Towards Enhanced Chlorine Control: Mathematical Modeling for Free Chlorine Kinetics During Fresh-cut Carrot, Cabbage and Lettuce Washing

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A B S T R A C T

In this study, we developed a novel produce-specific mechanistic model to predict free chlorine (FC) dynamics during washing of disk-cut carrots, cut cabbage, and cut iceberg lettuce, in 3 L and 50–100 L tanks, and of shredded iceberg lettuce in 3200 L pilot-plant trials. Ranges for two key parameters: β (L mg^{-1} min^{-1}) the apparent reaction rate constant of FC with produce constituents, and γ, the fraction of the increase of chemical oxygen demand (COD) contributing to the reaction, were determined at the 3 L scale. For disk carrots $\beta \in [0.05, 0.09]$ and $\gamma \in [0.054, 0.078]$, for cut cabbage $\beta \in [0.05, 0.10]$ and $\gamma \in [0.09, 0.12]$, and for cut iceberg lettuce $\beta \in [0.03, 0.06]$ and $\gamma \in [0.07, 0.14]$. Taking values from these ranges the model was able to consistently predict experimental FC dynamics (decay and replenishment), indicating robustness of the apparent reaction rate constants across scales. Comparing sequential changes in COD with turbidity and total dissolved solids (TDS) relative to produce washing rates, our results also illustrate that turbidity and TDS may not be reliable predictors of FC decay rates across produce types and experimental scales. In concert with future experiments, these models could serve as important tools aimed at validating FC compliance within operational limits as well as guiding large-scale commercial experiments focused on improving chlorine management strategies relevant for industry.

1. Introduction

As globalization has broadened the fresh produce supply chain and increased its complexity, more sophisticated methods of surveillance are needed to ensure the safety of fresh produce. Specifically, the produce washing juncture is a critical control point that has received much attention (Gil et al., 2009). Despite this, current understanding of the dynamics of sanitizer control during washing has still been limited. The most common sanitizer used by the industry to wash fresh produce in the United States is chlorine in the form of diluted sodium or calcium hypochlorite (Gombas et al., 2017). In order to maximize the disinfectant efficacy of hypochlorite *via* hypochlorous acid (HOCl), it is crucial for produce wash plants to maintain wash solutions at a slightly acidic pH [$\geq$ 4 as off-gassing for such pH levels may occur (Shen et al., 2013)]. In tandem, maintaining a minimal FC concentration for enough contact time is vital [see discussion in (Gombas et al., 2017)]. While dramatically increasing the FC concentration in wash water may result in enhanced pathogen control, it can result in rapid depletion of FC if large concentrations of reactants are present, as well as the formation of harmful disinfection by-products. Two recent studies have shown that this range of FC, to maximize microbial control on one hand and minimize disinfection by-products on the other, depends on multiple factors including, but not limited to the type and cut of produce being washed as well as the wash time (Garrido et al., 2019; Tudela et al., 2019b).

Considering the importance of maintaining such a balanced concentration of FC that is large enough to prevent cross-contamination during washing, many recent studies have provided correlative relationships between water quality parameters (e.g., total dissolved solids (TDS), turbidity, etc.) and chlorine demand as well as FC levels (Chen and Hung, 2018; López-Gálvez et al., 2019; Luo et al., 2018; Murray et al., 2018; Teng et al., 2018; Weng et al., 2016). In terms of chlorine sanitization, the residual FC concentration is critical in controlling pathogen inactivation and preventing cross-contamination (Beuchat et al., 2004; Gombas et al., 2017; Gómez-López et al., 2014; Luo et al., 2011; Shen et al., 2013). Maintaining such FC levels, however, is challenging as the continuous rise in organic load has been implicated as the primary consumer of FC during wash cycles. While

there are a few commercially-available chlorine management systems with automatic control on FC and pH regulation, due to differences in processing rate, produce & cut types, produce to volume ratios, and wash times across facilities in the industry, more insight towards chlorine control is needed (Tudela et al., 2019a). Thus, understanding the dynamic interactions between organic load and FC concentration is critical to develop practical sanitization strategies for maintaining safety of fresh-cut produce (Gil et al., 2009; Munther et al., 2015; Shen et al., 2013). Studies have demonstrated that COD is a good indicator of organic load at both the lab and pilot scales (Luo et al., 2012; Munther et al., 2015; Rice and Bridgewater, 2012; Van Haute et al., 2013; Zhou et al., 2014), but is impractical to measure during real-time processing (Zhou et al., 2014).

Therefore, several research groups have currently focused on identifying parameters which can predict and monitor FC levels in real-time for specific produce commodities. While many studies (Barrera et al., 2012; Chen and Hung, 2016, 2018; Pirovani et al., 2001; Van Haute et al., 2018, 2013) have examined the correlative strength of various water quality parameters such as UV254, turbidity, and TDS, more understanding is needed with regards to chlorine decay rates and produce type. This is because dosing and monitoring standards built on such correlations alone do not capture the scope of dynamic interactions between FC levels and organic material. Besides, considering the variations in scale of operation and applications in industry, this methodology may be difficult to generalize even when washing the same commodity.

Considering these issues, the objectives of this work are to first develop and validate a produce-specific (for cut carrots, cut green cabbage, and cut iceberg lettuce) mathematical model of FC dynamics that is consistent regarding FC replenishment as well as across multiple experimental scales (lab to pilot). The second aim is to determine whether TDS or turbidity are consistent predictors of FC decay (and hence FC levels) across experimental scales (lab to pilot) for cut carrots, green cabbage and iceberg lettuce. Finally, a discussion is presented on how the developed FC model could play an important role in validating FC compliance as well as in aiding experiments at the commercial-scale geared towards collecting key data for developing mathematical control strategies to optimize chlorine management during washing.

2. Materials and methods

2.1. Single-batch wash experiment and the initial model

Carrots (imperator type), green cabbage, and iceberg lettuce were purchased from a local supermarket and stored at 4 °C and used for experiments on the same day of purchase. Exterior leaves of the cabbage and lettuce were trimmed out and discarded. Cut specifications for cabbage and lettuce were 25.4 mm × 25.4 mm, and disk carrots were 6.35 mm thick.

Experiments for single-batch washing were carried out in a 3 L wash water system. Before starting each experiment, 1.5 mL of concentrated (4.5 %) sodium hypochlorite (BCS Chemicals, Redwood City, CA) was added to the wash tank to achieve approximately 25 mg/L of FC in the wash water (see Fig. 1 for FC data). The pH was simultaneously adjusted to 6.5 using 1 mol.L^{-1} citric acid. 100 g of cut produce were put into a sieve, submerged in the 3 L of tap water (20 °C), and washed (via manual agitation) for 30 s. The sieve was held over the 3 L wash water for another 30 s in order to minimize water loss from the system. This procedure was repeated three times for individual washes of disk carrots, cabbage, and lettuce, respectively.

Water quality parameters (FC, pH, and COD) were measured just before and after washing, as well as at 1, 2, 5, 10 and 20 min following the introduction of produce into the water. FC was measured via a DPD (N,N-Diethyl-p Phenylenediamine) method using a Chlorine Photometer (CP- 15, HF Scientific Inc., Ft. Myers, FL). The pH was measured using a digital pH meter. The COD was determined using a
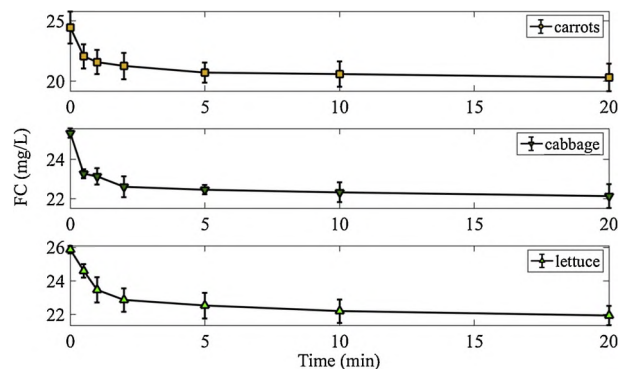

Fig. 1. FC decay data from 100 g single wash experiments in 3 L of water for disk carrots, cut cabbage, and cut iceberg lettuce, respectively. Data presented were the mean ± standard deviation of 3 replicates (with respect to each produce type). Note that COD values were measured at the same times as the FC data presented; following the introduction of produce, after 0.5 min, the COD values did not change for respective produce types.

reactor digestion method as detailed by Luo (Luo, 2007).

Most chlorination reactions can be formulated as $HOCL + B \rightarrow Products$, where B is an organic or inorganic compound (Deborde and Von Gunten, 2008). While there may be many constituents that react with chlorine during a produce wash, following the single wash experiment results for each produce type, we assume the chlorination reactions can be modeled via an averaged second-order reaction relative to the concentration of fresh-cut produce constituents in the wash water (Deborde and Von Gunten, 2008), and a first-order reaction describing FC decay in tap water (Hua et al., 1999). Note that while we do not know the concentration of the produce constituents reacting with FC, we estimate this concentration as a fixed fraction (γ) of the increase in COD due to washing a single batch of produce (for 30 s). Based on the single wash data and these assumptions, we model the FC decay as follows:

$$\frac{dC}{dt} = -\beta RC - \lambda C \tag{1.1}$$

$$\frac{dR}{dt} = -\beta RC \tag{1.2}$$

$$C(0) = C_0, R(0) = \gamma(\Delta COD) \tag{1.3}$$

where C (mg/L) is the FC concentration in the wash water at time t, R (mg/L) is the concentration of the reactant(s) coming from the produce at time t, β (L. mg^{-1}. min^{-1}) is the apparent second-order rate constant, λ (min^{-1}) is the first-order decay rate, ΔCOD (mg/L) represents the difference in COD prior to and after washing a single batch of cut produce, and γ is the fraction of COD increase in the wash water due to washing one batch of produce. We used γ to determine the initial concentration $R(0)$ of the reactant(s) entering the wash water from the cut produce. Related models in the context of produce washing were developed by (Munther et al., 2015) and in the context of drinking water by ((Kohpaei and Sathasivan, 2011) and references therein). We point out that the above model (equations 1.1–1.3) is distinct from the model in (Munther et al., 2015) regarding the dynamics of FC consumption with respect to the reactant(s) from the produce. In particular, from equations 1.2 and 1.3, one can see that R(t) (mg/L), for each $t > 0$, is not proportional to ΔCOD.

2.2. Parameter determination

Using our model (equations 1.1–1.2) and the assumption that reaction rate between R (mg/L) and C (mg/L) dominates FC consumption when R is present in the wash water, it follows that the change in FC concentration equals the change in reactant(s) concentration, i.e.

Table 1
Parameter range results from single-batch wash experiments. Parameter ranges were calculated by comparing model (equations 1.1–1.3) outputs with FC data across three single-batch wash experiments with respect to each produce type.

Produce	β (L mg^{-1} min^{-1})	γ
Disk Carrots	[0.05, 0.09]	[0.054, 0.078]
Cabbage	[0.05, 0.10]	[0.09, 0.12]
Iceberg lettuce	[0.03, 0.06]	[0.07, 0.14]

$\Delta C = \Delta R$. Using this relationship, the assumption that R (mg/L) is completely depleted within 5 min during the single wash experiments (see Fig. 1 and the discussion in section 3.1), and Eq. 1.3, the parameter γ can be calculated as $= \frac{C(0) - C(5)}{\Delta COD}$.

In order to determine the parameter β (L. mg^{-1}. min^{-1}), let D_k represent the data measurements (FC level in mg/L) at time t_k and let $C(t_k, X)$ signify the model output (FC level in (mg/L)) given parameter vector $X = [\gamma \beta]^T$ at time t_k (where γ is fixed by using the formula above). The calculated residuals are $e_{k,X} = D_k - C(t_k, X)$. To find the parameter β that provides the best model fit to the data, we use the *fminsearch* function, in MATLAB (MATLAB 2018b, The MathWorks, Natick, MA) to minimize the 2-norm of the function F defined as:

$$\|F(X)\|_2 = \left(\sum_k e_{k,X}^2 \right)^{1/2}$$

In order to account for the possibility of multiple minima, a multi-start procedure was utilized, dividing the interval $[1 \times 10^{-3}, 1 \times 10^{-1}]$ into n = 30 subintervals to specify initial guesses for β. Note also that model outputs were calculated using the ode45 solver in MATLAB (MATLAB 2018b, The MathWorks, Natick, MA). Table 1 gives the results in terms of parameter ranges for both parameters relative to produce type.

Recall that λ (min^{-1}) is assumed to be the first order decay rate of FC in tap water. It has been previously found that across a range of temperatures and time points, the bulk decay constant for FC in tap water is on average approximately 0.002 min^{-1} (Hua et al., 1999). This value is similar to the range of values for λ across our single wash experiments for varying produce types (using data from Fig. 1 from 5 min onward). Due to this and because the model output governing FC concentration is not sensitive to λ at this order of magnitude, we fixed $\lambda = 0.002$ min^{-1} for the rest of the study herein.

2.3. Multi-batch wash experiments and extended model

To test whether the model (equations 1.1–1.3) captures the essential dynamics involved in FC decay, it was first calibrated using data from single-wash experiments. That is, parameter ranges for β and γ were determined as mentioned in section 2.2. In order to validate the model, two types of experiments were performed: (i) multi-batch produce washing with periodic FC replenishment at the 3 L scale, and (ii) multi-batch produce washing with periodic FC replenishment at 50–100 L scale. In addition, published data from an experiment by Luo et al. was used to validate the model for shredded iceberg lettuce washing at the pilot scale of 3200 L (Luo et al., 2012). For each of these types of experiments, model inputs included only: (a) parameter values for β and γ determined from single wash experiments, (b) COD increase rates, (c) the initial FC concentration before any produce was washed (i.e. time t = 0), and (d) sodium hypochlorite dosing information between respective runs, for carrots, cabbage, and iceberg lettuce, respectively.

2.3.1. Small scale experiments (3 L)

Experiments for periodic FC replenishment were carried out in a 3 L wash system. Essentially, the procedure followed that described in section 2.1, but for 3 separate runs. Briefly, sodium hypochlorite was added to the wash tank to achieve approximately 20 mg/L of FC (specific measurements presented in Fig. 2) in the wash water. The pH was simultaneously adjusted to 6.5 using 1 mol.L^{-1} citric acid. 600 g of the respective cut produce types were washed during each experiment through 6 batches, each consisting of 100 g. Directly after the first run of washing 600 g of produce, the FC was replenished (via the addition of sodium hypochlorite) to reach approximately 20–30 mg/L (specific measurements presented in Fig. 2) and the pH was again adjusted to 6.5. The procedure above was followed until 1200 g of produce was washed batch-wise and then the FC was replenished a final time, the pH adjusted to 6.5, and the final 600 g of produce was washed. Thus, a total of 1.8 kg produce was washed for each trial with 3 separate runs. For specific amounts of sodium hypochlorite added between successive runs, refer to Table A3 in the Appendix A.

Water quality measurements included FC and total chlorine, COD, turbidity, TDS, and pH. FC was measured immediately after each batch based on a DPD method using a Chlorine Photometer. The pH, turbidity, and TDS were measured on-site using a digital pH meter, turbidity meter, and TDS meter, respectively. The COD was determined using a reactor digestion method as detailed elsewhere (Luo, 2007).

2.3.2. Larger scale experiments (50–100 L)

15 kg of produce were sliced using a mandolin slicer to 3.2 mm thick for carrots and 25.4 mm × 25.4 mm pieces for lettuce/ cabbage, and kept in sterilized containers immediately prior to being discharged into a wash tank (tap water; 100 L for carrots and 50 L for lettuce/ cabbage) at a rate of 0.5 kg/min (0.25 kg batch every 30 s). The experiment consisted of three 10 min runs, simulating a continuous wash operation with periodic replenishment of sodium hypochlorite. In each
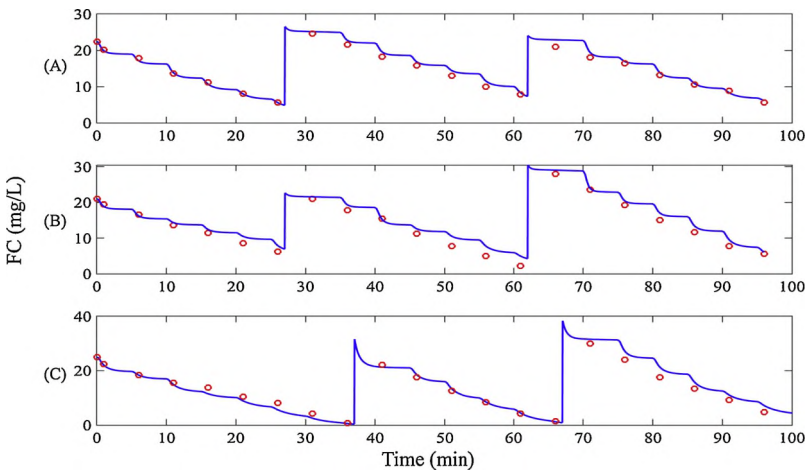
Fig. 2. Model prediction of FC kinetics for disk carrots (A), cut cabbage (B), and cut iceberg lettuce (C), at 3 L (multi-batch wash experiments). Panel A indicates the predictions for disk carrots (trial #1, see Table 3) corresponding to three experimental runs with redosing of FC in between each run. Similarly, panels B and C correspond to cut cabbage (trial #1, see Table 3) and cut iceberg lettuce (trial #1, see Table 3), respectively. The solid lines are the model predictions and the data points are FC measurements. Refer to Table 3 for RMSE values.
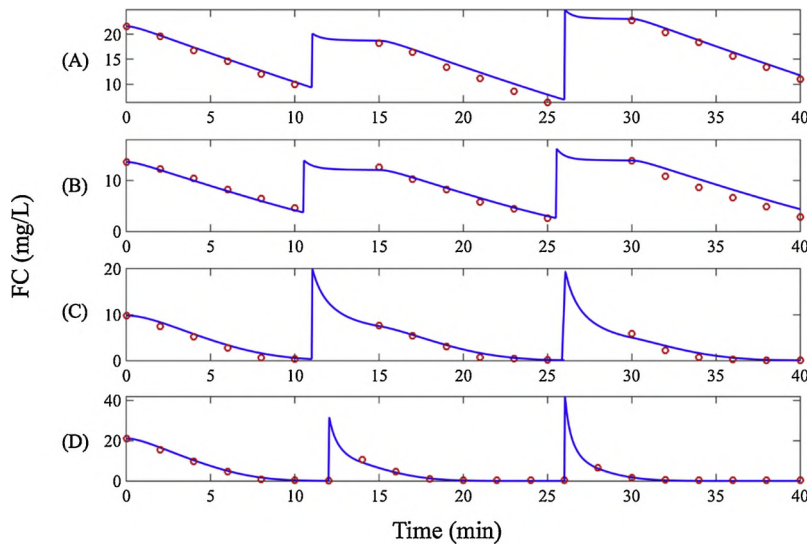
Fig. 3. Model prediction of FC kinetics for disk carrots, cut cabbage and cut iceberg lettuce at 50–100 L and 3200 L (continuous wash experiments). Panel A indicates the predictions for disk carrots corresponding to three experimental runs (at 100 L) with redosing of FC in between each run. Similarly, panels B and C correspond to cut cabbage (50 L) and cut iceberg lettuce (50 L), respectively. Finally, panel D illustrates the FC prediction for washing shredded iceberg lettuce at the pilot scale (3200 L). The solid lines are the model predictions and the data points are FC measurements. Refer to Table 3 for RMSE values.

run, 5 kg of produce was washed. Before the start of the first run, 60 mL (for carrots wash) or 18–20 mL (for cabbage/ lettuce wash) of concentrated (4.5 %) sodium hypochlorite was added to the wash tank to achieve approximately 21.5 mg/L FC (for carrots) and 13.5 mg/L of FC (cabbage/ lettuce) washing solution (specific measurements presented in Fig. 3). The pH was simultaneously regulated to 6.5 using citric acid. During washing, the carrots pieces were submerged into the water manually, washed for 30 s, and then removed via sieves. Water quality (COD, pH, Turbidity and TDS) and FC levels were measured every two minutes as detailed in section 2.3.1 for the 3 L experiments. After finishing the first run, the FC level was replenished by adding concentrated (4.5 %) sodium hypochlorite and the pH was again adjusted to 6.5 by adding citric acid. The experiment resumed after 5 min, and samples were collected every two minutes for the second segment. Similarly, after finishing the second run, concentrated (4.5 %) sodium hypochlorite was added to tank, pH readjusted to 6.5 using citric acid, and the same procedure was repeated for the third run. For specific amounts of sodium hypochlorite added between successive runs, refer to Table A3 in the Appendix A.

2.3.3. Model for multi-batch washing with chlorine replenishment

To account for a multiple batch wash process with FC redosing, equations 1.1–1.3 were naturally extended as follows:

$$\frac{dC}{dt} = D_n - \beta RC - \lambda C \qquad (2.1)$$

$$\frac{dR}{dt} = \gamma K_n - \beta RC \qquad (2.2)$$

$$K_n = \begin{cases} k_n, & \text{for during batch } n \text{ wash time} \\ 0, & \text{else} \end{cases} \qquad (2.3)$$

$$D_n = \begin{cases} d_n, & \text{for during } n^{th} \text{ dose time} \\ 0, & \text{else} \end{cases} \qquad (2.4)$$

$$C(0) = C_0, R(0) = 0 \qquad (2.5)$$

where d_n (mg.L^{-1}. min^{-1}) is the rate of FC increase depending on the addition of sodium hypochlorite to the wash water, and where k_n (mg.L^{-1}. min^{-1}) is the rate of COD increase during the nth wash. Notice that for larger scale ($>$ 3 L) washing experiments, $K_n = k_{avg}$, the average COD increase was used. We calculated d_n as follows: let S_0 be the initial volume of concentrated sodium hypochlorite added before any produce is washed, let S_n be the volume of sodium hypochlorite added at dose n, and let τ (min) represent the time to add sodium

hypochlorite (usually about 2.5 s), then $d_n = \dfrac{\frac{S_n}{S_0}C_0}{\tau}$. Note that the specific dosing information (S_i) relative to respective runs within each experiment is given in the Appendix A in Table A3.

2.4. Experimental procedure at the pilot plant scale

An experimental study (Luo et al., 2012) was conducted in a commercial pilot plant (New Leaf Food Safety Solutions, LLC, Salinas, CA). The setup consisted of a commercial double wash system, each tank with approximately 3200 L capacity, and equipped with rotating screens to ensure produce submersion, and air pumps to create turbulence in the wash water. While the larger goal of that study was to track *Escherichia coli* O157:H7 cross-contamination during lettuce washing, we considered here only the following components of that experiment relevant to tracking FC decay kinetics: (i) The entry rate of the shredded lettuce was approximately 45 kg/min; (ii) While the process water was continuously screened and re-circulated, produce spent an average of 26 s in each tank; (iii) The pH in the wash water was maintained at 6.5 using citric acid; (iv) Each test run involved three consecutive 12 min segments, simulating continuous processing with a periodic FC dosing scheme; (v) Sodium hypochlorite was added every 12 min (during a 2 min dosing period) with increasing dose volumes over a continuous wash period of approximately 40 min; and finally, (vi) pH, COD, turbidity and FC were monitored every 2 min.

In this study, we utilize the COD data [from Fig. 2A (Luo et al., 2012)] to inform the parameter k_{avg} (increase in COD in the wash water relative to incoming rate of lettuce) and the parameter values for β and γ (determined via the procedure in section 2.2 relative to iceberg lettuce) as inputs into model (equations 2.1–2.5) to predict the FC level data [from Fig. 3A in (Luo et al., 2012)].

2.5. Comparison of model predictions against data

Parameter values for β and γ (Table 2), taken from the ranges determined from the single batch wash experiments for respective

Table 2
Parameter values used for model predictions.

Produce	Tank Volume	β (L mg^{-1} min^{-1})	γ
Disk Carrots	3 L & 100 L	0.09	0.063
Cabbage	3 L & 50 L	0.10	0.11
Iceberg Lettuce	3 L & 50 L	0.049	0.11
Shredded Iceberg	3200 L	0.049	0.135

Table 3
Results of model prediction (quantified in terms of RMSE values) for multi-batch washing experiments at 3 L, multi-batch wash experiments at 50–100 L and continuous washing experiments at the pilot plant scale of 3200 L. Note that Trial # i = 1 or 2 in the 3 L context, indicates separate repeat experiments (3 runs each).

Produce	Volume	RMSE (Run1)	RMSE (Run2)	RMSE (Run3)	RMSE (exp.)
Disk Carrot #1	3 L	0.36	0.36	0.98	0.89
Disk Carrot #2	3 L	0.66	0.54	1.68	1.09
Cabbage #1	3 L	1.10	1.97	1.36	0.93
Cabbage #2	3 L	1.40	4.48	6.87	4.82
Iceberg Lettuce #1	3 L	0.97	0.99	0.93	0.96
Iceberg Lettuce #2	3 L	0.99	0.98	0.93	0.96
Disk Carrot	100 L	0.41	1.01	0.40	0.67
Cabbage	50 L	0.25	0.49	1.18	0.75
Iceberg Lettuce	50 L	0.65	0.42	0.67	0.59
Shredded Iceberg	3200 L	0.62	0.62	0.30	0.54

produce/cut types (as outlined in section 2.2), were used in the model (equations 2.1–2.5) to predict FC levels relative to each experiment conducted as described in sections 2.3 and 2.4. Note that λ is fixed to the same value for all predictive simulations (see section 3.1 for more details). Root mean square error (RMSE) values were computed relative to each run/produce type to quantify the quality of the respective predictions and were reported in Table 3.

2.6. Sensitivity analysis

To provide quantifiable evidence as to which model parameters have the greatest influence on the FC level, C, and the reactant level, R, we used Latin hypercube sampling (LHS) to build a sample matrix of parameter input values relative to each model output. Here we assumed that each parameter (β, γ, and k_{avg}) is sampled randomly from a uniform distribution across its respective range (coming from Tables 1 and A2). Using a sample size of n = 2000, and then rank-transforming the sample matrix and corresponding model output for C and R, the partial rank correlation coefficients (PRCCs) associated to each parameter were calculated (Marino et al., 2008). In general, the PRCCs take values between -1 and +1, and indicate a measure of monotonicity between each parameter and a respective model output. See section 3.5 for results and discussion regarding this analysis.

3. Results and discussion

3.1. Parameter fits from single-batch wash experiments

FC decay data during single-batch washes, corresponding to the respective produce type, were presented in Fig. 1. Notice that for each produce type the decay pattern is bi-phasic, *i.e.*, an initial fast decay (until about 5 min) followed by a transition to a relatively slower decay rate, similar to that observed by (Teng et al., 2018) as well as in the context of drinking/waste water by (Kastl et al., 1999) and (Clark, 1998). This observation as well as the fact that following the introduction of 100 g of cut produce into the water, after the first measurement (at 0.5 min), the COD values did not change for respective produce types, justify the assumption that the reactant(s) (denoted by R) must be depleted after the initial 5 min. Furthermore, the steady decay of FC after this point (Fig. 1) is assumed to follow purely first-order kinetics, which closely matched the decay rate of FC in tap water (Hua et al., 1999).

The results of the calculations for γ and the fitting procedure for, outlined in section 2.2 along with data to determine ranges for each parameter across the three experiments were presented in Table 1. The

results for respective β values did not depend on the initial conditions administered during the minimization algorithm. Also, see the end of section 3.5 for further discussion concerning the differences in the parameter ranges relative to produce type.

3.2. Model predictions at the 3 L and 50 − 100 L scale

Using commodity-specific values of the model parameters β, γ (Table 2), and COD data [used to inform the change in COD, k_n, after each batch of produce washed for multi-batch washing (Table A1), k_{avg} for larger scale washing experiments (Table A2)] as inputs, as well as sodium hypochlorite dosing information (Table A3), our model (equations 2.1–2.5) was able to successfully predict the FC decay kinetics at both the 3 L (multi-batch washing experiments) and 50 − 100 L scales. Table 2 indicates the parameters used for predictions against all wash experiments for respective produce types and scales. Note that for each of the produce types, the values for β and γ fall within the ranges determined from the single wash experiments (Table 1).

3.2.1. Model results at 3 L

Table 3 summarizes the performance of our model (equations 2.1–2.5) against FC decay data during the respective cut produce washing with FC replenishment at the 3 L scale, listing RMSE values for each respective run as well as a total RMSE value considering all three runs. Fig. 2 provides a clear visual of the success of this model in predicting FC decay for disk carrots (panel A), cut cabbage (panel B), and cut lettuce (panel C), respectively, at the 3 L scale. The model output for FC levels replicates a bi-phasic type decay pattern between successive data points as washing is batch-wise (Fig. 2). Mathematically, this is accounted for via the on/off input function K_n (Eq. 2.3). Furthermore, Fig. 2 indicates the dynamics of FC during the dosing interval (using specific dosing input information given in Table A3), in between successive washing runs. For instance, Fig. 2(A) shows that sodium hypochlorite was added at $t=$ 27 min with carrot washing restarted at $t=$ 35 min for the second run. Sodium hypochlorite was additionally added at $t=$ 62 min with carrot washing restarted at $t=$ 70 min. Dosing for washing the other produce types followed a similar pattern as seen in Fig. 2(B) and (C).

It is important to mention that the only chlorine related data the model takes as inputs are the FC measurement at time $t = 0$ min and the amount of sodium hypochlorite added between runs. Thus, the results in Fig. 2 as well as those presented in Table 3 (3 L experiments) illustrate the success of the model predictions in terms of both FC increase due to dosing and FC decay due to washing respective produce types. Notice that the total RMSE < 1.1 (Table 3 last column) for each prediction except for cabbage trial #2, which had a total RMSE of 4.82. The issue here was that the model underestimated the FC decay during the second half of each washing run. The interesting part is that the data for cabbage trial #2 indicated faster FC decay rates at the end of each washing run with corresponding slower COD increase rates than during the first half of each washing run. It is not yet clear why this occurred, however, this inconsistency was not seen with any of the other experiments at 3 L or at the larger scale.

3.2.2. Model results at 50–100 L

For the large-scale multi-batch wash experiments, 50 L of water was used for lettuce and cabbage, and 100 L of water was used for disk carrots, respectively. We used 100 L of water for carrots vs. 50 L for cabbage and iceberg lettuce merely to make sure that similar sodium hypochlorite dosing amounts could be used for the experiments involving the three produce types. In particular, the water volumes were chosen so that rate of COD increase from washing the respective produce types would be similar. This ensured that the FC would not be completely depleted half-way through a washing run.

Using the same parameter values listed in Table 2 for respective produce types, COD input (tracked as the average increase in COD, k_{avg},

from Table A2 in the Appendix A), and sodium hypochlorite dosing (Table A3), our model (equations 2.1–2.5) was again able to successfully predict the FC dynamics (Table 3). Fig. 3 illustrates the model's prediction versus data for disk carrots at 100 L (panel A), cabbage washing at the 50 L scale (panel B), and for iceberg lettuce at 50 L (panel C).

The results in Table 3 (and Figs. 2 & 3) suggest a few points. First, they indicate that the model (equations 2.1–2.5) captures the main mechanisms governing observed FC decay relative to the produce/cut type. Second, the model can consistently account for the FC levels in between runs relative to the amount of sodium hypochlorite added and the rate of decay of FC due to the amount of reactant(s) R (mg/L) remaining at the start of the dosing period. Third, FC reaction rates relative to the produce/cut type appear to be robust with respect to scaling as the same rates used by the model at both the 3 L and $50-100$ L resulted in accurate predictions. Finally, the parameter γ, the fraction of the COD increase due to repeated produce washing, is a relatively consistent predictor of the associated produce/cut type organic load in wash water, and therefore a reliable predictor (via the model) of FC decay rates.

3.3. Model predictions for iceberg lettuce at a pilot-plant scale

Using the values of the model parameters β, γ (Table 2), for shredded iceberg lettuce, and COD data from (Luo et al., 2012) [used to inform the change in COD, $K_n = k_{avg}$ (Table A2), during continuous washing], as well as the volume/timing of 12.5 % sodium hypochlorite additions (Table A3) as inputs, our model (equations 2.1–2.5) successfully predicted the FC dynamics at the pilot plant scale (3200 L). The comparison of the model output versus FC data were presented in the last row of Table 3 and panel D of Fig. 3.

It is noteworthy that in the 3 L and 50 L iceberg lettuce experiments, the wash water temperature was 20 °C and the lettuce cut size was 25.4 mm × 25.4 mm, whereas the water temperature during the pilot scale experiments was about 5 °C and the lettuce was shredded (6 mm shreds). Furthermore, the lettuce used in the 3 L and 50 L experiments was purchased from a local supermarket whereas the lettuce in the pilot plant study was washed within 24 h of harvesting. Our model (equations 2.1–2.5) results (Table 3) indicate that FC reaction rates associated with iceberg lettuce washing appear to be robust despite differences in produce supply-chain history, cut size, and experimental scale, as the same value for the apparent second order rate constant $\beta = 4.9 \times 10^{-2}$ (L. mg^{-1}. min^{-1}) utilized in the model at the 3 L, 50 L and 3200 L resulted in accurate predictions. Note that for the 3200 L prediction, $\gamma = 0.135$ whereas for the 3 L and 50 L predictions, the model input used was $\gamma = 0.11$. While both values for γ are in the range determined from the 3 L single wash experiments, it may be that since the lettuce in the pilot plant study was used within a day of harvesting, soil and other such materials contributed to a slightly higher value of γ. These results also indicate that the temperature differences (20 °C vs 5 °C, respectively) between the lab and pilot scale experiments did not significantly influence the observed FC decay rates. Furthermore, cut type seemed to not be significant as single wash 3 L experiments using shredded iceberg lettuce yielded almost identical ranges for γ and β as those for square cut lettuce (data not shown).

Additionally, Fig. 1A [from (Luo et al., 2012)] indicates that the average initial COD value in the 3200 L tank was about 300 mg/L. This value is extremely high for a mixture of tap water, citric acid for pH control, and an initial dose of 700 mL of 12.5 % sodium hypochlorite. If for instance some initial produce washing occurred before the start of the recorded runs, the initial condition for the reactant(s) concentration $R(0) > 0$. Notice that if we let $R(0) = 8$ mg/L, set $\gamma = 0.115$ and $\beta = 4.9 \times 10^{-2}$ (L. mg^{-1}. min^{-1}), then the model predictions are similar to those in Fig. 3D, with an overall RMSE of 1.45.

Finally, even though the rate of iceberg lettuce coming into the wash tanks differed across experimental scale (and thus the COD

increase rate differed across scales), the range for γ from the single wash lettuce studies at 3 L, used to represent the corresponding increase in organic load, resulted in faithful FC predictions at all scales.

3.4. Predictors of FC decay

Note that the success of the model predictions (as discussed in sections 3.2 and 3.3) depends on the fact that the change in organic load (relative to produce/cut type) was quantified *via* a model that utilizes the *successive increase in COD* rather than current COD levels. More specifically, the consistency of the model (equations 2.1–2.5) predictions stem from two main assumptions (built from the single wash experiments), namely: (i) as cut produce enters the wash tank, the increase in constituents in the wash water that rapidly deplete FC [represented as the concentration of R in the model (equations 2.1–2.5)] is fairly well approximated as a fraction of the change in COD in the wash water (due to washing more and more produce), and (ii) the slower depletion of FC (via tap water or possibly other constituents) can be well approximated by a first-order decay term (see Eq. 2.1).

In terms of practically predicting FC levels, unfortunately, real-time COD measurements are not currently feasible during produce washing and therefore surrogate parameters which can be measured in real-time have been sought after. Many studies have examined the correlative strength of various water quality parameters such as UV254, turbidity, TDS, to name a few, with regards to chlorine demand and produce type [e.g., (Barrera et al., 2012; Chen and Hung, 2016; López-Gálvez et al., 2019; Pirovani et al., 2001; Van Haute et al., 2018, 2013)]. In particular, because of the ease of real-time measurements and because of their inclusion in multiple wash water studies, we chose to examine if the change in turbidity (Δ TUR) or the change in total dissolved solids (Δ TDS) could be consistently used to predict the change in COD (Δ COD) relative to consecutive multi-batch and continuous washing of disk carrots, cabbage and lettuce. That is, we seek functions U and T such that Δ COD $= U(\Delta$ TUR$)$ and Δ COD $= T(\Delta$ TDS$)$. Furthermore, it is logical that these functions be monotonic (i.e. an increase in the input corresponds to an increase in the output).

To evaluate if such functions U and T can be determined in practice, wash data for carrots/ cabbage/ lettuce obtained at both the 3 L and $50-100$ L level (as detailed in section 2.3) was utilized as well as wash data for shredded lettuce at the pilot plant scale (as discussed in section 2.4). In particular, the Spearman's rank correlation coefficient (ρ) between (a) Δ TUR and Δ COD, and (b) Δ TDS and Δ COD, for respective produce types and experimental runs was calculated. Essentially, ρ indicates how well the connection between two variables can be described by a monotonic function.

Using the *corr* function in MATLAB, the corresponding ρ value for each experimental run and across respective experiments was calculated. The results are presented in Tables A4 and A5 (3 L scale), Tables A6 and A7 ($50-100$ L scale), and Table A8 (pilot scale 3200 L), providing strong evidence that there is no consistent relationship between Δ TUR and Δ COD as well as between Δ TDS and Δ COD for cut carrots, cabbage and iceberg lettuce across the scales considered. While a complicated mechanistic relationship that links real-time Δ TUR or Δ TDS with FC decay kinetics might exist, the analysis above illustrates that this relationship is not readily evident.

3.5. Sensitivity analysis

Following the procedure outlined in section 2.6, uniform distributions – typically used to build a sample matrix relative to each model output and with respect to each produce type – were constructed from parameter information as presented in Tables 1 and A2. This analysis was performed only in the context of larger scale wash experiments (50–100 L, and pilot scale). Recall that β is the apparent second order rate constant, k is the rate of COD increase due to produce entering the wash water, and γ is a fraction so that γk indicates the entry rate of
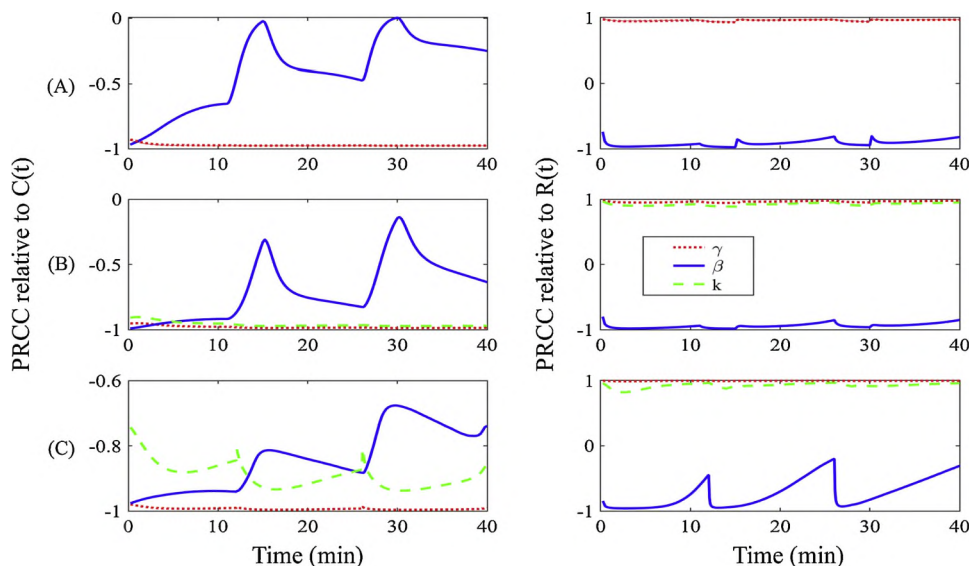
Fig. 4. PRCCs in the context of continuous produce washing. Panel A corresponds to washing disk cut carrots (100 L), where the left figure in panel A illustrates PRCC values (vs time) between the corresponding parameter (see the legend in the bottom panel) and the model FC output at time t, $C(t)$. The right figure in panel A illustrates PRCC values (vs. time) between the corresponding parameter and the model reactant output at time t, $R(t)$. Similarly, panel B is for cut cabbage (50 L) and panel C corresponds to shredded iceberg at the pilot scale (3200 L). Notice in panel A the curve for γ essentially overlays the curve for k.

reactant(s) into the wash water from cut produce.

Fig. 4 (panel A) illustrates PRCC values (over time) relative to disk carrot washing (100 L) and Fig. 4 (panel B) depicts PRCC values for cut cabbage washing (50 L). These figures show that the sensitivity of the model's FC output as well as the reactant(s) output depends similarly on variations in the parameters, γ, and k. Recall that PRCC values close to 0 indicate low sensitivity whereas values closer to 1 or -1 illustrate high sensitivity. Notice that fresh sodium hypochlorite was added at time t =11 min and t =26 min for both carrot and cabbage washing. While the sensitivity of the FC levels relative to γ and k was not noticeably affected during dosing, the sensitivity of the FC levels with respect to β dramatically decreases (PRCC values quickly increase towards 0) during the chlorine replenishment periods (see the left most figures in panel A and B of Fig. 4). This is because during the replenishment periods, the reactant(s) (denoted by R in Eq. 2.2) are totally depleted.

In contrast to this, the model predicts that for shredded iceberg lettuce washing at the pilot plant (3200 L) scale, even though the sensitivity of the FC levels relative to β decreases during the dosing periods, the PRCC values stay well below -0.6 (see the leftmost figure in panel C of Fig. 4). The reason for this is that due to the relatively high input rate of shredded lettuce (45 kg/min), the COD increase rate is 1.5–2 times faster than those in the carrot/cabbage experiments, leading to substantial levels of R remaining in the wash water, even just after the dosing period.

While it is clear from Fig. 4 that model predictions of the FC levels are strongly influenced by the parameter γ across the board, the above discussion indicates that as the produce input rate to wash water volume ratio increases, the influence of apparent reaction rate β becomes more significant. These sensitivity results stress the importance of thoroughly understanding the second-order reaction dynamics of FC with produce constituents (coming from carrots, cabbage and iceberg) and further reinforce the utility of lab experiments to gain more insight in this regard.

Referring to the ranges for β (with respect to produce type) as determined from single wash experiments at 3 L (Table 1), it seems logical that reaction rates of FC with respect to produce type would differ as their constituents differ [USDA's Food Data Central website]. However, Table 1 indicates that the reaction rates for disk carrots and cabbage have similar larger ranges, whereas the range of reaction rates for iceberg lettuce is narrower with smaller values. On the other hand, the

ranges for the fraction of the COD increase (γ), which governs the input rate of reactant(s) R into the wash water as cut produce enters the wash tank, are relatively similar for iceberg lettuce and cabbage but differ substantially for carrots. While it is not immediately clear how to explain these findings in terms of the constituent make up of each type of produce, the data on the USDA's Food Data Central website may be useful for future detailed study in this regard.

4. Conclusions

The wash model developed in this study applies to a variety of produce types in the context of predicting FC dynamics (*decay* and *re-dosing*) across various scales. The fact that its predictions of FC levels hold at multiple experimental scales and across three produce types strongly suggests that the model illustrates fundamental chlorine dynamics that occur during fresh cut carrot/cabbage and iceberg lettuce washing. In particular, these findings give validity to performing future lab scale experiments to quantify FC kinetics associated with different produce/cut types as well as experiments aimed at understanding the impact of continuous FC dosing on FC dynamics during produce washing. Specifically, the effect of pH dynamics should be explored in this context as the results from this paper were obtained via wash water pH levels near 6.5.

In addition, it was shown that turbidity and TDS measurements may not be reliable in predicting *FC levels* (as opposed to chlorine demand) as there is no consistent, observable relationship linking the increase in organic load (in terms of change in COD) from cut carrots/cabbage/lettuce entering the wash tank (across various scales 3 L, 50–100 L, and 3200 L) and the corresponding increase in turbidity or TDS. Recall that the model's predictive success across scales and produce types provides a strong case that COD information is much more dependable than that of turbidity or TDS. In particular, the fraction γ of the COD increase due to washing produce was shown to be a consistent predictor of the associated produce/cut type organic load in wash water and therefore a reliable predictor (via the model) of FC decay rates.

While the inability to measure COD dynamics in real-time during commercial washing limits the model's ability to be directly implemented into online FC control, the model can be used as a key tool for validating FC compliance within operational limits at the industrial scale. However, due to the model's sensitivity to γ (see section 3.5),

care must be taken in determining the range for this parameter relative to the washing setup and produce type in question.

Finally, the results from this work demonstrate the utility of using mathematical models as tools to elucidate fundamental mechanisms, like FC reaction rates associated with various produce cut types. To minimize expensive experiments at the commercial scale, the model developed herein, and particular lab scale experiments could aid in planning the logistics of how and what should be measured during commercial scale experimentation.

Declaration of Competing Interest

The authors declare that there are no conflicts of interest.

Appendix A

Table A1

Example of determination of k_n (increase of COD corresponding to the nth batch wash), for iceberg lettuce trial 1, run 1 at 3 L. The following values for k_n were used as inputs into model (equations 2.1–2.5), whose resulting FC output is shown in Fig. 2 panel (C), corresponding to the first 36 min of washing. Note that since k_n = (successive change in COD)/(0.5 min) there are only eight entries in the 3rd column.

Number of batch wash	COD (mg/L)	k_n (mg L^{-1} min^{-1})
1	30	92
2	76	46
3	99	84
4	141	38
5	160	68
6	194	82
7	235	94
8	282	110
9	337	

Table A2

Values determined for k_{avg} from the average increase of COD during continuous wash experiments. The ranges for k_{avg} are given as mean ± standard deviation.

Produce/ Vol	k_{avg} (mg L^{-1} min^{-1})
Disk Carrot, 100 L	19.07 ± 3.31
Cabbage, 50 L	10.11 ± 1
Iceberg, 50 L	19.03 ± 0.76
Iceberg, 3200 L	30.72 ± 3.29

Table A3

Amounts of concentrated sodium hypochlorite added for respective experimental runs. Note that 4.5 % sodium hypochlorite was used for all experiments except for shredded iceberg, during which Luo et al. used 12.5 % sodium hypochlorite (Luo and others 2012). Also, note that Trial # i = 1 or 2 in the 3 L context, indicates separate repeat experiments (3 runs each).

Produce	Volume	Initial Dose	Dose 2	Dose 3
Disk Carrot #1	3 L	1.5 mL	1.5 mL	1.125 mL
Disk Carrot #2	3 L	1.5 mL	1.125 mL	0.75 mL
Cabbage #1	3 L	1.5 mL	1.125 mL	2.25 mL
Cabbage #2	3 L	1.5 mL	1.5 mL	1.5 mL
Iceberg Lettuce #1	3 L	1.5 mL	1.875 mL	2.25 mL
Iceberg Lettuce #2	3 L	1.5 mL	1.875 mL	1.875 mL
Disk Carrot	100 L	60 mL	30 mL	50 mL
Cabbage	50 L	20 mL	15 mL	20 mL
Iceberg Lettuce	50 L	18 mL	36 mL	36 mL
Shredded Iceberg	3200 L	750 mL	1050 mL	1400 mL

Table A4
Spearman's rank correlation coefficient ρ for Δ COD vs Δ TUR data from experiments at the 3 L scale.

Produce	Experiment	Run 1	Run 2	Run 3	Total
Carrots	1	0.149	0.3714	-0.029	0.1376
Carrots	3	0.4569	-0.1218	0.5385	0.3659
Cabbage	1	0.3714	-0.4928	-0.1160	0.1894
Cabbage	3	-0.3361	0.381	0.2101	0.2699
Iceberg	1	0.3353	0.700	-0.2319	0.1402
Iceberg	2	0.2224	0.7143	0.8469	0.4011

Table A5
Spearman's rank correlation coefficient ρ for Δ COD vs Δ TDS data from experiments at the 3 L scale.

Produce	Experiment	Run 1	Run 2	Run 3	Total
Carrots	1	-0.1177	0.3381	0.6269	0.4054
Carrots	3	0	-0.1674	-0.2582	-0.0181
Cabbage	1	0	0.8454	-0.1791	0.3119
Cabbage	3	-0.7307	0.2520	-0.1967	0.0274
Iceberg	1	0.0782	0.2887	0.1852	0.2191
Iceberg	2	-0.3203	-0.0976	0.1456	-0.0884

Table A6
Spearman's rank correlation coefficient ρ for Δ COD vs Δ TUR data from experiments at the 50 L for cabbage and iceberg, and 100 L scale for disk carrots. * indicates significance where $p < 0.05$.

Produce	Experiment	Run 1	Run 2	Run 3	Total
Carrots	1	0.300	0.100	0.100	0.2538
Cabbage	1	-0.300	-0.600	0.100	-0.1251
Cabbage	2	0.8208	-0.100	0.500	0.0413
Iceberg Lettuce	1	-0.6325	-1.00	0.400	-0.1053
Iceberg Lettuce	2	-0.6669	0.300	-1.00*	-0.4293

Table A7
Spearman's rank correlation coefficient ρ for Δ COD vs Δ TDS data from experiments at the 50 L for cabbage and iceberg, and 100 L scale for disk carrots. * indicates significance where $p < 0.05$. NaN indicates no number returned for \rho

Produce	Experiment	Run 1	Run 2	Run 3	Total
Carrots	1	0.7071	0.7182	0.5798	0.4080
Cabbage	1	0	-0.2887	-0.8660	-0.3233
Cabbage	2	0.8652	0.7379	0.2887	0.5911*
Iceberg Lettuce	1	NaN	-0.4472	0.2582	0.1913
Iceberg Lettuce	2	-0.1579	-0.3536	-1.00*	0.0088

Table A8
Spearman's rank correlation coefficient ρ for Δ COD vs Δ TUR average data from pilot plant experiment [from Fig. 2A of (Luo et al., 2012)].

Produce	Run 1	Run 2	Run 3	Total
Iceberg Lettuce	-0.3714	0.4857	-0.0286	0.0588

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