive evaporation. During each jar experiment, an equal salt brine dose was added to each of six containers prior to lime addition. Lime was then added at increasing quantities to the containers to increase pH to different levels. The mixtures were then rapidly mixed for 1 min at 150 rpm then slowly mixed at 25 rpm for 15 min to achieve flocculation then settled for 30 min. A portion of the supernatant from each container was siphoned out for water quality analysis.

The initial levels of contaminants whose removals were assessed as a function of the quantity of Mg^{2+} precipitate are presented in Table 1.

The pH was measured in the jars at the end of the settling period. Turbidity was measured immediately after siphoning the supernatant from the jars. A portion of the supernatant was immediately filtered through a 0.45 μm filter for further analysis. The pH was adjusted to between 6.5 and 7.5 in a portion of the supernatant using 10 N H_2SO_4 and the supernatant was used for color, total hardness, Ca^{2+} hardness and soluble phosphorus measurements. Total phosphorus was measured using unfiltered supernatant samples after digestion with sulphuric and nitric acids. All analytical procedures were following standard procedures.

3. Results and discussion

3.1. Precipitation of $Mg(OH)_2$

Precipitation of significant quantities of Mg^{2+} as $Mg(OH)_2$ from water using lime requires raising the pH above approximately 10 to 10.5, depending on Mg^{2+} availability in the water. To assess the impact of the quantity of Mg^{2+} precipitate (Mg_{ppt}) on contaminants removal, we divide total removal (R_T) into two components: (1) removal associated with lime addition to raise the pH up to pH 10 to 10.5 but without significant Mg^{2+} precipitation (referred to herein as R_{Lime}), and (2) removal associated with quantity of Mg^{2+} precipitated (referred to as $R_{Mg_{ppt}}$). Therefore, total removal can be described as in Eq. (1):

$$R_T = R_{Lime} + R_{Mg_{ppt}} \quad (1)$$

Contaminants removal without significant $Mg(OH)_2$ precipitation can be attributed to mechanisms such as gravity settling and precipitation of constituents such as calcium carbonate and calcium phosphate. On the other hand, removals associated with Mg^{2+} precipitation ($R_{Mg_{ppt}}$) can be brought about by enhanced clarification, adsorption or co-precipitation, or due to other removal mechanisms, including pH-related removal mechanisms.

The quantity of Mg^{2+} precipitate (Mg_{ppt}) depends on two independent variables; initial Mg^{2+} available for precipitation (Mg_0) and pH. Therefore, the use of Mg_{ppt} as an indicator of contaminants removals combines the effects of both, Mg_0 and pH, as in Eqs. (2) and (3):

Table 1
Select wastewaters characteristics considered in this study and maximum removals achieved

Parameter	WW type	Initial value	Max R_{Lime} (%)	Max $R_{Mg_{ppt}}$ (%)	Max R_T (%)
TCOD (mg/L)	Slaughter house	5740	40.6	26.4	67.0
TSS (mg/L)	Slaughter house	4860	54.0	29.1	83.1
Color (PtCo)	Domestic	220	69.7	20.8	90.5
Turbidity (NTU)	Domestic	48	56.3	39.1	95.4
$P_{soluble}$ (mg/L)	Domestic	6.0	99.2	0.8	100
P_{Total} (mg/L)	Domestic	8.2	74.9	21.6	96.5

$$R_{Mgpppt} = f(Mg_{pppt}) \quad (2)$$

$$Mg_{pppt} = f(pH, Mg_o) \quad (3)$$

The amount of Mg^{2+} precipitate (i.e., Mg_{pppt}) can be estimated from the difference between initial Mg^{2+} (Mg_o) and residual Mg^{2+} , which is determined by the solubility of $Mg(OH)_2$ as a function of pH. The $Mg(OH)_2$ solubility can be expressed using the well-known solubility-product equation and solubility-product constant (K_{sp}), as shown in Eqs.(4) and (5) below.



$$K_{sp} = [Mg][OH]^2 \quad (5)$$

Substituting for $[OH]$ from the water dissociation equation, $K_w = [H][OH]$, and noting that $K = \text{Log}(K_{sp}/K_w^2)$, Eq. (5) becomes:

$$[Mg] = 10^{(K - 2pH)} \quad (6)$$

For convenience, Eq. (6) can be rewritten in terms of mass concentration of Mg (i.e., mg/L), as follows:

$$Mg = 24,305 \times 10^{(K - 2pH)} \quad (7)$$

The quantity of Mg_{pppt} at any pH can be estimated from Mg_o and soluble Mg [Eq.(7)], as follows:

$$Mg_{pppt} = Mg_o - 24305 \times 10^{(K - 2pH)} \quad (8)$$

When lime is used to raise the pH, the precipitation of $CaCO_3$ is typically limited by available carbonate alkalinity. Similarly, calcium phosphate precipitation is limited by the availability of reactive phosphate in solution. Modeling the impact of $Mg(OH)_2$ precipitation requires separating removal associated with quantity of Mg^{2+} precipitate (R_{Mgpppt}) from removal associated with lime addition (R_{Lime}). Fortunately, R_{Mgpppt} typically occurs above the pH required to precipitate the other major precipitates, such as calcium carbonate and calcium phosphate, which makes it possible to separate the impact of R_{Mgpppt} from R_{Lime} .

The data in Fig. 1a show the experimental solubility of $Mg(OH)_2$ in the two wastewaters tested for a variety of Mg_o concentrations (added Mg^{2+} plus Mg^{2+} originally in the two wastewaters). The solubility curves reveal that unless initial Mg^{2+} is unpractically high, the minimum pH required for Mg^{2+} precipitation is above 10.5. From the data in Fig. 1, the experimental K value in Eq. (7) was estimated to be 18.9, such that:

$$Mg = 24,305 \times 10^{(18.9 - 2pH)} \quad (9)$$

The data in Fig. 1b show that precipitating a certain quantity of $Mg(OH)_2$ can be achieved through either increasing the pH at a given Mg^{2+} level, or increasing Mg^{2+} at a given pH, or both. Fortunately, the results confirmed that the same $Mg(OH)_2$ -associated treatment efficiency can be achieved regardless of how the desired quantity of

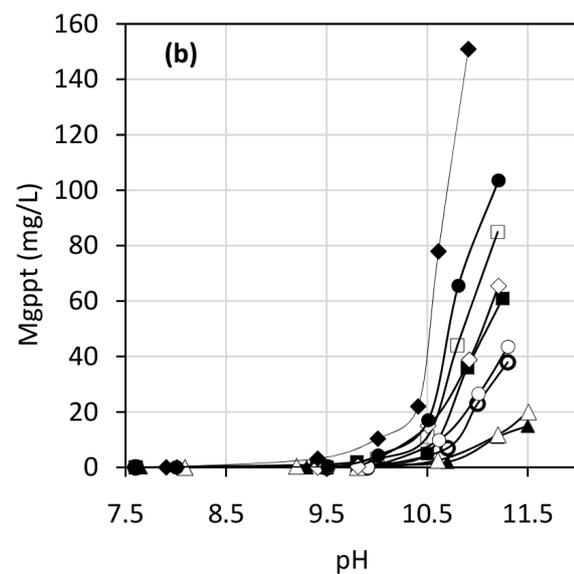
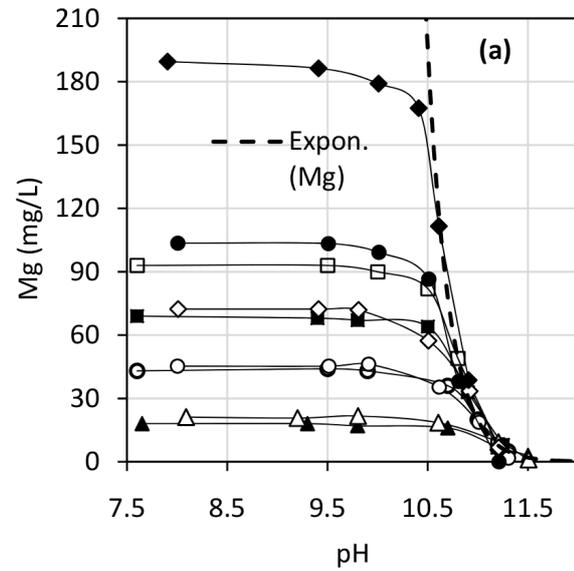


Fig. 1. Solubility of Mg^{2+} according to Eq. (7) and precipitation of $Mg(OH)_2$ starting with various Mg_o values.

$Mg(OH)_2$ precipitate was formed, which allows modeling R_{Mgpppt} as a function of the quantity of Mg^{2+} precipitate.

3.2. TSS Removal as a function of Mg^{2+} precipitate

The TSS reduction from slaughterhouse wastewater as a function of pH is shown in Fig. 2a and as a function of Mg_{pppt} in Fig. 2b. Using lime to raise the pH to precipitate Mg^{2+} and PO_4^{3-} also results in precipitation of calcium carbonate and possibly other substances. However, the precipitation of calcium carbonate and calcium phosphate is virtually complete at pH values less than those required for $Mg(OH)_2$ precipitation. Therefore, it is possible to separate the effect of $Mg(OH)_2$ precipitation from the effects of calcium carbonate and calcium phosphate precipitation. The data in Fig. 2a show that significant TSS reduction was achieved

between pH 7.5 and pH 10.5 without significant $\text{Mg}(\text{OH})_2$ precipitation. The data scatter in Fig. 2a disappeared in Fig. 2b as the TSS reduction data were normalized against the quantity of Mg^{2+} precipitate (i.e., Mg_{ppt}), with Mg_{ppt} quantity accounting for the effects of both, pH and initial Mg^{2+} (i.e., Mg_0), on TSS removal, as indicated in Eq. (3).

The data in Fig. 2b show a clear logarithmic TSS decline trend with Mg_{ppt} quantity. The observed logarithmic decline trend suggests that most of the benefits of Mg^{2+} precipitation can be achieved through precipitating relatively small quantities of Mg^{2+} (i.e., less than 30 mg/L) and that precipitation of large quantities of Mg^{2+} does not significantly contribute to significant additional TSS removal. The model that best describes the remaining TSS as a function of Mg_{ppt} quantity [i.e., R_{Mgppt} in Eq. (1)], excluding TSS removal at pH values less than 10.5 [i.e., excluding R_{Lime} in Eq. (1)] is presented in Eq. (10):

$$\text{TSS} = 2,630 - 1,000 \times \text{Log}(\text{Mg}_{\text{ppt}}) \quad (R^2 = 0.9) \quad (10)$$

The data in Table 1 indicate that the maximum TSS removal associated with Mg precipitation (R_{Mgppt}) reached 29% compared to 54% associated with lime addition (R_{Lime}), such that the maximum TSS removal (R_T) reached 83%.

3.3. TCOD Removal as a function of Mg^{2+} precipitate

Total chemical oxygen demand (TCOD) consists of a soluble component and a particulate component. The removal of the particulate component generally follows TSS removal, as the TSS particles contribute to TCOD. Furthermore, precipitating $\text{Mg}(\text{OH})_2$ may contribute to soluble COD removal through adsorption and co-precipitation. The reduction in TCOD from the slaughterhouse wastewater as a function of pH is shown in Fig. 3a, b as a function

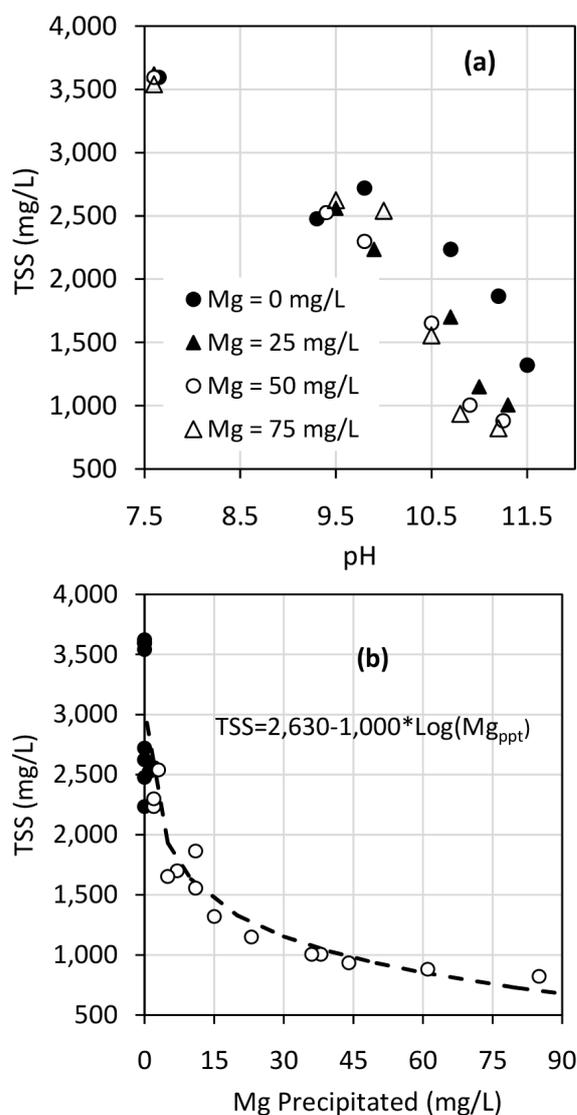


Fig. 2. TSS reduction from slaughter house wastewater as a function of pH (a) or Mg^{2+} precipitated (b). Legend shows external Mg^{2+} dosages.

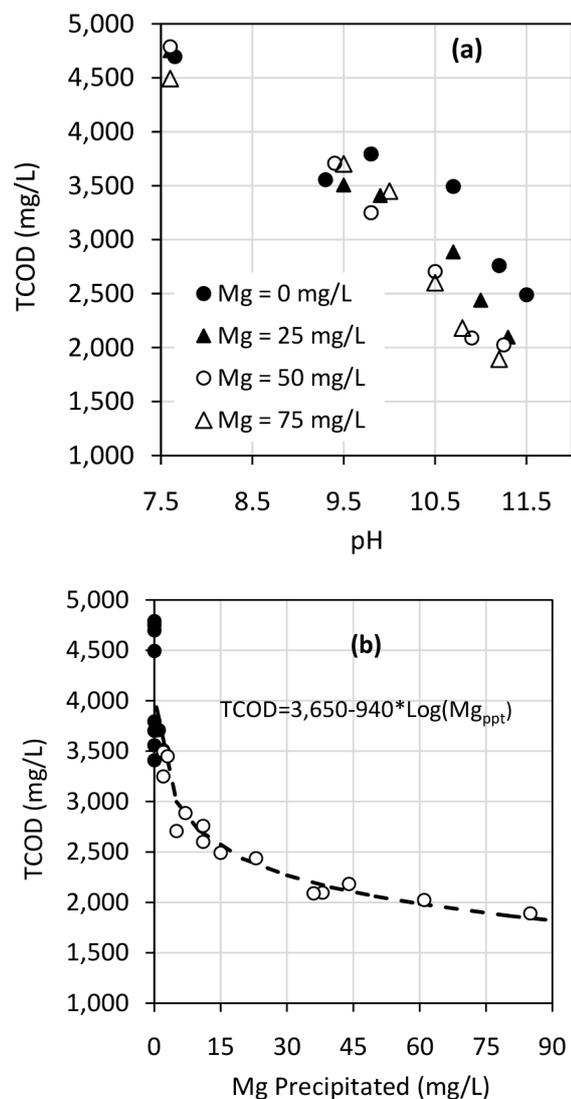


Fig. 3. TCOD Removal from slaughter house wastewater as a function of pH (a) or Mg^{2+} precipitated (b). Legend shows external Mg dosages.

of quantity of Mg_{ppt} . As with TSS removal, the data in Fig. 3a show significant TCOD reduction between pH 7.5 and pH 10.5 without significant Mg^{2+} precipitation. The data in Table 1 indicate that the maximum TCOD removal associated with Mg precipitation ($R_{Mg_{ppt}}$) reached 26% compared to 41% associated with lime addition (R_{Lime}), such that the maximum TCOD removal (R_T) reached 67%.

The data in Fig. 3b show a clear logarithmic decline in TCOD concentration with increased Mg^{2+} precipitation. The logarithmic reduction trend is described in Eq. (11) below:

$$TCOD = 3,650 - 940 \times \text{Log}(Mg_{ppt}) \quad (R^2 = 0.9) \quad (11)$$

3.4. Turbidity removal as a function of Mg^{2+} precipitate

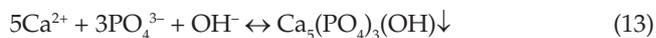
Precipitating $Mg(OH)_2$ particles favorably interact with turbidity particles in solution resulting in effective turbidity removal. The data presented in Fig. 4a show a dramatic decline in residual turbidity as the pH increased above approximately 9.5. The normalised data in Fig. 4b show the impact of $Mg(OH)_2$ precipitation, which is manifested by an logarithmic decline in turbidity to below 5 NTU. The data in Table 1 indicate that the maximum turbidity removal associated with Mg^{2+} precipitation ($R_{Mg_{ppt}}$) reached 39% compared to 56% associated with lime addition (R_{Lime}), such that the maximum TSS removal (R_T) reached 95%. The logarithmic decline trend suggests that precipitating more than 30 mg/L Mg did not significantly contribute to additional turbidity removal. It is important to limit the quantity of Mg^{2+} precipitate, not only to reduce the cost, but also due to its unfavorable gelatinous nature and poor settling characteristics.

The observed logarithmic reduction in residual turbidity is described in terms of the quantity of Mg precipitated as in Eq. (12). The correlation is generally positive, as indicated by $R^2 = 0.7$, however it is rather poor, which suggests that turbidity removal may be influenced by factors other than Mg^{2+} precipitation.

$$Turbidity = 18.5 - 7.1 \times \text{Log}(Mg_{ppt}) \quad (R^2 = 0.7) \quad (12)$$

3.5. Phosphorus removal as a function of Mg^{2+} precipitate

Lime precipitation of phosphates results in the formation of calcium hydroxyapatite according to the following reaction:



In the above reaction, precipitation increases as the pH increases above 8 and becomes virtually complete above pH 10. As soluble phosphorus ($P_{Soluble}$) removal occurs below pH 10.5, the precipitation of Mg^{2+} , which typically occurs above pH 10.5, does not contribute to soluble phosphorus removal. The independence of soluble phosphorus removal from Mg^{2+} precipitation is clearly demonstrated by the data presented in Fig. 5a. On the other hand, soluble phosphorus removal is pH dependent, as shown in Fig. 5b, and the model that describes soluble phosphorus removal above pH 9 is presented in Eq. (14).

$$P_{Soluble} = 2.18 \times 10^{11} \times 10^{1.19 \times pH} \quad (R^2 = 0.9) \quad (14)$$

As the majority of total phosphorus (P_{Total}) in the wastewater was soluble, the total phosphorus reduction as a function of pH was highly influenced by the pH, Fig. 5c. On the other hand, particulate phosphorus removal requires effective clarification and Mg^{2+} precipitation improves the removal of particulate matter, including those containing phosphorus, as shown in Fig. 5d. The logarithmic total phosphorus removal trend in Fig. 5d that best describes total phosphorus removal as a function of Mg^{2+} precipitation in Fig. 5d is presented in Eq. (15) below:

$$P_{Total} = 2.9 - 1.4 \times \text{Log}(Mg_{ppt}) \quad (R^2 = 0.8) \quad (15)$$

The data in Table 1 indicate that soluble phosphorus removal associated with Mg^{2+} precipitation ($R_{Mg_{ppt}}$) was less than 1% compared to 99% associated with lime addition

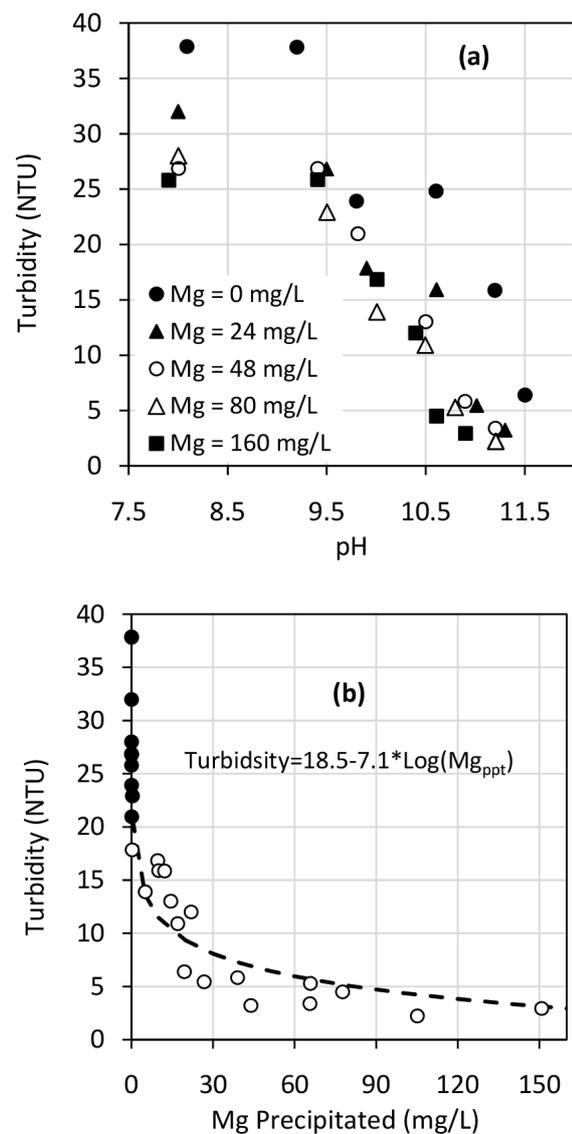


Fig. 4. Turbidity removal from domestic wastewater as a function of pH (a) or Mg precipitated (b). Legend shows external Mg dosages.

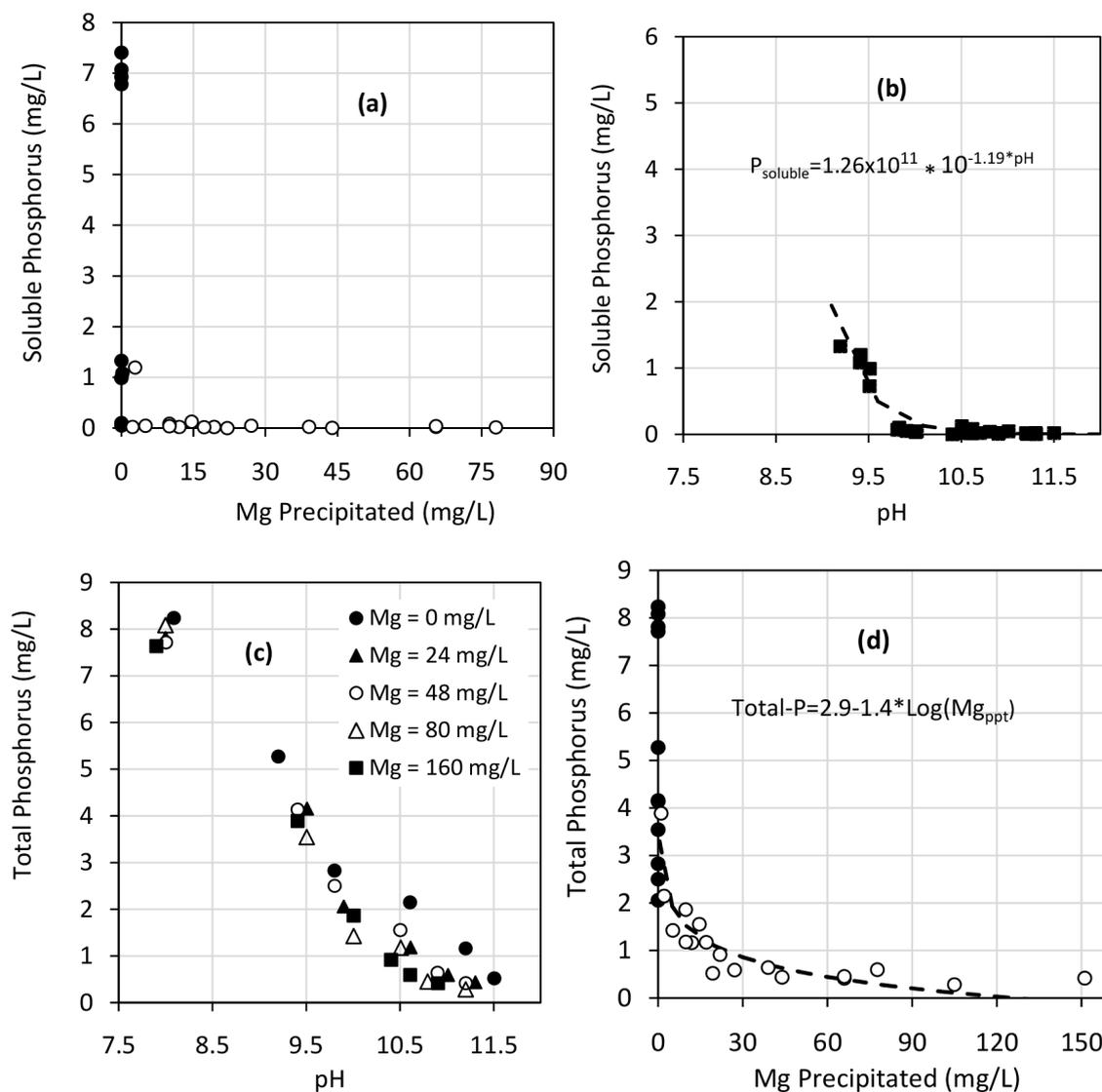


Fig. 5. Soluble and total phosphorus removal from domestic wastewater as a function of pH (b and d) or quantity of Mg_{ppt} (a and c). Legend shows external Mg dosages.

(R_{Lime}). The maximum total phosphorus removal associated with Mg^{2+} precipitation ($R_{Mg_{ppt}}$) reached 22% compared to 75% associated with lime addition (R_{Lime}), such that the maximum total phosphorus removal (R_T) reached 97%.

3.6. Color removal as a function of Mg^{2+} precipitate

Color removal can be attributed to precipitation, coagulation and adsorption onto precipitating particles, in addition to possible precipitation of trace color-causing substances. Furthermore, the pH can impact color and therefore, color measurement was conducted on filtered samples after adjusting the pH to near neutrality. The data in Fig. 6a show that significant color removal occurred below pH 10.5, with additional removal achieved above pH 10.5 together with Mg precipitation (Fig. 6b). The maximum color removal associated with Mg precipitation ($R_{Mg_{ppt}}$) reached 21% compared to 70% associated with lime

addition (R_{Lime}), such that the maximum TSS removal (R_T) reached 91%. The color reduction trend above pH 10.5 as a function of Mg^{2+} precipitation is presented in Eq. (16). On the other hand, color reduction above pH 9 as a function of pH is presented in Eq. (17). The data show that both relationships, color vs. Mg_{ppt} and color vs. pH, show positive to very good correlations, as indicated by the R^2 values.

$$\text{Color} = 71.3 - 24.5 \times \text{Log}(Mg_{ppt}) \quad R^2 = 0.7 \quad (16)$$

$$\text{Color} = 2.18 \times 10^4 \times 10^{0.23 \times \text{pH}} \quad R^2 = 0.9 \quad (17)$$

A summary of the various models that describe the removal of TSS, TCOD, color, turbidity, $PO_4\text{-P}$ and Total-P as functions of either Mg_{ppt} quantity or pH is presented in Table 2.

Although the models presented in Table 2 are specific to the experimental data described in this study, the

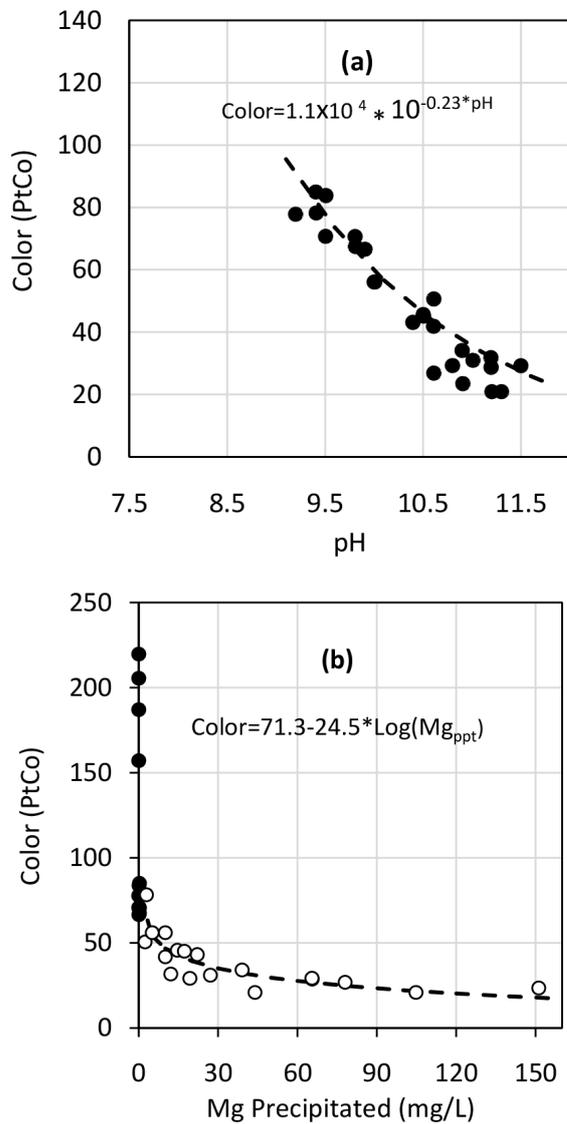


Fig. 6. Color removal from domestic wastewater as a function of pH (a) and quantity of Mg_{ppt} (b).

modeling approach can be applied to other experimental data involving precipitation of magnesium to remove wastewater contaminants. The general applicability of the modeling approach stems from the observed correlations between removal of particulate contaminants and quantity of Mg(OH)₂ precipitate. The observed logarithmic reductions of contaminants from their initial values reflects the logarithmic precipitation trend of magnesium.

Inspection of the R² values in Table 2 suggest that the goodness of the presented models in representing the experimental data ranged between moderate (R² = 0.7) to very good (R² = 0.9). It should be noted that there are two types of models presented in Table 2: (1) models describing contaminants (i.e., color and PO₄-P) removals as a function of pH; and (2) models describing contaminants (i.e., TSS, turbidity, TCOD, and Total-P) removals as a function of quantity of Mg precipitate. For color, the data in Table 2

Table 2
Summary of models developed in this study

Parameter	Wastewater type	Model	R ²
TSS	Slaughterhouse	TSS = 2,630–1,000 × Log(Mg _{ppt})	0.9
TCOD	Slaughterhouse	TSS = 3,650–940 × Log(Mg _{ppt})	0.9
Turbidity	Domestic	Turbidity = 1 8.5–71 × Log(Mg _{ppt})	0.7
Color	Domestic	Color = 1.1 × 10 ⁴ × 10 ^{-0.23 × pH}	0.9
Color	Domestic	Color = 71.3–24.5 × Log(Mg _{ppt})	0.7
P _{Total}	Domestic	P _{Total} = 2.9–1.4 × Log(Mg _{ppt})	0.8
P _{Soluble}	Domestic	P _{Soluble} = 1.26 × 10 ¹¹ × 10 ^{-1.19 × pH}	0.9

suggest that describing removal as a function of pH (R² = 0.9) is more appropriate than describing it as a function of quantity of Mg precipitate (R² = 0.7). For turbidity, the relatively low R² value of 0.7 suggests that factors other than quantity of Mg precipitate influenced turbidity removal significantly.

4. Summary and conclusions

Modeling the impact of Mg(OH)₂ precipitation on removal of TCOD, TSS, turbidity, color, and total phosphorus was achieved first through separating removals associated with lime addition (R_{Lime}) from removal associated with Mg(OH)₂ precipitation (R_{Mgpppt}), then finding the best model that fitted the experimental data. Logarithmic models adequately (R² = 0.7–0.9) represented reductions of the various contaminants from their levels remaining prior to Mg(OH)₂ precipitation. Color removal was dependent on both, pH and Mg precipitation, and models describing color removal dependence on both Mg²⁺ precipitation and pH were presented. On the other hand, soluble phosphorus (i.e., PO₄-P) removal was independent of Mg(OH)₂ precipitation. The logarithmic nature of the models confirmed that precipitation of excessive quantities of Mg was not needed to achieve efficient clarification, which helps reduce Mg sludge production. The removals associated with Mg(OH)₂ precipitation reached 26.4% TCOD, 29.1% TSS, 39.1% turbidity, 20.8% color, and 21.6 total phosphorus and the total removals (i.e., R_T = R_{Lime} + R_{Mgpppt}) reached 67% TCOD, 83.1% TSS, 95.4% turbidity, 90.5% color, 96.5% total phosphorus and almost 100% soluble phosphorus.

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