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Flume and single-pass washing systems for fresh-cut produce processing: Disinfection by-products evaluation

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Abstract

Free chlorine is widely used in fresh-cut produce washing to minimize microbial cross-contamination, but its application can also lead to the formation of harmful disinfection byproducts (DBPs). This study compared DBPs in process water and washed produce during commercial operation. Samples of shredded lettuce and diced cabbages washed in chlorinated water in the traditional double-flume wash and novel immersion-free single-pass wash systems were collected at multiple time points and locations from the processing lines. A suite of 33 conventional and emerging DBPs, including 4 trihalomethanes (THMs), 9 haloacetic acids (HAAs), 9 nitrogenous DBPs (N-DBPs), and 11 carbonaceous DBPs (C-DBPs), were investigated. Overall, N-DBPs and HAAs dominated the concentration of detected DBPs. Wash water in the flumes contained 7-111 folds higher DBP levels compared to that in the single-pass system, with the abundance of DBPs following the order of N-DBPs > HAAs > C-DBPs > THMs. DBPs were detected on washed produce, presenting primarily in the forms of N-DBPs and HAAs followed by C-DBPs and THMs. While higher levels of DBPs were detected in flume-washed produce, the final potable water rinse significantly reduced the DBP concentration in these products. The concentrations of THMs and HAAs on these washed products were used to estimate potential exposure via consumption of fresh-cut produce and the levels were below the daily allowable exposure for drinking water standard established by the U.S. Environmental Protection Agency. The detected high concentration and potential toxicity of N-DBPs indicate the need for further research. The findings of this study advance the understanding of the formation and removal of DBPs during wash processes, and identify additional research opportunities for further reduction in DBPs and food safety improvement.

Keywords: Fresh-cut produce, produce washing, disinfection byproducts, emerging DBPs, flume wash, single-pass wash

1. Introduction

To maintain product quality and safety, fresh-cut vegetable processing usually relies on sanitizers to reduce microbial counts and minimize water mediated microbial cross-contamination (Luo et al., 2011). Chlorine is the most widely used sanitizer in the fresh-cut industry due to high disinfection efficiency, but poses risks of generating harmful disinfection byproducts (DBPs) in produce wash water and on washed produce (Rico et al., 2007; Simpson and Mitch, 2020). Exposure to harmful DBPs is a potential health concern (Richardson and Kimura, 2015). The U.S. Environmental Protection Agency (USEPA) has set the maximum contaminant levels (MCL) for the sum of four trihalomethanes (THMs; chloroform, bromodichloromethane, dibromochloromethane, and bromoform) and five haloacetic acids (HAAs; chloroacetic acid, bromoacetic acid, dichloroacetic acid, dibromoacetic acid, and trichloroacetic acid) in drinking water at 80 and 60 μg/L, respectively (USEPA, 2006). However, the identification of new DBPs with higher toxicity in recent decades raises concern that the regulated THMs and HAAs are inadequate to address the health risk of DBP exposure (Li and Mitch, 2018). Indeed, studies with bioassays have shown that many emerging DBPs, such as haloacetonitriles, halonitromethanes and haloacetamides (i.e., members of the so-called nitrogenous DBPs (N-DBPs)), are far more genotoxic and cytotoxic than THMs and HAAs (Muellner et al., 2007; Plewa et al., 2008; Stalter et al., 2016; Wagner and Plewa, 2017). Currently, the health concern of emerging DBPs is being closely evaluated within the USEPA for possible future actions (USEPA, 2017). Compared to drinking water, there are no regulations related to DBPs for food products including fresh-cut produce. Even so, the occurrence of DBPs in produce processing with chlorine sanitizers is expected, and effective strategies to minimize DBP levels should be developed and implemented.

Following the previous focus of THMs and HAAs in drinking water, literature about DBPs in fresh-cut produce and wash water also mainly focused on THMs and HAAs. The detected DBP levels in produce wash water varied widely and strongly depended on the employed washing conditions and produce types. For instance, a commercial-scale study conducted by Lopez-Galvez and co-workers (2019) found average THM concentrations at 34.1±27.4, 54.4±24.9, and 2635.2±824.9 μg/L in the wash water with chemical oxygen demand (COD) of 34-112, 297-375, and 1012-7092 mg O_2/L for baby spinach leaves, fresh-cut lettuce and shredded products (including carrot, cabbage, and fresh-cut tomato), respectively. The THM concentration in the wash water was quite stable in the baby leaves and fresh-cut lettuce lines, while it decreased in the shredded vegetables line. In a lab-scale tests, Tudela and co-workers (2019) observed that both THM and HAA concentrations in produce wash water from the highest to the lowest levels as cabbage > onion > baby leaves > lettuce at different free chlorine levels of 8-36 mg/L and COD levels up to 2220, 3150, 266, and 942 mg O_2/L for cabbage, onion, baby leaves, and lettuce, respectively. Moreover, Van Haute and co-workers (2013) evaluated DBPs in fresh-cut lettuce process water after one hour of washing and found 27.8±5.4 µg/L and 124.5±13.4 µg/L total THMs in process water with COD of 500 and 1000 mg O_2/L, respectively.

Regarding DBP residues in washed produce, the commercial-scale study conducted by Lopez-Galvez and co-workers (2019) found average THM concentrations of 19.9±8.5, 16.2±11.5, and 19.4±20.2 µg/kg in baby leaves, fresh-cut lettuce, and shredded vegetables, respectively, from several produce processing lines. In a lab-scale study, Gomez-Lopez and co-workers (2013) evaluated the washing of baby spinach with 2-4 mg/L chlorine and reported the detection of 3.9±0.7 ng/g chloroform, 1.4±0.1 ng/g chlorodibromomethane, and 1.5±0.2 ng/g bromoform in the washed spinach. Cardador and Gallego (2012) measured 9 HAAs in various vegetables and ready-to-eat salads from markets. More than 100 samples were analyzed and 23 samples showed positive results, among them, dichloroacetic acid (0.5-6.4 µg/kg) and trichloroacetic acid (0.5-193 µg/kg) were found in almost all of the positive samples.

Recently, Lee and co-workers investigated a broad range of conventional and emerging DBPs for their formation during fresh-cut lettuce washing with chlorine and other sanitizers (Lee and Huang, 2019; Lee et al., 2018; Lee et al., 2019). The results showed that dominant DBPs formed during chlorine wash were HAAs, N-DBPs and aldehydes in the wash water, and THMs, HAAs and aldehydes in the washed lettuce. In addition, the distributions of major DBPs were different in wash water versus in washed lettuce. Considering the higher toxicity of emerging N-DBPs compared to the regulated DBPs, many knowledge gaps still remain to be addressed regarding the generation of emerging DBPs during fresh-cut processing and the suitable strategies to control DBPs overall.

In the commercial line of fresh-cut processing, produce is washed conventionally using a flume washing system which typically consists of double flumes with produce moving sequentially from the primary to the secondary flumes (Bornhorst et al., 2018; Luo et al., 2012). Water is often reused and recirculated (with some fresh water make up) throughout each shift (usually 8 hours), leading to significant buildup of organic matter in the wash water over time (Manzocco et al., 2015; Van

Haute et al., 2015). The high organic loads can significantly hinder the effectiveness of chemical sanitizers by exerting oxidant demand and may promote DBP formation (Gil et al., 2009). The single-pass washing system is a novel design to address some of the problems encountered with the flume washing system. The single-pass washing system utilizes water sprays to clean and wash produce at multiple tiers and stages (McEntire et al., 2016). Wash water is not recirculated and thus the produce only interacts with clean wash water. Research has shown the effectiveness of the single-pass washing system for reducing pathogen cross-contamination and maintaining product quality and shelf life (Bornhorst et al., 2018). However, how DBP formation varies between the two washing systems remain unknown.

This work investigated DBP formation during the chlorine wash of shredded iceberg lettuce and diced green cabbage using traditional flume wash and novel single-pass wash. The objective was to compare the two washing systems for their impacts on DBP formation by assessing a wide range of conventional and emerging DBPs. The DBP formation was evaluated from several aspects, including different washing systems, types of produce, and wash water versus washed produce. The abundance of DBPs was compared based on individual compounds and subgroups of compounds for their distribution in wash water and produce, respectively.

2. Materials and Methods

2.1 Fresh-cut produce processing plant

Samples of shredded iceberg lettuce (*Lactuca sativa* var capatata) and diced green cabbages (*Brassica oleracea* var. capitata) and the associated produce wash water were collected from the washing lines at a medium-size fresh-cut produce processing plant in the USA during its commercial operation. The processing plant was equipped with several traditional double flume systems and single-pass washing systems in operation side-by-side. For this study, the average processing rate of the plant was about 30 and 50 kg min^{-1} in the flume system and 20 and 30 kg min^{-1} in the single-pass system for iceberg lettuce and cabbage, respectively. The whole plant was operated under approximately 4°C. Before washing, iceberg lettuce was shredded into 6-mm strips using a TranSlicer® 2510 Cutter (Urschel Laboratories Inc., Chesterton, IN, USA) and cabbage was diced into 6-mm squares using a Diversa Cutter (Urschel Laboratories Inc.). The input water for both washing systems was potable water chilled to 4°C, dosed with chlorine (~20 mg/L) and pH controlled at 5.7-6.2. The potable water consisted of reclaimed water that was treated by an on-site water treatment facility and blended with water obtained from a groundwater well and/or from the city's drinking water supply. The spent wash water was collected at the on-site water treatment facility (with capacity of 750 L min^{-1}) and treated by conventional processes of coagulation, flocculation and sedimentation, followed by advanced processes of ultrafiltration, reverse osmosis and ultraviolet photolysis before reclamation.

As shown in Figure 1a, the flume system consisted of a primary flume (9000 L, labeled as 1st-flume) and a secondary flume (7000 L, 2nd-flume) (Luo et al., 2018). The designed contact time of fresh-cut produce and wash water was ~30 sec for both flumes. During operation, the spent wash water was collected in the catch tank and recirculated to the flume after reconditioning (i.e., adding potable water, replenishing chlorine, adjusting pH, and chilling to 4°C). The recirculated water was not mixed across different catch tanks. A final rinse of produce with potable water (no additional chlorine added) was applied after the 2nd-flume wash. In the single-pass washing system, as shown in Figure 1b, a series of sprayers was installed over the incline belt (incline-wash) and a set of vibrating screens (vibra-wash) to deliver the input water (Bornhorst et al., 2018). The designed contact time of fresh-cut produce and sprayed input water was ~30 sec for both incline-wash and vibra-wash. The spent wash water in the single-pass system was collected and discharged to the on-site water treatment facility.

2.2 Sample collection and storage

Two sampling events were conducted from the same processing plant at different times on different days, i.e., the first sampling occurred in the middle point of the first shift (11 am to 12 pm), and the second sampling occurred within one hour after processing started (8 am to 9 am). The first time of sampling was done for shredded lecture, whereas the second time of sampling was done for shredded lecture and diced cabbage. The second sampling event also included sample duplicates. During the 1st sampling, the wash water in the flume had residual chlorine level fluctuated between 3.7 and 20.0 mg/L (average 8.5±3.0 mg/L) and pH varied between 5.1 and 6.2 (average 5.7 ±0.2). Meanwhile, the input water in the single-pass system had more stable residual chlorine level between 18.2 and 21.3 mg/L (average 20.0±0.6 mg/L) and stable pH between 6.0 and 6.4 (average 6.2±0.07).

As shown in Figure 1, the fresh-cut produce samples were collected (i) from the de-watering shaker (of the 1st/2nd-flume) and after the final rinse in the flume system, and (ii) from the incline belt and the vibrating screens in the single-pass system. The samples of unwashed shredded lettuce and diced cabbage were collected as controls. The produce samples of shredded lettuce and diced cabbage were immediately sealed into Whirl-Pak^{TM} sterile bags and stored in a cooler with ice after collection. The produce wash water samples were collected from (i) the 1st and 2nd troughs of the flume system, and (ii) beneath the incline belt and the vibrating screens in the single-pass system where water had flowed pass the cut produce. Wash water samples were collected in clean amber borosilicate bottles (500 mL) with sodium thiosulfate (to yield ~6 mM) to quench residual chlorine immediately upon water collection, and then stored in a cooler with ice. No headspace was allowed in the sample bottle to minimize volatile loss. Samples of well water, city potable water, and reclaimed water from the on-site water treatment facility were collected as controls in a similar manner. All samples were transported to the research lab on the same day and stored at 4-5°C (water samples) or 0°C (produce samples). Samples were extracted within three days and analyzed within two weeks.

2.3 Sample analysis

Table 1 lists the 33 DBPs investigated, including 4 THMs, 9 HAAs, 9 N-DBPs, and 11 carbonaceous DBPs (C-DBPs) and their abbreviations. The sources of DBP standards and other chemicals and reagents are provided in Supporting Information (SI) Text S1. The quantification of the DBPs in wash water and produce samples was conducted following the analytical methods established previously (Lee et al., 2018; Lee et al., 2019). Briefly, the DBPs were divided into two groups (Group I included THMs, N-DBPs and C-DBPs; Group II included HAAs) due to the higher polarity and lower volatility of HAAs than the rest of DBPs. The produce samples were homogenized by a food processor. Then, homogenized produce and produce wash water went through the same liquid-liquid extraction process using methyl *tert*-butyl ether (MTBE) as organic phase. For Group I compounds, the extracted MTBE layer was analyzed directly by capillary column GC/ECD with detailed parameters described in the previous work (Lee et al., 2018). For Group II compounds, the extracted MTBE layer was mixed with 10% (v/v) sulfuric acid in methanol and heated at 51±2 °C for 2 h to derivatize the HAAs to their methyl esters. Afterward, the HAAs methyl esters were analyzed using capillary column GC/ECD. The details of analytical method including instrument operation parameters, method detection limits and extraction recoveries were documented in the previous publications (Lee et al., 2018; Lee et al., 2019).

3. Results and Discussion

The results of detected DBPs in the water and produce samples are presented by individual compounds (Tables 2 and 3) and by the subgroups of THMs (4), HAAs (9), N-DBPs (9), and C-DBPs (11) (Figures 2-5 and S1). As detailed below, DBP formation is evaluated based on (i) different washing systems, (ii) different types of produce and the same type of produce but collected from different sampling events, (iii) groups/subgroups of DBPs and individual DBPs, and (iv) DBPs detected in produce wash water versus in washed produce. DBP formation may also be affected by produce cut size, but this effect was not investigated in this study.

As a control to check the background DBPs in potable water (before dosing chlorine for the washing systems), the DBP levels in well water, city water, and reclaimed water were measured and compared with the DBP levels in the input water (chlorine dosed). It was found that the total DBP level in input water was higher than those in city water and reclaimed water in the 1st sampling event, but comparable or lower in the comparison in the 2nd sampling event (Figure S1). Negligible amounts of DBPs were detected in well water in both sampling events. In general, all the water sources including input water contained a low level of total DBPs (≤163.3 μg/L) and did not exceed the MCLs of THMs and HAAs for drinking water.

3.1 Formation of DBPs in wash water

3.1.1 DBPs in wash water from flume and single-pass washing systems

In the flume wash of shredded lettuce, high abundance of total DBPs (165.6-2173.2 μg/L) was measured in the wash water, indicating a significant increase from the level of 41.1-163.3 μg/L in input water (Figures 2a and 2b). The total DBP levels were higher in the first flume wash water than in the second flume wash water, likely due to a greater amount of exudates leached from cut produce into wash water in the first flume. This finding is consistent with the earlier work by Bornhorst and co-workers on a similar flume washing line that reported average chemical oxygen demand (COD) of 1650 mg/L and 1150 mg/L in the first and second flume wash water, respectively, during a 2.7-hour continuous monitoring (Bornhorst et al., 2018). Interestingly, the two sampling events yielded different results, with total DBP levels in lettuce flume wash water to be higher in the 1st sampling event (Figure 2a) than in the 2nd sampling event (Figure 2b).

Comparing shredded lettuce and diced cabbages during the second sampling event, wash water in the flume with diced cabbage contained 10 times more total DBPs (5898.8-8115.8 μg/L) than that of shredded lettuce (Figure 2c). Bornhorst and co-workers (2018) observed a much higher COD level in diced cabbage wash water than that of the shredded lettuce wash water. This is likely attributable to the higher solid content (dry weight) and chlorine demand in cabbages (Teng et al., 2021), as well as the higher throughput in its wash water, when compared to the shredded lettuce. The total DBP level was also higher in the first flume water than in the second flume water similar to shredded lettuce washing.

Compared to the flume washing system, the single-pass washing system generated much lower levels of total DBPs (41.6-186.1 µg/L) in the wash water (Figure 3), which were around 7-12 times and 111 times lower for shredded lettuce and diced cabbage, respectively. The significantly lower DBP formation was likely because water was used one time without reuse or recirculation in the single-pass system. Indeed, a lower COD level was observed in incline-wash water (1250 mg/L for lettuce and 1920 mg/L for cabbage) and vibra-wash water (830 mg/L for lettuce and 1390 mg/L for cabbage) compared to those in flume wash water (1150-1650 mg/L for lettuce and 2000-4550 mg/L for cabbage) based on a study with a similar single-pass washing line (Bornhorst et al., 2018). Furthermore, when free chlorine concentration in the wash water was compared between the two washing systems, the single-pass system had smaller fluctuation in chlorine concentration (18.2-21.3 mg/L) compared to that in the flume system (3.7-20.2 mg/L). As shown in Figure 3, only a modest increase of total DBPs was observed in incline-wash water and vibra-wash water compared to input water for both lettuce and cabbage during the single-pass wash. Additionally, there was no obvious increase of total detected DBPs along the washing line in the single-pass system, i.e., similar total DBP concentrations were observed in incline-wash water and vibra-wash water.

3.1.2 Major DBPs detected in wash water

In the flume wash, among all samples of lettuce wash water, the abundance of detected DBP subgroups showed the trends of N-DBPs > HAAs > C-DBPs > THMs (Figures 2a and 2b; Table 2). For the group of N-DBPs, DCAN, DCAAm, and TCAAm were detected in high abundance in lettuce wash water. For instance, (1) 783.6-1089.2 µg/L DCAN and 246.2-295.2 µg/L DCAAm were detected during the 1st sampling event; (2) 166.5 µg/L DCAN and 310.3 µg/L TCAAm were detected during the 2nd sampling event. For the group of HAAs, DCAA and TCAA dominated the high abundance of detected HAAs in lettuce wash water. Specifically, 312.0/298.2 µg/L DCAA and 72.7/122.9 µg/L TCAA were detected in the 1st/2nd-flume, respectively, during the 1st sampling event. In the group of C-DBPs, PentaCPN dominated the high abundance with detected concentrations of 156.8/249.2 µg/L in the 1st/2nd-flume, respectively, during the 1st sampling event.

In the flume wash, the subgroup of N-DBPs was also the most abundant DBPs in cabbage wash water (Figure 2c). More specifically, the high concentration of detected N-DBPs was largely attributed to the high abundance of TCAAm (5794.4 μg/L in the 1st-flume and 5351.8 μg/L in the 2nd-flume) and DCAN (1069.0 μg/L in the 1st-flume and 318.7 μg/L in the 2nd-flume) (Table 2). In addition, the high abundance of THMs was observed in cabbage wash water during the flume wash. The high abundant THMs in the 1st-flume was dominated by DBCM (468.0 μg/L).

In the single-pass wash, the increase of DBP level in the wash water was determined by subtracting the DBP concentration in the input water (Figure 3). The results indicated that the subgroups of C-DBPs and HAAs contributed predominantly to the detected DBPs in lettuce wash water (Figures 3a and 3b). Specifically, 30.6 μg/L and 31.8 μg/L TetraCPN were detected in incline-wash water and vibra-wash water, respectively during the 2nd sampling event (Table 2). DCAA contributed predominantly to the detected HAAs in lettuce wash water from the single-pass wash. In cabbage wash water, HAAs concentrations increased while other DBP concentrations remained consistent compared to those in input water as background (Figure 3c). To be noted, the increase of detected HAAs in cabbage vibra-wash water was attributed to BDCAA (31.6 μg/L) which was not observed to be important in other water wash samples (Table 2). Overall, all DBPs in wash water samples collected during the single-pass wash were detected with insignificant concentrations. In addition, unlike the flume system, N-DBPs were not observed to be formed significantly in the single-pass wash across all collected wash water samples.

3.2 Detected DBPs in washed produce

3.2.1 DBPs in washed produce from flume and single-pass washing systems

Varying amounts of DBPs were detected in shredded lettuce from both sampling events and various wash stages, ranging from 189.4 ng/g to 1983.4 ng/g (Figure 4). Higher DBPs in washed lettuce were observed in the 1st sampling event than in the 2nd sampling event (Figures 4a and 4b). This is consistent with the higher level of DBPs observed in produce wash water (Figures 2a and 2b). Meanwhile, the total detected DBPs in washed lettuce in the 1st and the 2nd flumes varied without a consistent trend (Figures 4a and 4b), suggesting that many factors may impact the final DBPs on the washed produce, in addition to the DBP level in the wash water. It is important to note that, washed lettuce samples collected after the final rinse step significantly reduced the amount of total DBPs in lettuce, compared to that after the 2nd-flume wash. This is consistent for both sampling events, with a reduction of DBPs from 1983.4 ng/g to 1044.6 ng/g in the 1st sampling event and from 310.0 ng/g to 189.4 ng/g in the 2nd sampling event (Figures 4a and 4b).

Diced cabbage contained slightly less DBPs in comparison with shredded lettuce during the same sampling event (Figures 4b and 4c), despite of the 10-times higher DBPs present in the cabbage wash water (Figures 2b and 2c). This disparity may be attributable to the following multiple factors: 1) cabbage has much higher density and solid content, and thus smaller surface to mass ratio than lettuce; 2) cabbage surface has more waxy cuticles that are more resistant to water uptake; and 3) possibly weaker absorption of DBPs to cabbage. The result indicated that the very high abundance of N-DBPs (5774.7-7203.6 μg/L) in cabbage wash water in the flume wash did not impact the N-DBP concentration in cabbage samples.

Same as flume, DBPs in produce washed from the single-pass system varied among sampling events and produce types (Figure 5). In most samples, DBP concentration was lower in washed lettuce collected from the single-pass than those from the flume (Figures 4 and 5), although the difference was not as pronounced as that in the wash water. Similarly, lower DBP concentration was observed in diced cabbage washed in the single-pass system than that in the flume. Overall, the relatively higher DBP level in samples from the flume system and the increase of DBPs with the increase of processing time in the flume system suggested that the single-pass system could be beneficial in maintaining a lower DBP level in water and washed produce in situations when the anticipated DBPs were high.

3.2.2 Major DBPs detected in washed produce

In general, the washed produce samples of shredded lettuce and diced cabbage were all detected with subgroups of HAAs and N-DBPs as dominant detected DBPs (Figures 4 and 5) in both flume wash and single-pass wash systems. However, washed lettuce contained a higher abundance of N-DBPs than HAAs, but washed cabbage contained more HAAs than N-DBPs. More specifically, as shown in Table 3, DCAA, CDBAA, and BDCAA were the dominant HAAs, and DCAAm and TCAAm were the dominant N-DBPs, detected in washed lettuce. In washed cabbage, DCAA and BDCAA were detected as dominant HAAs, and DCAAm as the dominant N-DBP.

In addition, the dominant DBPs in each subgroup were compared between produce wash water and washed produce. For instance, in the subgroup of HAAs, DCAA and TCAA were detected with high abundance in lettuce wash water; in comparison, DCAA, CDBAA, and BDCAA were detected with relatively high abundance in washed lettuce. In the case of cabbage, DCAA dominated the subgroup of HAAs in both cabbage wash water and washed cabbage. For the subgroup of N-DBPs, while DCAN, DCAAm and TCAAm were the most dominant ones detected in wash water and washed produce of lettuce and cabbage, DCAAm and TCAAm exhibited much higher abundance in washed produce than DCAN. Effort was made to try to correlate the dominated DBPs in produce wash water and washed produce. However, although there were the same dominant DBPs observed in produce wash water and washed produce, there was no clear correlation obtained by comparing the wash water samples and the washed produce samples collected from the same sampling locations. A lab-scale study of washing fresh-cut lettuce with chlorine sanitizers conducted in our previous study also observed differences of dominant DBPs in wash water versus in washed produce (Lee and Huang, 2019). The differences could be related to (i) varied formation potential of different DBPs in wash water and on washed produce during chlorine reactions and (ii) some properties (e.g., hydrophobicity) of the DBP compounds.

To evaluate the risk of DBPs ingestion via the consumption of fresh-cut produce, the estimation of the DBPs exposure via fresh-cut produce and drinking water was calculated and compared. The HAAs and N-DBPs were the primary DBPs found in this study, and they averaged 131.4 ng/g and 218.4 ng/g in washed lettuce and 64.9 ng/g and 28.5 ng/g in washed cabbage, respectively. Based on an estimation of ~150 g consumption of vegetables per day (Cardador and Gallego, 2012), a person could ingest up to 19.7 μg HAAs and 32.8 μg N-DBPs via washed lettuce or 9.7 μg HAAs and 4.3 μg N-DBPs via washed cabbage. In comparison, a person could intake up to 120 μg HAAs per day from the consumption of 2 L of tap water on the basis of the MCL of HAAs (60 μg/L) (EPA, 2006). Therefore, the HAAs exposure from the consumption of commercially washed fresh-cut produce could be equivalent to one third of the maximum HAAs exposure via tap water on a daily base. To date, there are no regulatory limits for N-DBPs in drinking water and more research is needed to understand the potential health risks associated with exposure to this group of emerging DBPs.

4. Conclusions

This research investigated the formation of DBPs in produce wash water and washed produce during commercial processing of fresh-cut vegetables using different washing systems (traditional double-flume wash and novel immersion-free single-pass wash). In general, the total DBP concentration in wash water was 7-111 folds higher in the flume than in the single-pass, 2-12 folds higher in the late processing stage than the earlier stage, and 0.5-36 folds higher for diced cabbage than for the shredded lettuce. DBPs were also detected in all washed produce, with higher abundance associated with shredded lettuce at later processing stage, and diced cabbage washed in flume. Flume system contained much higher DBP levels in wash water than single-pass, but the difference was less pronounced in washed produce. While higher levels of DBPs were observed with produce washed in flume, the final potable water rinse significantly reduced the DBP concentration in flume washed produce.

Overall, HAAs and N-DBPs dominated the concentration of DBPs detected in wash water and washed produce in both washing systems and produce types. The major HAAs were dichlorinated and trichlorinated acetic acid (DCAA and TCAA), while the major N-DBPs were dichloroacetonitrile (DCAN) and chlorinated acetamides (DCAAm and TCAAm).

Additionally, results obtained within this study suggest that the estimated exposure to HAAs via fresh-cut vegetable ingestion did not exceed the daily allowable exposure for drinking water standard, while more research is needed for emerging N-DBPs due to their higher detected concentrations and potentially greater toxicity than THMs and HAAs. Research findings of this study advance the understanding of the DBP formation and migration as impacted by fresh-cut produce wash system design and operation. It also highlights the importance of consistent application of final portable water rinse in minimizing DBPs when using the flume washed produce.

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Disclaimer

All opinions expressed in this paper are the authors and do not necessarily reflect the policies and views of USDA, ARS, DOE, or ORISE. The authors declare no competing financial interest.

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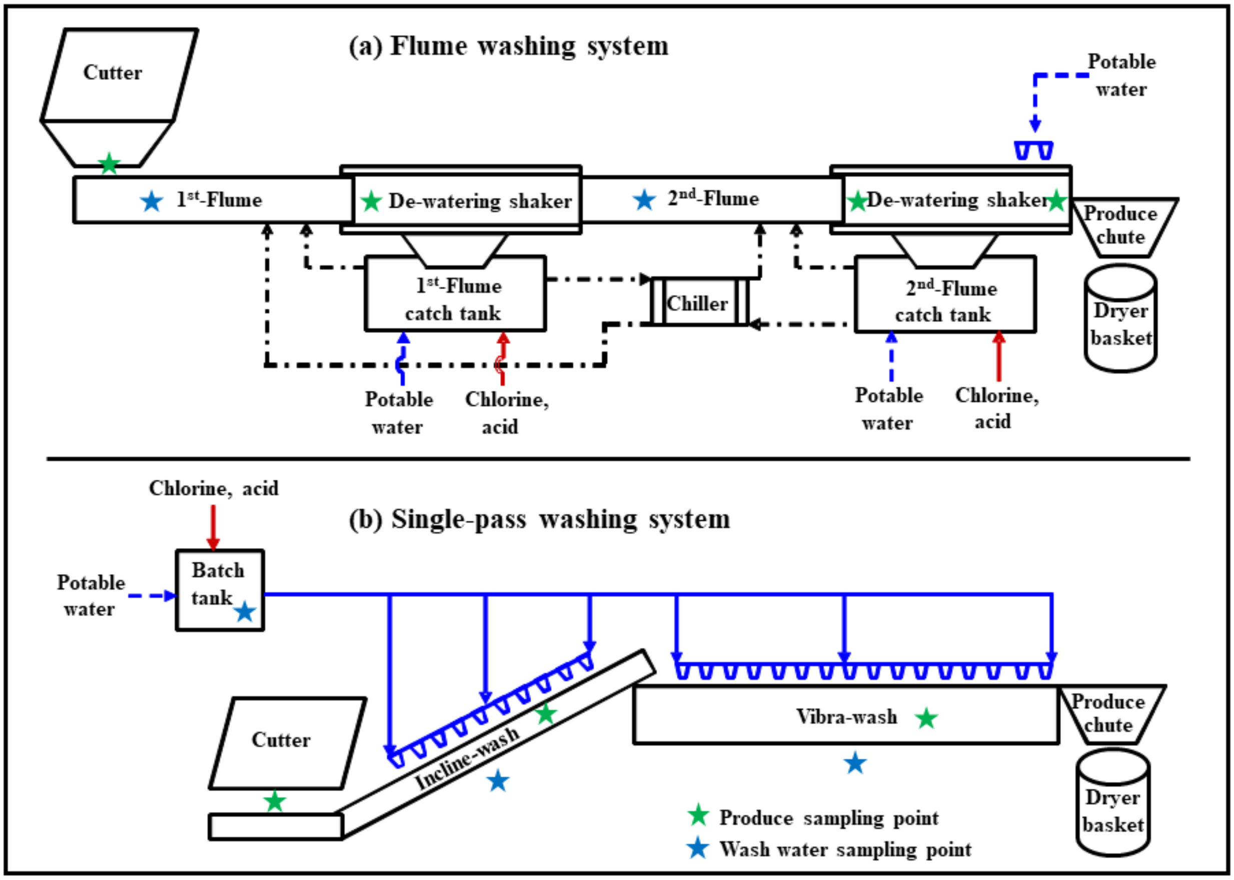

Figure 1. Diagram of two commercial fresh-cut produce washing processes, (a) flume washing system and (b) single-pass washing system (Adaptd with permission from Bornhorst et al., 2018. Copyright 2018 Elsevier). The sampling locations of wash water and produce were labelled with blue star and green star, respectively.

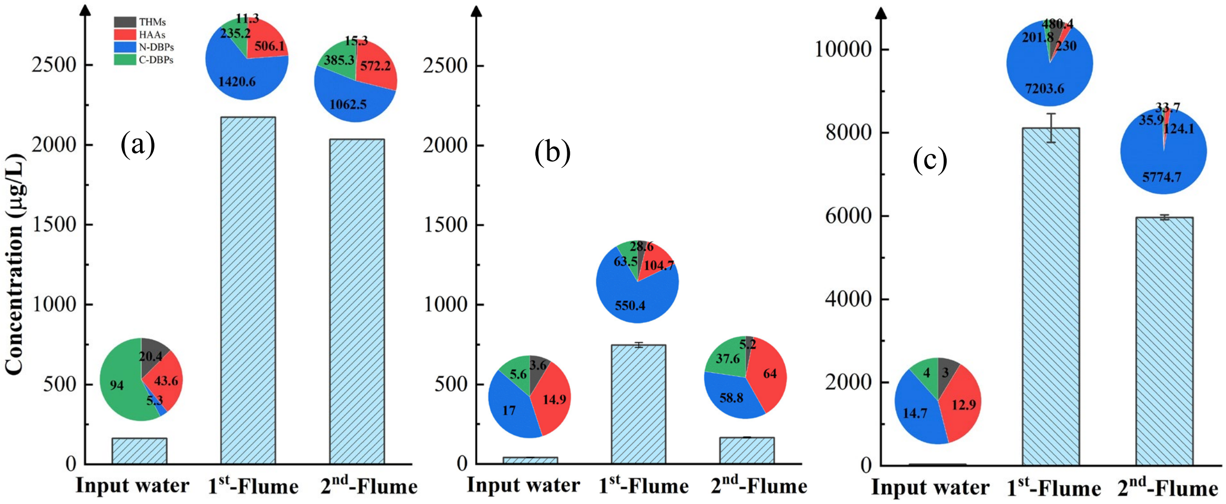

Figure 2. DBP concentrations in the wash water from the washing of shredded iceberg lettuce (a/b) and diced green cabbage (c) in a commercial flume wash system. Numbers in pie charts represent the concentrations of subgroup DBPs in each sample. Samples in (a) were collected during the 1st sampling event, and samples in (b/c) were collected during the 2nd sampling event.

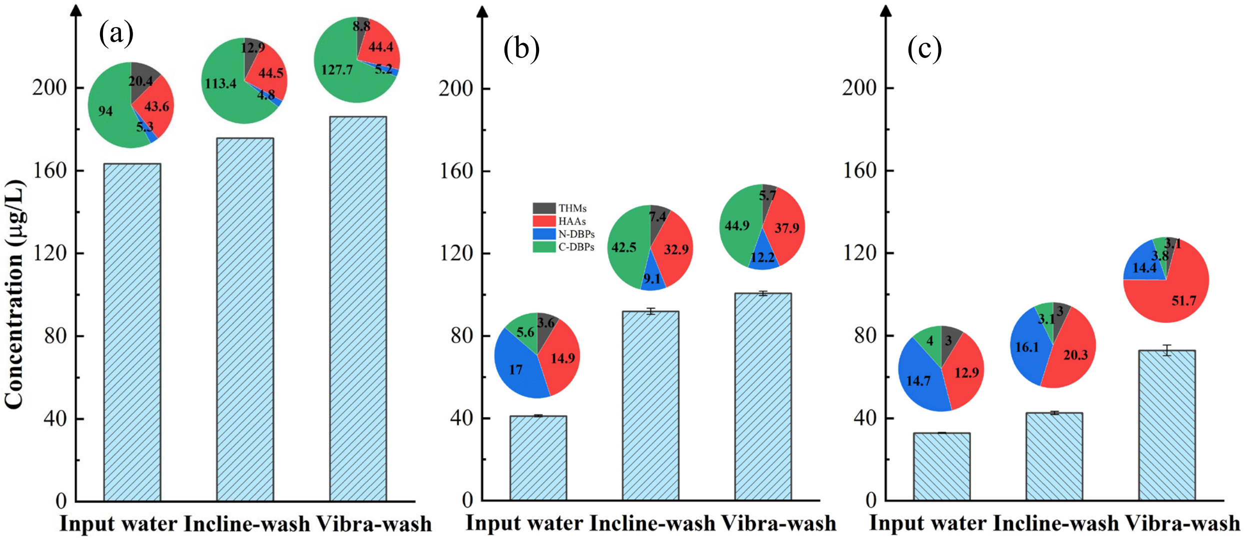

Figure 3. DBP concentrations in the wash water from the washing of shredded iceberg lettuce (a/b) and diced green cabbage (c) in a commercial single-pass system. Numbers in pie charts represent the concentrations of subgroup DBPs in each sample. Samples in (a) were collected during the 1st sampling event, and samples in (b/c) were collected during the 2nd sampling event.

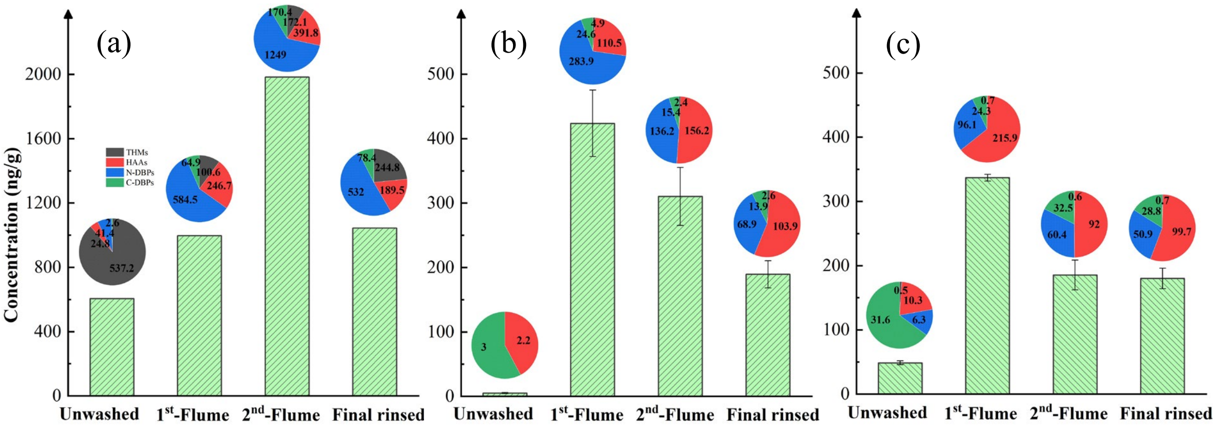

Figure 4. DBP concentrations in shredded iceberg lettuce (a/b) and diced green cabbage (c) collected from a commercial flume wash system. Numbers in pie charts represent the concentrations of subgroup DBPs in each sample. Samples in (a) were collected during the 1st sampling event, and samples in (b/c) were collected during the 2nd sampling event.

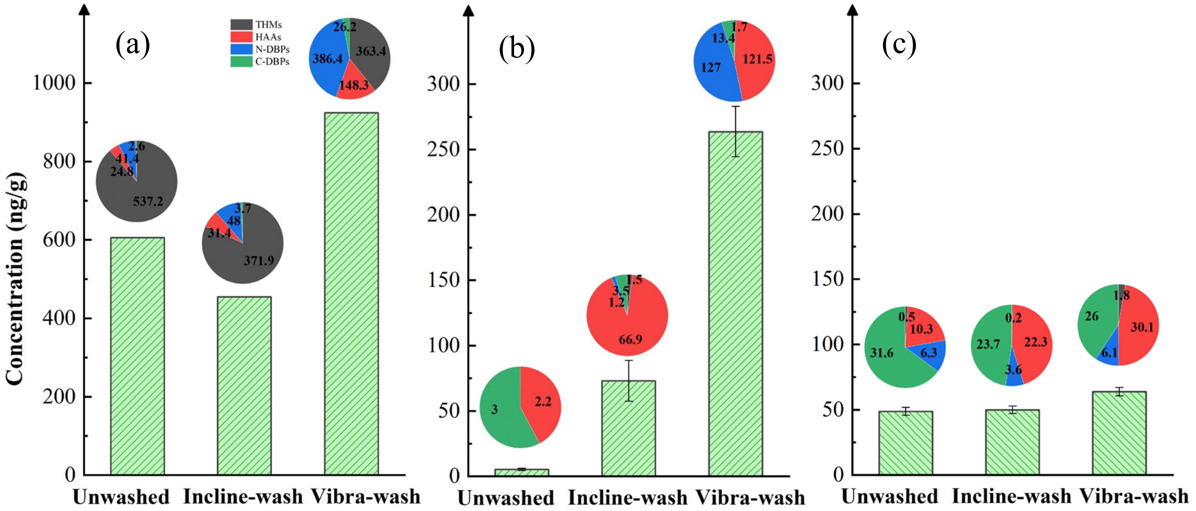

Figure 5. DBP concentrations in shredded iceberg lettuce (a/b) and diced green cabbage (c) collected from a commercial single-pass wash system. Numbers in pie charts represent the concentrations of subgroup DBPs in each sample. Samples in (a) were collected during the 1st sampling event, and samples in (b/c) were collected during the 2nd sampling event.

Table 1. The 33 DBPs investigated in this study.

Group	Target DBPs	Abbreviation	CAS No.
THMs	Chloroform	CF	67-66-3
	Bromoform	BF	75-25-2
	Bromodichloromethane	BDCM	75-27-4
	Dibromochloromethane	DBCM	124-48-1
HAAs	Chloroacetic acid	MCAA	79-11-8
	Bromoacetic acid	MBAA	79-08-3
	Dichloroacetic acid	DCAA	79-43-6
	Trichloroacetic acid	TCAA	76-03-9
	Bromochloroacetic acid	BCAA	5589-96-8
	Dibromoacetic acid	DBAA	631-64-1
	Bromodichloroacetic acid	BDCAA	71133-14-7
	Chlorodibromoacetic acid	CDBAA	5278-95-5
	Tribromoacetic acid	TBAA	75-96-7
N-DBPs	Dichloroacetonitrile	DCAN	3018-12-0
	Trichloroacetonitrile	TCAN	545-06-2
	Bromochloroacetonitrile	BCAN	83463-62-1
	Dibromoacetonitrile	DBAN	3252-43-5
	Dichloronitromethane	DCNM	7119-89-3
	Trichloronitromethane	TCNM	76-06-2
	Bromonitromethane	BNM	563-70-2
	2,2-dichloroacetamide	DCAAm	683-72-7
	Trichloroacetamide	TCAAm	594-65-0
C-DBPs[a]	Chloral hydrate	CH	302-17-0
	Carbon tetrachloride	CTC	56-23-5
	1,1-dichloro-2-propanone	DCPN	513-88-2
	1,1,1-trichloro-2-propanone	TCPN	918-00-3
	1,1,3,3-tetrachloro-2-propanone	TetraCPN	632-21-3
	1,1,1,3,3-pentachloro-2-propanone	PentaCPN	1768-31-6
	1,2-dibromo-3-chloropropane	DBCPN	96-12-8
	1,1,1-trichloroethane	TCE	71-55-6
	Trichloroethylene	TriCEL	79-01-6
	Tetrachloroethylene	TCEL	127-18-4
	1,2-dibromoethane	DBE	106-93-4

[a] C-DBPs (carbonaceous DBPs) excluding THMs and HAAs.

Table 2. Concentrations of DBPs (µg/L) detected in shredded lettuce and diced cabbage wash water from commercial processing.

Group	Target DBPs	Lettuce wash water										Cabbage wash water				
		First sampling event					Second sampling event					Second sampling event				
		Input water	1st-Flume	2nd-Flume	Incline-wash	Vibra-wash	Input water	1st-Flume	2nd-Flume	Incline-wash	Vibra-wash	Input water	1st-Flume	2nd-Flume	Incline-wash	Vibra-wash
THMs	CF	12.1	9.6	12.0	8.7	6.0	1.8 ± 0.1	5.9 ± 0.1	3.9 ± 0.0	6.0 ± 0.1	4.6 ± 0.1	1.4 ± 0.1	10.4 ± 0.0	3.8 ± 0.1	1.0 ± 0.0	1.5 ± 0.4
	BF	0.3	0.2	0.3	0.1	0.2	1.3 ± 0.0	1.2 ± 0.1	0.9 ± 0.0	0.8 ± 0.0	0.6 ± 0.0	1.2 ± 0.0	1.6 ± 0.1	1.6 ± 0.1	1.5 ± 0.0	0.9 ± 0.2
	BDCM	5.6	1.5	1.1	2.8	1.6	0.3 ± 0.0	0.3 ± 0.0	0.2 ± 0.0	0.4 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.4 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.5 ± 0.1
	DBCM	2.4	ND	1.9	1.4	1.0	0.2 ± 0.0	21.2 ± 0.8	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	0.2 ± 0.0	467.9 ± 114.5	28.0 ± 4.6	0.2 ± 0.0	0.2 ± 0.0
HAAs	MCAA	2.1	31.6	35.4	1.5	1.5	0.4 ± 0.1	9.4 ± 0.2	5.0 ± 0.2	4.3 ± 0.6	3.9 ± 0.3	0.3 ± 0.0	10.2 ± 1.5	8.0 ± 0.8	0.5 ± 0.1	0.7 ± 0.1
	MBAA	0.3	1.1	1.3	0.4	0.4	0.5 ± 0.0	1.1 ± 0.0	1.2 ± 0.1	0.5 ± 0.0	0.5 ± 0.0	0.5 ± 0.0	ND	1.3 ± 0.1	0.8 ± 0.1	0.4 ± 0.0
	DCAA	21.7	312.0	298.2	22.5	23.0	8.4 ± 0.2	41.6 ± 0.1	22.5 ± 0.4	16.8 ± 0.3	17.5 ± 0.0	6.9 ± 0.0	145.5 ± 1.6	36.3 ± 0.7	9.3 ± 0.0	10.5 ± 0.3
	TCAA	7.2	72.7	112.9	7.0	7.3	2.1 ± 0.1	26.5 ± 0.3	14.9 ± 1.8	3.5 ± 0.2	5.0 ± 0.3	2.0 ± 0.2	33.7 ± 0.1	14.0 ± 1.0	1.8 ± 0.4	4.6 ± 0.3
	BCAA	5.4	15.1	16.8	4.8	4.8	1.3 ± 0.2	6.6 ± 0.1	4.1 ± 0.1	1.9 ± 0.4	2.5 ± 0.0	1.2 ± 0.0	15.7 ± 0.6	9.3 ± 0.4	1.8 ± 0.2	1.6 ± 0.0
	DBAA	1.6	3.0	3.3	1.6	1.7	2.2 ± 0.0	7.7 ± 0.1	7.6 ± 0.8	3.1 ± 0.0	2.7 ± 0.1	2.1 ± 0.0	16.4 ± 0.1	17.9 ± 1.1	3.9 ± 0.4	2.3 ± 0.2
	BDCAA	2.3	6.8	8.4	2.3	2.9	ND	11.8 ± 0.7	8.7 ± 0.1	2.8 ± 0.1	2.8 ± 0.1	ND	8.5 ± 0.9	4.2 ± 0.9	2.2 ± 0.2	31.6 ± 1.0
	CDBAA	0.8	21.4	33.9	2.0	2.0	ND	ND	ND	ND	3.0 ± 0.9	ND	ND	ND	ND	ND
	TBAA	2.0	42.6	62.0	2.5	1.0	ND	ND	ND	ND	ND	ND	ND	33.1 ± 8.2	ND	ND
N-DBPs	DCAN	1.6	1089.2	783.6	1.5	1.4	2.4 ± 0.1	166.5 ± 6.1	5.4 ± 0.1	1.3 ± 0.0	1.4 ± 0.0	2.8 ± 0.0	1069.0 ± 4.2	318.7 ± 7.5	2.2 ± 0.1	1.8 ± 0.3
	TCAN	0.1	3.3	3.2	0.1	0.1	ND	ND	ND	ND	ND	ND	5.8 ± 0.3	ND	ND	ND
	BCAN	0.9	ND	ND	0.7	0.7	0.4 ± 0.0	1.4 ± 0.0	0.8 ± 0.0	ND	0.5 ± 0.0	0.4 ± 0.0	10.5 ± 0.3	3.2 ± 0.1	0.6 ± 0.0	0.5 ± 0.1
	DBAN	0.3	0.3	0.3	0.3	0.3	5.1 ± 0.4	23.4 ± 0.1	13.0 ± 0.9	2.7 ± 0.1	4.3 ± 0.1	4.0 ± 0.1	57.2 ± 1.0	55.5 ± 0.8	6.8 ± 0.1	4.3 ± 1.4
	DCNM	ND	ND	ND	ND	ND	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	ND	ND	0.1 ± 0.0	0.3 ± 0.0	0.2 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
	TCNM	0.5	0.6	0.7	0.3	0.2	ND	0.3 ± 0.0	ND	ND	ND	ND	4.3 ± 0.0	ND	ND	ND
	BNM	0.1	ND	ND	0.1	0.1	ND	0.1 ± 0.0	0.1 ± 0.0	ND	ND	ND	ND	ND	ND	ND
	DCAAm	1.3	295.2	246.2	1.3	1.9	6.8 ± 0.0	48.2 ± 0.5	15.5 ± 0.1	3.7 ± 0.0	4.7 ± 0.0	5.6 ± 0.0	262.2 ± 14.2	45.4 ± 2.1	6.0 ± 0.3	7.2 ± 1.7
	TCAAm	0.6	32.0	28.4	0.6	0.5	2.3 ± 0.0	310.3 ± 14.0	23.8 ± 1.0	1.4 ± 0.1	1.2 ± 0.0	1.8 ± 0.0	5794.4 ± 324.9	5351.8 ± 58.6	0.4 ± 0.0	0.5 ± 0.3
C-DBPs	CH	1.9	8.5	15.4	1.7	2.0	0.3 ± 0.0	2.8 ± 0.1	1.7 ± 0.0	0.5 ± 0.0	0.8 ± 0.0	0.3 ± 0.0	34.0 ± 2.1	5.3 ± 0.1	0.3 ± 0.0	1.3 ± 0.3
	CTC	0.3	ND	ND	0.3	0.1	0.1 ± 0.0	0.2 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	ND	ND	0.1 ± 0.0	ND
	DCPN	4.7	5.2	6.2	6.6	7.2	0.8 ± 0.0	3.3 ± 0.1	3.2 ± 0.1	2.1 ± 0.0	2.3 ± 0.0	0.9 ± 0.0	2.2 ± 0.2	2.5 ± 0.1	0.3 ± 0.0	0.5 ± 0.1
	TCPN	12.7	16.0	25.0	18.2	18.7	1.7 ± 0.1	8.9 ± 0.3	8.4 ± 0.1	9.2 ± 0.1	9.6 ± 0.1	1.6 ± 0.0	2.7 ± 0.1	6.1 ± 0.2	1.2 ± 0.1	1.4 ± 0.3
	TetraCPN	29.1	40.0	76.7	32.4	35.6	2.7 ± 0.1	34.1 ± 0.6	23.8 ± 0.6	30.6 ± 1.3	31.8 ± 0.3	1.1 ± 0.0	14.6 ± 0.7	12.4 ± 0.1	1.0 ± 0.0	0.6 ± 0.2
	PentaCPN	45.4	156.8	249.2	54.0	63.9	ND	14.2 ± 2.7	0.3 ± 0.4	ND	0.3	ND	143.0 ± 19.2	8.8 ± 0.2	ND	ND
	DBCPN	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TCE	ND	8.8	12.9	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TriCEL	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TCEL	ND	ND	ND	0.2	0.2	ND	ND	ND	0.1 ± 0.0	ND	0.1 ± 0.0	5.4 ± 1.3	0.8 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
	DBE	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND = not detectable; n = 2 for values with standard deviation.

Table 3. Concentrations of DBPs (ng/g) detected in shredded lettuce and diced cabbage from commercial processing.

Group	Target DBPs	Shredded lettuce												Diced cabbage					
		First sampling event						Second sampling event						Second sampling event					
		Unwashed	1^{st}-Flume washed	2^{nd}-Flume washed	Final rinsed	Incline-washed	Vibra-washed	Unwashed	1^{st}-Flume washed	2^{nd}-Flume washed	Final rinsed	Incline-washed	Vibra-washed	Unwashed	1^{st}-Flume washed	2^{nd}-Flume washed	Final rinsed	Incline-washed	Vibra-washed
THMs	CF	537.2	100.6	171.9	244.8	371.9	363.4	ND	3.1 ± 2.1	0.8 ± 1.0	1.0 ± 1.2	1.5 ± 2.0	0.2 ± 0.0	0.5	0.1 ± 0.1	ND	0.1 ± 0.1	0.2	1.8 ± 0.4
	BF	ND	ND	ND	ND	ND	ND	ND	0.1 ± 0.0	0.1	0.04	ND	ND	ND	ND	ND	ND	ND	ND
	BDCM	ND	ND	0.2	ND	ND	ND	ND	0.9 ± 0.0	0.8 ± 0.0	0.8 ± 0.0	ND	0.9 ± 0.0	ND	ND	ND	ND	ND	ND
	DBCM	ND	ND	0.03	ND	ND	ND	ND	0.7 ± 0.0	0.7 ± 0.0	0.7 ± 0.0	ND	0.7 ± 0.0	ND	0.7 ± 0.0	0.6 ± 0.1	0.6 ± 0.0	ND	ND
HAAs	MCAA	3.1	12.9	15.4	11.5	ND	5.2	ND	ND	11.5 ± 0.1	11.8 ± 2.5	ND	8.0 ± 0.8	9.6 ± 2.8	19.2 ± 1.8	12.8 ± 1.1	11.9 ± 0.0	9.5 ± 1.1	10.1 ± 1.5
	MBAA	0.4	ND	0.5	0.05	0.3	0.2	ND	ND	0.2	0.1	ND	0.1 ± 0.1	ND	ND	ND	ND	ND	ND
	DCAA	5.3	211.3	343.3	154.4	8.1	129.2	2.2 ± 0.8	59.1 ± 0.8	58.2 ± 0.5	23.3 ± 4.4	1.2 ± 0.3	54.7 ± 1.3	0.7 ± 0.0	181.1 ± 1.8	63.6 ± 1.4	71.0 ± 4.8	4.5 ± 0.7	5.8 ± 0.3
	TCAA	ND	14.5	20.5	13.8	ND	4.6	ND	ND	5.0 ± 0.5	ND	ND	ND	ND	15.6 ± 3.4	7.4 ± 0.0	8.7 ± 2.7	2.5 ± 1.9	2.7 ± 0.9
	BCAA	ND	2.2	5.1	2.2	ND	1.5	ND	ND	ND	ND	ND	ND	ND	ND	8.2 ± 0.2	8.1 ± 1.6	5.8 ± 1.2	4.1 ± 0.4
	DBAA	ND	ND	0.5	ND	ND	ND	ND	1.7 ± 0.2	2.8 ± 1.1	ND	ND	1.5 ± 0.1	ND	ND	ND	ND	ND	ND
	BDCAA	0.3	1.4	2.7	1.4	1.0	1.2	ND	18.9 ± 10.5	36.7 ± 2.4	25.9 ± 4.3	28.6 ± 6.0	24.2 ± 3.5	NA	NA	NA	NA	NA	NA
	CDBAA	15.8	0.8	1.0	1.9	21.9	1.3	ND	30.9 ± 2.1	41.8 ± 2.9	42.9 ± 16.0	37.2 ± 14.2	33.0 ± 2.7	ND	ND	ND	ND	ND	7.3 ± 0.4
	TBAA	ND	3.8	2.7	4.2	ND	5.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
N-DBPs	DCAN	ND	23.2	100.4	23.6	0.08	0.2	ND	1.7 ± 0.2	1.2 ± 0.1	1.1 ± 0.0	0.9 ± 0.0	1.0 ± 0.0	ND	1.2 ± 0.0	0.9 ± 0.1	0.8 ± 0.0	ND	ND
	TCAN	ND	0.2	0.8	0.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	BCAN	1.7	9.4	7.8	6.4	1.9	4.3	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	DBAN	0.6	ND	0.6	0.8	0.6	0.2	ND	ND	0.2 ± 0.1	0.2 ± 0.2	0.2 ± 0.2	0.1 ± 0.0	ND	ND	ND	ND	ND	ND
	DCNM	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TCNM	ND	5.8	5.0	4.6	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	BNM	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	DCAAm	39.1	379.4	612.9	321.9	45.4	381.7	ND	282.2 ± 50.3	134.9 ± 44.7	67.6 ± 11.7	ND	125.8 ± 18.7	6.3 ± 1.2	92.1 ± 3.2	58.3 ± 22.7	49.1 ± 14.8	3.6 ± 0.8	6.1 ± 0.4
	TCAAm	ND	166.6	521.6	174.5	ND	ND	ND	ND	ND	ND	ND	0.1 ± 0.0	ND	2.8 ± 0.2	1.2 ± 0.5	0.9 ± 0.5	ND	ND
C-DBPs	CH	ND	6.6	13.9	6.7	0.7	1.3	ND	1.7 ± 0.3	1.1 ± 0.1	1.0 ± 0.0	ND	0.8 ± 0.0		2.8 ± 0.1	1.5 ± 0.4	1.2 ± 0.1	ND	0.6 ± 0.0
	CTC	ND	ND	ND	ND	ND	ND	1.2 ± 0.0	1.2 ± 0.0	1.2 ± 0.0	1.2 ± 0.0	1.2 ± 0.0	1.2 ± 0.0	1.7 ± 0.2	1.5 ± 0.0	1.5 ± 0.0	1.5 ± 0.0	1.5 ± 0.0	1.5 ± 0.0
	DCPN	0.9	1.9	3.5	2.0	0.6	1.2	1.8 ± 0.2	2.4 ± 0.2	1.9 ± 0.1	2.1 ± 0.3	2.0 ± 0.2	1.8 ± 0.1	1.8 ± 0.3	1.6 ± 0.0	1.8 ± 0.0	1.8 ± 0.2	1.5 ± 0.1	1.7 ± 0.0
	TCPN	ND	4.6	9.3	7.3	0.2	2.4	ND	1.2 ± 0.0	0.8 ± 0.2	1.0 ± 0.1	ND	1.6 ± 0.1	ND	0.8 ± 0.0	0.5 ± 0.1	ND	ND	0.4 ± 0.0
	TetraCPN	1.7	34.8	84.6	36.2	2.0	13.1	ND	18.0 ± 1.0	10.1 ± 3.1	8.3 ± 3.2	ND	7.7 ± 0.8	ND	ND	6.2 ± 3.2	5.1 ± 0.8	3.4 ± 0.6	4.1 ± 1.6
	PentaCPN	ND	17.0	59.2	26.1	ND	8.1	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	DBCPN	ND	ND	ND	ND	0.2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TCE	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TriCEL	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	TCEL	ND	ND	ND	ND	ND	ND	ND	ND	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	0.3 ± 0.0	28.2 ± 0.4	17.6 ± 0.1	21.1 ± 2.6	19.2 ± 2.0	17.3 ± 0.8	17.7 ± 2.0
	DBE	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

ND = not detectable; NA = not available (due to analytical interference); n = 2 for values with standard deviation.