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Assessment and speciation of chlorine demand in fresh-cut produce wash water

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ABSTRACT

For the fresh-cut produce industry, a critical area of concern is potential pathogen cross-contamination during wash operations when wash water is reused and re-circulated in wash systems continuously imputed with fresh-cut produce. However, little research has focused on the chemical properties of wash water. Organic input from residual soil and vegetable material deteriorates water quality and creates increasing chlorine demand within this wash water.

This study evaluated the origins of chlorine demand input and chlorine decay kinetics of fresh-cut produce wash water. Using a model system, vegetable juice released per kg of processed produce for shredded romaine lettuce, shredded iceberg lettuce, shredded carrot and baby spinach was 82.1 mL/kg, 94.5 mL/kg, 158 mL/kg, and 2.26 mL/kg, respectively. Batch water analysis revealed a rapid reaction between constituents in the wash water and chlorine where over a 90 min observation period, 50% of chlorine demand occurred within first 5 min, underscoring the challenge for any water treatment process to reduce chlorine demand once vegetables are deposited into washing systems. Moreover, the results also showed sustained chlorine demand over 90-min periods, indicating an accumulative effect on chlorine consumption with continuous organic input. Additionally, HPLC-SEC analysis showed that the constituents contributing to chlorine demand are predominantly dissolved small molecules (<3400 Da), which will challenge water reuse treatment approaches. This study provides quantitative information of chlorine demand origins and chlorine decay kinetics in wash water and provides baseline data critical for integrating water reuse in the fresh produce processing industry.

1. Introduction

The demand of fresh-cut leafy green vegetables has continued to expand as consumers have integrated healthy diets with the concept of ready to eat meals. Reports by the World Health Organization (WHO, 2005) document that consumption of vegetables has benefits to human health by providing high levels of minerals and vitamins with the goal of preventing chronic diseases. Consuming uncooked food, however, has risks, such as those associated with food-borne illness outbreaks. As outlined by Center for Food Safety and Applied Nutrition, 72 outbreaks were reported to be associated with fresh produce from 1996 to 2006 with 18 out of these outbreaks due to fresh-cut produce (CFSAN, 2008). Additional outbreak investigations conducted in recent years continue to indicate that consumption of contaminated fresh-cut produce can be problematic (Barton Behravesh et al., 2011; Buchholz et al., 2011; Greene et al., 2008; Hanning, Nutt, & Ricke, 2009; MacDonald et al., 2012; Nygård et al., 2008; Söderström et al., 2008). To reduce microbial

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contamination and improve produce safety, it is clear that proper sanitation is essential during fresh-cut produce postharvest processing. In the absence of a practical decontamination method to directly remove/kill pathogens from produce, washing with sanitizing water has been widely adopted by the fresh produce food industry. Although other disinfectants or disinfection methods such as ozone, organic acids, chlorine dioxide, and UV irradiation have been used (Gil, Selma, López-Gálvez, & Allende, 2009; Ölmez & Kretzschmar, 2009), addition of chlorine (or other forms of hypochlorous acid) in wash water is still the most common practice. Chlorine is predominant because of its efficacious disinfection capability against a wide spectrum of microorganisms and its economic accessibility compared to other disinfectants (Arana, Santorum, Muela, & Barcina, 1999; Baxter, Hofmann, Templeton, Brown, & Andrews, 2007; Bohrerova & Linden, 2006; Corona-Vasquez, Samuelson, Rennecker, & Mariñas, 2002; Cromeans, Kahler, & Hill, 2010; Li, Xin, Wang, Zheng, & Chao, 2002; Luh & Mariñas, 2007).

The vegetable washing process in food processing facilities is a mechanism by which water-borne pathogens can be dispersed throughout wash water. Allende, Selma, López-Gálvez, Villaescusa, and Gil (2008) reported that pathogens could carry over from previous processed produce to a subsequent batch of produce via wash water if insufficient residual chlorine is maintained in the washing tank. Luo et al. (2011) also described the importance of maintaining a minimum chlorine level during the wash process to avoid cross-contamination. However, during washing of fresh-cut produce, large amounts of organic compounds from soil debris and from exudates of produce are deposited into wash water (Luo et al., 2012). As these compounds react with chlorine, there is a rapid decrease in the level of free available chlorine as well as the potential generation of disinfection byproducts (Chen, Zhu, Zhang, Niu, & Du, 2010; López-Gálvez et al., 2010). Although chlorine replenishment is practiced during commercial produce washing operations, the level of organic compounds generated during continuous produce feeding rapidly deteriorates wash water quality in terms of depletion of chlorine and increasing turbidity and chemical oxygen demand (Luo et al., 2012). Maintaining adequate residual chlorine levels in the washing tank is challenging in commercial produce wash processes, and the resulting low chlorine level often increases the risk of cross-contamination (Luo et al., 2012).

Previous studies have predominantly focused on the association between water quality and the potential of cross contamination (Allende et al., 2008; Luo et al., 2011; Van Haute, Sampers, Holvoet, & Uyttendaele, 2013). Minimal information is available regarding quantitative assessment of the fraction of wash water responsible for observed chlorine demand, particularly in regards to organic loading. Therefore, this study evaluated the organic input per unit of processed produce during a washing process by using chemical properties (*e.g.*, chemical oxygen demand or total organic carbon) of wash water from four different vegetables (romaine lettuce, iceberg lettuce, carrots, and baby spinach). The data generated is useful in terms of estimating the quantity of chlorine demand input per mass of vegetable following different produce processing techniques. Moreover, the assessment of chlorination kinetics of these vegetable wash waters was conducted to characterize the behavior of chlorine demand in terms of reaction time and chloramine formation. By generating these data, existing and alternative industrial raw vegetable wash water processes should be able to be evaluated for the best treatment strategy. Further, use of this information could enable conceptual engineering designs for safe water reuse during leafy green washing processes.

2. Materials and methods

2.1. Vegetable wash water and vegetable extracts

Romaine lettuce (*Lactuca sativa* L.), iceberg lettuce (*L. sativa*), and carrots (*Daucus carata* L.) were purchased from a wholesale market located in Baltimore, MD. Fresh-cut samples were prepared freshly on the same day that experiments were conducted by shredding (0.32 cm in width) or chopping (6.45 cm^2) for both romaine lettuce and iceberg lettuce and shredding (0.32 cm by 0.32 cm) or slicing (0.32 cm in thickness) for carrots using a commercial food slicer (ECD-302; Nichimo International Inc., WA). Freshly harvested baby spinach leaves were received from Taylor Farms (Jessup, MD) and were processed without cutting.

Washing experiments were conducted by submerging prepared produce in a washing tank containing sterile deionized water at a produce:water ratio of 1 kg:20 L for prepared romaine lettuce, iceberg lettuce, and baby spinach samples and a ratio of 1 kg:40 L for prepared carrot samples at room temperature (22 °C). During each simulated wash cycle, 1 kg of prepared produce sample was packed in a mesh bag and completely submerged into washing tank for 1 min. The romaine lettuce, iceberg lettuce, and baby spinach were processed for a total of 15 simulated wash cycles and the carrots for 10 simulated wash cycles. A new batch of prepared produce was used for each simulated wash cycle and additional fresh water was added as needed to maintain a fixed volume between simulated wash cycles, which simulated the continuous operation in a commercial setting. Thus, a batch of 15 kg of produce was used in total during washing experiments for romaine lettuce, iceberg lettuce, and baby spinach and 10 kg in total during washing experiments for carrots. Samples were collected at the end of experiment or as needed between simulated wash cycles.

Vegetable liquid exacts of all four vegetables were prepared following the method of Shen et al. (2012) with a commercial household juice maker (Breville Model BJE200XL Juice Fountain, Shanghai, China). The liquid portion was collected, filtered with cheesecloth to remove coarse vegetable fragments and stored at −20 °C until use.

2.2. Analytical methods

Samples from washing experiments and from vegetable extracts were prepared with a proper dilution ratio to fit the detection range of analytical methods. The analysis included chemical oxygen demand (COD) by colorimetric method (method 5220; APHA-AWWA-WEF (2012)) using the TNT 822 COD Kit (Hach Company, CO), nitrogen composition analysis by colorimetric method using TNT 880 simplified TKN kit (Hach Company, CO), total organic carbon (TOC) by persulfate-ultraviolet oxidation method (Method 5310; APHA-AWWA-WEF (2012)) using a Sievers 900 series TOC analyzer (GE Analytical Instrument, CO), ultraviolet light absorbance at wavelength of 254 nm (UV_{254} abs.) using a DR4000U spectrophotometer (Hach Company, CO), and anions analysis by IC25 ion chromatography system with IonPac© A17 column (Thermo Scientific, CA).

2.3. Chlorination experiments

The final wash water samples were collected from washing experiments using four vegetable types as described above and were subjected to chlorination where 1 mL of sample was mixed with 0.02 mL of diluted chlorine stock solution (80,000–8000 mg/L as Cl_2) in form of sodium hypochlorous acid (NaOCl, 10–15%, Sigma–Aldrich). The applied chlorine concentration varies from 1600 to 160 mg/L as Cl_2 based on the chlorine demand in various wash waters and the feasibility for analysis by selected methods. Time-course sampling was conducted over sequential 90 min periods to assess the rate of chlorine consumption and chloramine formation. Chlorine demand was calculated by subtracting the chlorine consumption in the blank sample (deionized water) from the chlorine consumption in wash water samples. During chlorination experiments, a pH of 7.0 was maintained by addition of phosphate buffer (50 mM) as needed. The N,N-diethyl-*p*-phenylene diamine (DPD) colorimetric method (APHA-AWWA-WEF, 2012) was used to measure residual chlorine and total chlorine; chloramine formation was calculated by subtracting the residual chlorine reading from total chlorine measurement. Samples from chlorination experiments were collected and diluted with deionized water at a proper ratio according to the applied dose to accommodate the best detection range of the analytical method for chlorine and chloramine measurements. The effects of filtration on reduction of chlorine demand and chloramine formation were also evaluated and the filtered samples were prepared by using a 0.45 μm pore size filter (PVDF; Fisher Scientific, PA). Moreover, the effects of numbers of wash cycles were also evaluated with shredded iceberg lettuce following 5, 10, and 15 washing cycles.

The analysis of HPLC-Size Exclusion Chromatography (HPLC-SEC; Ultimate 3000 system; Thermo Scientific, CA) equipped with BioSep-SEC-S3000 column (300 mm × 7.8 mm; Phenomenex, CA) was applied on the final wash water samples from four vegetables with/without chlorination. The mobile phase was a mixture of 4.0 mM phosphate buffer solution and 0.1 M sodium sulfate solution with a flow rate of 1.054 mL/min, isocratic. All samples for HPLC-SEC were filtered with 0.45 μm pore size filter (PVDF; Fisher Scientific, PA) before analysis to prevent clogging in HPLC columns.

3. Results and discussion

3.1. Assessment of vegetable wash

Table 1 shows COD, nitrogen composition, TOC, UV_{254} absorbance, and anion measurements of four vegetable extracts and wash water of three shredded vegetables (romaine lettuce, iceberg lettuce and carrot) and baby spinach. Chopped and sliced produce samples had less vegetable exudate compared to shredded vegetables resulting in less of an issue for wash water processing and thus were not further evaluated (data not shown). From Table 1, the results reveal that wash water from carrots contained high concentrations of COD and TOC, but low concentrations of nitrogen, compared to romaine lettuce and iceberg lettuce. For wash waters and vegetable extracts, an Equivalent Extract Ratio (EER) (%, volume/volume) was calculated by the measurement of water quality parameters (*e.g.*, COD and TOC) of vegetable wash water divided by the same measurements on vegetable extracts. This ratio represents the percentage of a diluted vegetable extract solution that contains the same amount of target constituents as present in wash water. The average and standard deviation of equivalent extract ratio for four vegetable wash waters are shown in Table 2. Outlier measurements potentially resulting from extraneous soil debris were excluded from data analysis where the outlier measurement was defined as greater than three times the standard deviation.

The equivalent volume of vegetable juice released per weight of processed produce was calculated based on equivalent extract ratio, volume of wash water, and mass of total processed produce, as shown in Equation (1).

$$\text{Equivalent Vegetable Juice Released}\left(\frac{\text{mL}}{\text{kg}}\right)=\frac{\text{Average of Equivalent Juice Ratio }(\%)\times\text{Volume of Wash Water(mL)}}{\text{Total Mass of Processed Produce(kg)}} \quad (1)$$

Tables 1 and 2 reveal that baby spinach wash water has the lowest equivalent extract ratio, which could be due to the whole-plant being washed as compared to the other vegetables that were chopped or shredded. Minimal vegetable juice was recovered, only 2.26 mL per kg of processed produce. Wash waters from the other three shredded vegetable washes contained much higher concentrations of targeted analytes and increased release of vegetable juice compared to baby spinach wash waters, 82.1 mL/kg for romaine lettuce, 94.5 mL/kg for iceberg lettuce, and 158 mL/kg for carrots. Enhanced release of vegetable juice resulted from increased vegetable tissue surface area due to cutting or shredding. Nou and Luo (2010) have proposed washing whole-leaf produce to reduce the rate of water quality deterioration during the washing process with a subsequent reduction in potential microbial cross-contamination.

The analysis of vegetable juice released during the wash process provides information regarding organic loading into wash water. Zhou, Luo, Nou, and Millner (2014) established an empirically-based equation to calculate the chlorine requirement for vegetable wash, but the equation was established based on the vegetable extracts, not the amount of processed produce. Our study provides quantitative information on the volume of vegetable fluid extracts released by washing per unit of produce, which can be used in the empirical algorithm equation to calculate chlorine demand based on the amount of processed produce.

3.2. Chlorination kinetics of vegetable wash

Chlorine demand of wash water from three vegetables (romaine lettuce, iceberg lettuce and carrots) following two different cutting techniques (chopped and shredded) as well as from whole baby spinach was evaluated at pH 7.0 with time-course sampling over 90 min. In addition, the behavior of chloramine formation was also evaluated. The results are reported in Fig. 1. Shredded-vegetable wash water had higher chlorine demand than chopped/sliced-vegetable wash water due to increased exposure of vegetable tissue generated by shredding compared to chopping/slicing, resulting in enhanced vegetable juice release. As shown in Fig. 1, wash water from shredded carrots had the highest chlorine demand. This is consistent with the data in Tables 1 and 2 where the wash water from shredded carrots had the highest COD and TOC and the largest volume of vegetable juice released per mass of processed produce among four vegetables that were analyzed.

Fig. 1 also shows rapid chlorine consumption and fast chloramine formation following chlorination of all four vegetable wash waters. Over a 90 min observation period, 50% of chlorine demand occurred within the first 5 min, except wash water from baby spinach (47%), as shown in Fig. 2. Chloramine formation, which includes both organic chloramines and inorganic chloramines, was almost complete within 5 min for all four vegetable wash waters with only modest increases in chloramine concentration observed subsequent to this time period. These results are consistent with previous findings in the literature. Deborde and von Gunten (2008),

Table 1
Characteristics of vegetable wash water and vegetable extracts.

Sample	COD (mg/L)		Nitrogen composition			TOC		UV absorbance at wavelength of 254 nm	
	Non-filtered	Filtered	Total N (mg/L)	$NO_3 + NO_2$ (mg/L)	TKN (mg/L)	TOC (mg/L)	DOC (mg/L)	Non-filtered	Filtered
Iceberg lettuce extracts	38,100	34,400	708	293	415	13,200	12,600	24.9	18.1
Shredded iceberg lettuce wash	2420	2420	39.9	16.9	23	686	640	1.19	0.865
Equivalent extract ratio (%)	6.35	7.03	5.64	5.77	5.54	5.20	5.08	4.78	4.78
Romaine lettuce extracts	34,200	22,900	985	208	777	9750	7760	60.2	20.8
Shredded romaine lettuce wash	2580	2420	43.4	19.5	24	692	690	2.41	1.73
Equivalent extract ratio (%)	7.54	10.57	4.41	9.38	3.09	7.10	8.89	4.00	8.34
Carrot extracts	95,600	87,800	664	322	342	29,400	28,200	67.6	42.1
Shredded carrot wash	4830	4530	15.1	11.1	4.02	1930	1780	2.46	1.52
Equivalent extract ratio (%)	5.05	5.16	2.27	3.45	1.18	6.56	6.31	3.64	3.62
Baby spinach extracts	46,400	35,900	2950	544	2410	14,100	11,200	143	86
Baby spinach wash	94.8	48	3.17	0.754	2.42	11.7	9.9	0.292	0.166
Equivalent extract ratio (%)	0.20	0.13	0.11	0.14	0.10	0.08	0.09	0.20	0.19

Sample	IC				
	Fluoride (F^-; mg/L)	Chloride (Cl^-; mg/L)	Nitrate (NO_3^-; mg/L)	Sulfate (SO_4^{2-}; mg/L)	Phosphate (PO_4^{3-}; mg/L)
Iceberg lettuce extracts	n.d.[a]	205	211	14.9	193
Shredded iceberg lettuce wash	n.d.[a]	15.0	15.2	1.04	16.3
Equivalent extract ratio (%)		7.31	7.22	7.00	8.42
Romaine lettuce extracts	17.6	324	253	17.9	105
Shredded romaine lettuce wash	4.21	25.8	17.0	5.16	20.8
Equivalent extract ratio (%)	23.92[b]	7.96	6.73	28.83[b]	19.85[b]
Carrot extracts	70.5	735	n.d.[a]	149	741
Shredded carrot wash	5.12	16.6	n.d.[a]	3.40	17.3
Equivalent extract ratio (%)	7.27	2.25		2.28	2.34
Baby spinach extracts	n.d.[a]	417	721	5.96	306
Baby spinach wash	n.d.[a]	1.41	1.92	2.12	n.d.[a]
Equivalent extract ratio (%)		0.34	0.27	35.50[b]	

[a] n.d.: not detected or below detection limit.
[b] These measurements were excluded in the data analysis of Table 2.

for example, reported that chlorination of nitrous compounds involves an electrophilic substitution reaction, where a chloride atom substitutes a hydrogen atom on amine functional group and results in rapid formation of chloramines.

Effects of filtration (0.45 μm) on reduction of chlorine demand and chloramine formation were also evaluated. Filtration has been integrated into water treatment settings for decades and has shown high efficiency to remove solids from water (Zularisam, Ismail, & Salim, 2006). Moreover, Nelson, Singh, Toledo, and Singh (2007) investigated the application of a submerged membrane microfiltration system in a fresh-cut vegetable washing operation and showed high removal of particles and total solids. However, our results demonstrated a low efficiency for filtration to reduce chlorine demand of wash waters from iceberg lettuce, romaine lettuce and carrots following two different cutting techniques (chopped and shredded), ranging from 4.7% to 25.9% after 90 min contact time (black bars in Fig. 3). This modest reduction was most likely due to removal of chlorine demand originating from coarse particles, such as pieces of vegetable tissue. The results indicate that among these three vegetable wash waters, most of the chlorine demand comes from dissolved compounds, not from particles, with the dissolved compounds mostly generated by the release of vegetable juice during washing. This result is consistent with the finding of Van Haute, Uyttendaele, and Sampers (2015), where little or no dissolved organic was removed by removing particles (turbidity). Moreover, the reduction of chloramine formation by filtration is higher than removal of chlorine demand, ranging from 12.1% to 43.7% among three vegetable wash waters (grey bars, Fig. 3).

Interestingly, the highest chlorine demand removal efficiency and chloramine formation following filtration is found in baby spinach wash water; 37.6% and 59.2%, respectively (Fig. 3). These results are most likely due to whole-leaf washing applied to baby spinach. A greater portion of chlorine demand in whole-leaf wash water comes from foreign compounds (*e.g.*, soil) than in fresh-cut vegetable wash water. Those foreign compounds were usually induced in the form of small solids that could be removed by filtration.

Data from Table 1 and Fig. 2 were analyzed for correlations among water quality parameters and chlorine demand for four vegetables. Among all parameters which were examined in this study, the concentrations of COD and TOC in wash water had the best linear correlations with chlorine demand, as shown in Table 3. The high correlation between COD and chlorine demand can be explained in part because the COD concentration represents the concentration of compounds that could be chemically oxidized and chlorine is a strong oxidant available to oxidize those compounds

Table 2
The average and standard deviation of equivalent juice ratio and the equivalent volume of vegetable juice released from three fresh-cut vegetable wash waters and baby spinach wash water.

Sample	Average of equivalent juice ratio (%)	Standard deviation	Equivalent volume of vegetable juice released (mL/Kg)
Shredded iceberg lettuce wash water	6.16	1.15	82.1
Shredded romaine lettuce wash water	7.09	2.37	94.5
Shredded carrot wash water	3.95	1.94	158
Baby spinach wash water	0.17	0.08	2.26

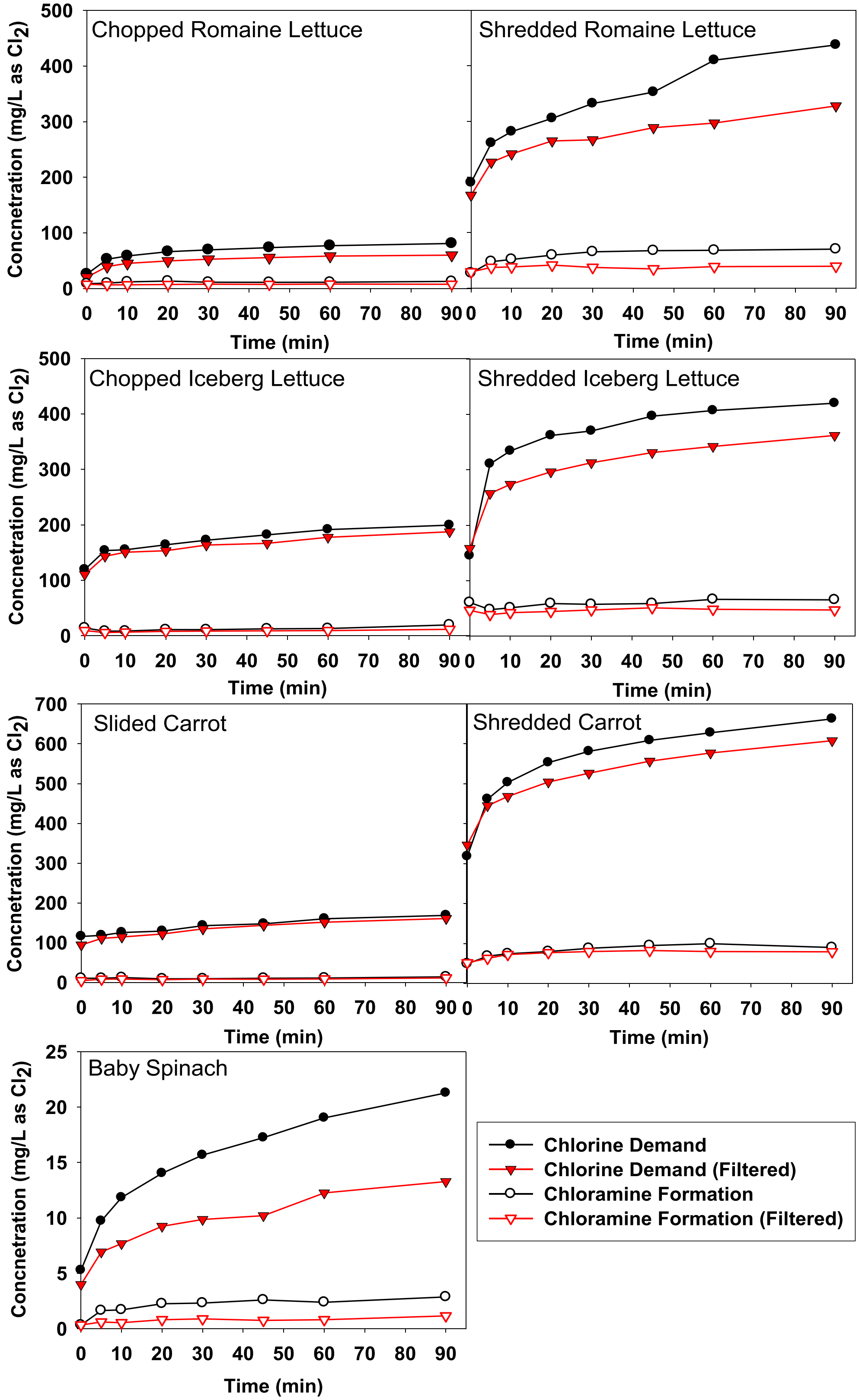

Fig. 1. Kinetics of chlorine demand and chloramine formation over 90 min contact time from chlorination of fresh-cut vegetable wash waters with or without filtration.

contributing to COD. Moreover, since organic compounds are the major source of COD in wash water, the high correlation between TOC and chlorine demand is expected. The third highest correlation is between chlorine demand and UV_{254} absorbance (a standard water quality parameter analyzed in drinking water). UV_{254} absorbance is used to estimate organic content. Compounds with UV_{254} absorbance, however, only represent part of organic wash water content, resulting in a slightly lower correlation of UV_{254}

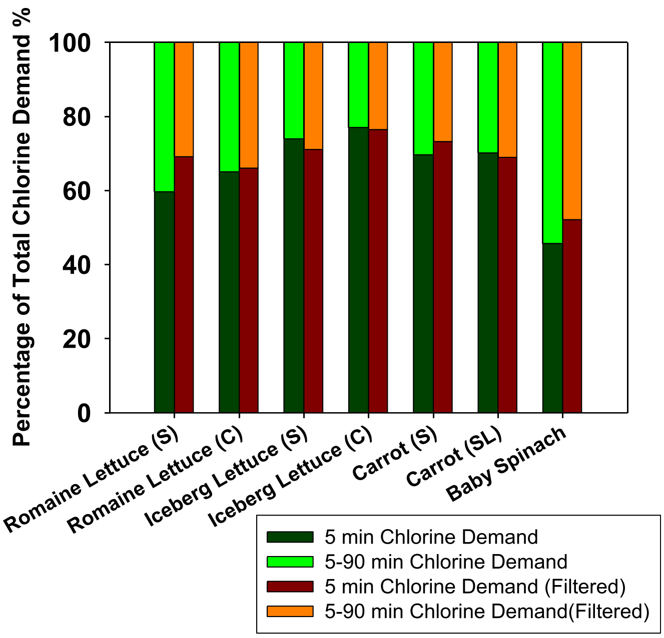

Fig. 2. The distribution of time-dependent chlorine demand of vegetable wash waters with or without filtration over 90 min contact time. S = shredded; C = chopped; SL = sliced.

absorbance than of COD or TOC. These results imply that the concentration of COD and TOC could be used as indicators for chlorine demand during washing process with an online real-time monitoring system.

Additionally, the effects of the number of simulated wash cycles on chlorination kinetics and chloramine formation were also evaluated. Romaine lettuce was selected as the target vegetable and washing experiments were conducted as described above. Water samples were collected after the 5th, 10th and 15th simulated wash cycle, and these samples underwent chlorination experiments in triplicate to assess chlorine demand and chloramine formation.

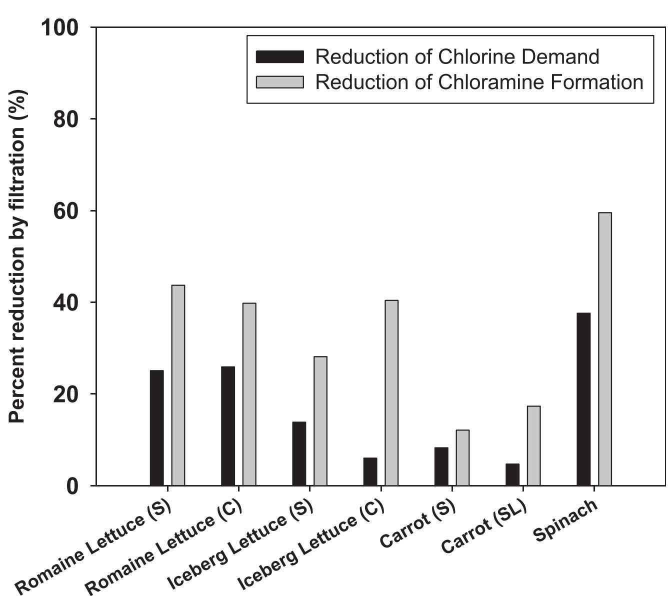

Fig. 3. The effects of filtration on reduction of chlorine demand and chloramines formation over 90 min in four vegetable washing waters. S = shredded; C = chopped; SL = sliced.

Table 3
Correlations between chemical properties of vegetable washing water and chlorine demand at 90 min.

Parameters	R^2
COD	0.99
Total N	−0.15
$NO_3^- + NO_2^-$	0.19
TKN	−0.31
TOC	0.89
UV_{254} absorbance	0.73
Cl^-	0.36
NO_3^-	−0.35
SO_4^{2-}	0.45
PO_4^{3-}	0.63

With increasing numbers of simulated wash cycles, chlorine demand increased (Fig. 4(a)), as did chloramine formation (Fig. 4(b)). The behavior of chlorination kinetics over 90 min was consistent among three different numbers of simulated wash cycles; 59.6–64.5% of chlorine demand had been fulfilled within 5 min contact time with chlorine. Moreover, the efficiency of chlorine demand removal by filtration is similar (18.1%, 17.4%, and 19.7% for 5 simulated wash cycles, 10 simulated wash cycles, and 15 simulated wash cycles, respectively). Since demand was caused by chlorine-demanding substances in wash water, our results indicate that more of these substances were induced into wash water from vegetables with higher number of simulated wash cycles, which could be translated to the amount of processed vegetables. Our results were consistent with the observation by Luo (2007) that concentrations of COD and turbidity had a positive correlation with the mass of processed produce.

3.3. Changes to wash water quality after chlorination

HPLC-SEC analysis separates compounds based on molecular size resulting in the ability to identify groups of compounds with specific molecular weights by comparing the target analyte with the retention time of a series of standardized compounds (polystyrenesulfonic acid sodium salt; Fluka) with a known molecular weight. This method has been used to study natural organic matter which is known to contribute to TOC and to be a source of chorine demand in drinking water (Chow, Fabris, Leeuwen, Wang, & Drikas, 2008; Her et al., 2002). In this study, HPLC-SEC was applied on vegetable wash water to evaluate which group of compounds contributes to chlorine demand based on molecular weight. The HPLC-SEC chromatographs from wash waters from four vegetables with/without chlorination were compared, and the results are shown in Fig. 5. The majority of compounds in four wash waters have molecular weights around 4300 Da or less (Figure S1 in Supporting Material). The differences in chromatographs from samples with and without chlorination reveal compounds in wash water that had been modified by chlorine. Among four vegetables, the minimum chromatographic difference was observed with spinach wash water, due to the low chlorine demand. On the other hand, wash water from romaine lettuce showed the largest change in chromatographs before and after chlorination. These results indicate a large composition change by chlorine which is consistent with our previous observation of high chlorine demand in romaine lettuce wash water. Although wash water from carrots and iceberg lettuce also presented high chlorine demand, there were only moderate changes in chromatographs before and after chlorination, indicating most of the compounds structurally did not change. The results suggest that chlorine, which was consumed, underwent only electrophilic substitution, instead of completely degrading the compounds

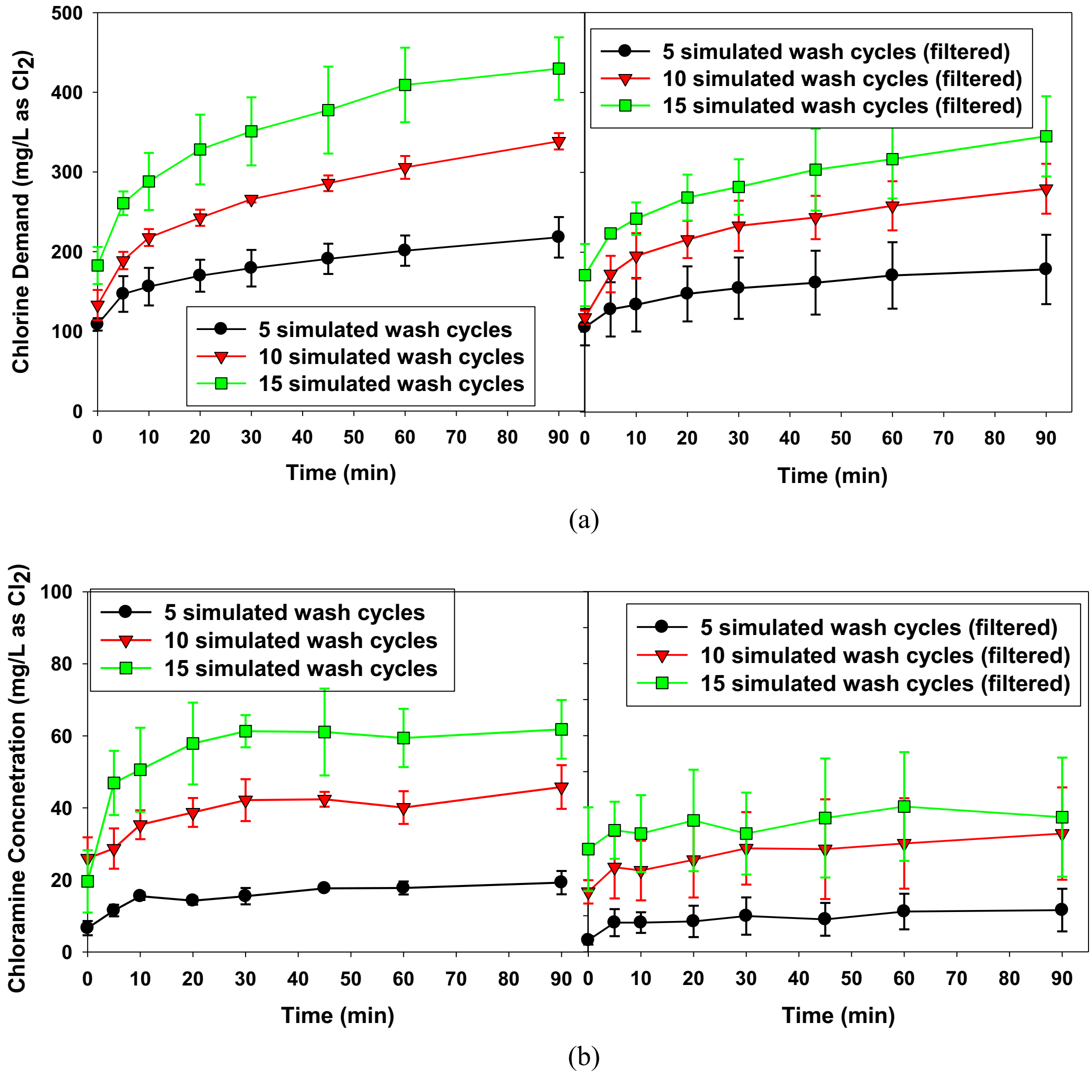

Fig. 4. (a) Chlorine demand kinetics and (b) chloramines formation over 90 min from chlorination of shredded romaine lettuce wash water after 5 simulated wash cycles, 10 simulated wash cycles, and 15 simulated wash cycles with and without filtration.

(Deborde & von Gunten, 2008). Nevertheless, a portion of compounds in iceberg lettuce and carrot wash waters were degraded to smaller molecular weight compounds after chlorination, as shown in a hump at the right side of the main peaks. The HPLC-SEC results show the fraction of compounds that contribute to chlorine demand which could facilitate development of treatment process that specifically target interfering substances within this fraction. It is important to note that our results represent the change of compounds with absorbance only at wavelength of 254 nm, which does not represent all constituents in wash water. A comprehensive study with multi-wavelength (or more detail compound separation) would be necessary for complete molecular-weight-based chlorine demand profiling. These findings indicate that UV_{254} absorbance is not the only parameter that should be used to evaluate the chlorine demand. From Table 3, the correlation between chlorine demand and UV_{254} absorbance is 0.73, which indicates UV_{254} absorbance is not a perfect parameter to predict chlorine demand. Other parameters need to be considered (*e.g.*, COD and TOC). This information is particularly useful when integrating membrane filtration with certain molecular weight cutoffs as a treatment option for chlorine demand removal during the wash process or for reuse of wash water.

4. Conclusion

Fundamental studies of the characteristics of vegetable wash water are necessary for fresh-cut produce industry to thoroughly understand the impact of water quality on the efficacy of its washing processes. The characteristics of a target water quality are essential for selecting a feasible treatment system (Johns, 1995). As such, our study estimated the rate of constituents entering the wash water in terms of volume of vegetable juice per unit mass of processed produce. This information could be used to estimate chlorine demand in conjunction with an existing model (Zhou et al., 2014). Moreover, this study assessed the chemical properties of produce wash water with a focus on how selected water quality parameters affected chlorination. TOC and COD showed good correlations to chlorine demand regardless of the type of vegetables; consequently, these two parameters have the potential to estimate the amount of chlorine required during washing process. Additionally, SEC results showed changes in molecular weight of the fractions of constituents after chlorination, such changes lend insight into the molecular weight fractions that contribute to the overall chlorine demand of wash water.

The desired outcome is development of an approach that will

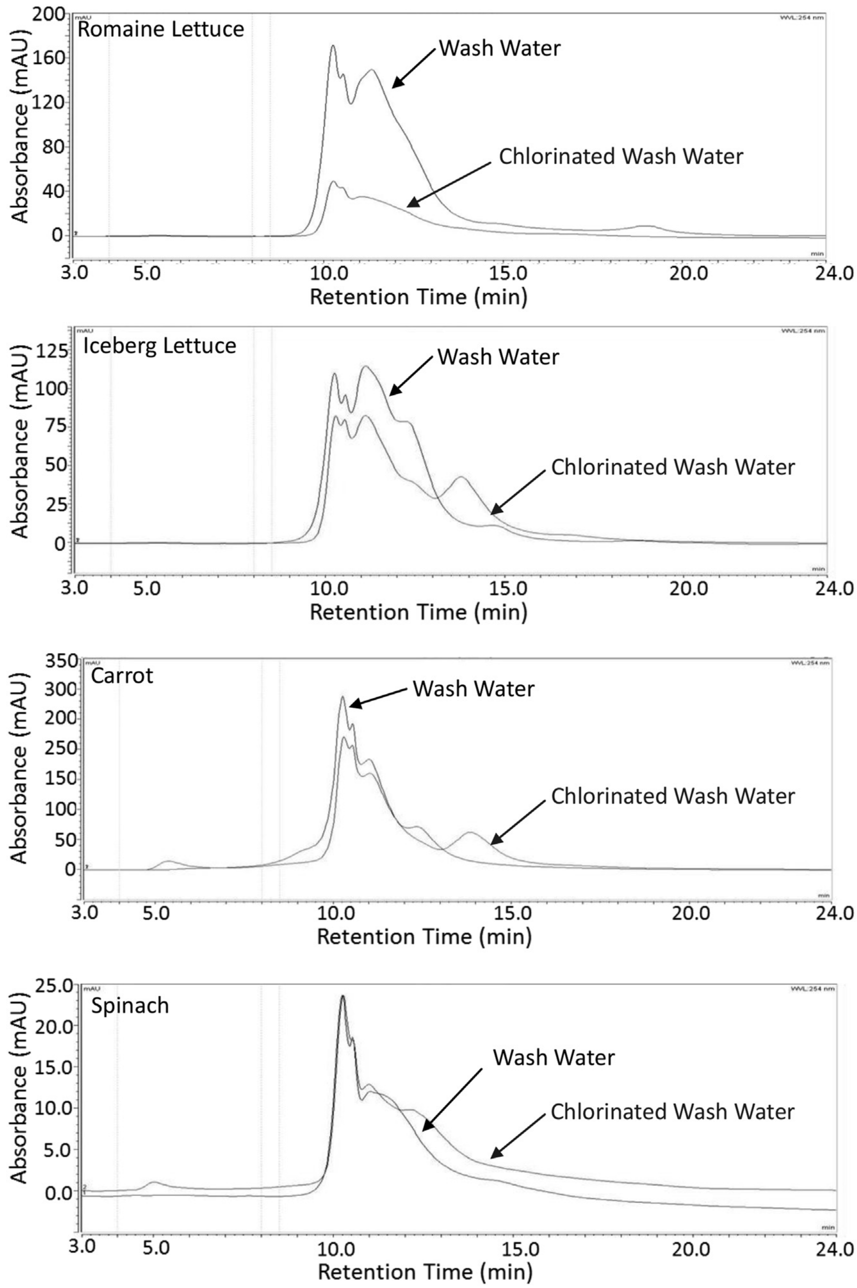

Fig. 5. The HPLC-SEC chromatographs of four vegetable wash waters before (black line) and after (blue line) chlorination at wavelength of 254 nm.

reduce chlorine demand, thus enabling adequate residual chlorine levels to prevent cross-contamination during the entire wash cycle process. The results from this study showed a fast reaction between constituents in wash water and chlorine among four target vegetables. These interactions will be a challenge for any water treatment process designed to reduce chlorine demand once the constituents enter a chlorinated washing basin. From an engineering aspect, the alternative solution is to reduce organic loading input, potentially by having a chlorinated pre-wash or rinse for fresh-cut vegetables or by first having whole-leaf washes (Nou & Luo, 2010).

Substantial quantities of water are used for various purposes during food processing (Dupont & Renzetti, 1998; Fähnrich, Mavrov, & Chmiel, 1998; Poretti, 1990). Due to the large volume used, wash water is becoming significant economic and environmental issues for fresh produce industry, especially when many areas in the U.S. are experiencing water shortages and severe droughts. Despite the limitations and challenges from regulatory and treatment technologies (Casani, Rouhany, & Knøchel, 2005), many factors (*e.g.*, economical and environmental ones) have driven food industry to expand the scale of water reuse. Our study provides information for engineering design of water treatment for water reuse and has implications for future development of treatment strategies for fresh-cut produce industry.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.foodcont.2015.08.031.

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