Lead in Maple Syrup Produced in Connecticut

David E. Stilwell* and Craig L. Musante

Department of Analytical Chemistry, The Connecticut Agricultural Experiment Station, P.O. Box 1106, New Haven, Connecticut 06504

The lead content of 44 maple syrup samples from 23 Connecticut producers was determined over a period of two seasons. The results ranged from 38 to 948 μg/kg and averaged 291 μg/kg. In addition, sap and syrup-processing samples were taken from two producers. The lead content (μg/L) in all maple grove site sap samples ($n = 48$) ranged from <0.5 to 19 and averaged 2.6, while those taken exclusively from plastic grove site tanks ($n = 35$) ranged from <0.5 to 5.4 and averaged 1.1. The higher values were associated with samples taken from galvanized containers. Further contamination of the sap, shown to be caused by the use of a bronze gear pump employed in sap transfer steps, resulted in an average lead content of 19 μg/L in sap samples ($n = 7$) from the final stainless steel storage tanks. These data indicate that contact of any lead-bearing metal by the acidic sap should be avoided. Lead and phosphorus analyses of the syrup-processing samples showed that during the processing of the sap into syrup, significant amounts of lead precipitate out of solution in a form consistent with lead phosphate. Due to the formation of this precipitate, the importance of distinguishing between mixed phase and solution phase samples is emphasized. Finally, on a dry weight basis, the final syrup was no higher in lead content than the incoming sap.

Keywords: *Maple syrup; syrup; sap; lead*

INTRODUCTION

Increasing awareness of the toxicity and widespread environmental distribution of lead (Biddle, 1982; WHO, 1989) has prompted many investigations into its levels in the general food supply (Jelinek, 1982; Wolnik *et al.*, 1985; Chen and Gao, 1993; Dabeka and McKenzie, 1995), including syrups (Robinson *et al.*, 1989; Dabeka and McKenzie, 1995). In maple syrup produced in Canada, Robinson *et al.* (1989) found higher average lead contents compared to the average amounts in other dietary components. Subsequently, many of the maple syrup-producing states in the United States initiated surveys of the lead content in maple syrup. In this report, we present our findings regarding the lead content in maple syrup produced in Connecticut over a 2 year period (1994−1995). In addition, we identified potential sources of lead in the syrup based on our analysis of sap and syrup-processing samples.

MATERIALS AND METHODS

Collection and Analysis of Maple Syrup Samples. Forty-four grade A maple syrup samples were obtained from a total of 23 Connecticut producers over two seasons (1994−1995). Syrup samples from 12 of the 23 producers were obtained during each of the two seasons, while samples from the remaining 13 were obtained only once. Replicate samples were received from two producers in 1994 and three producers in 1995. The samples came from retail stock, in producer-supplied containers (typically plastic), ranging in size from 84 to 475 mL. In Connecticut there are about 120 syrup producers who sell retail. In order to compare the lead concentrations in maple syrup to other syrups, 8 commercially available table (corn) syrups and 10 molasses products were purchased. Statistical analysis by matched pair *t*-test (Ott, 1984) was carried out on the results from the 12 producers sampled in both the 1994 and 1995 seasons.

The 1994 syrup samples were prepared for analysis by microwave digestion (MDS 81D, CEM Corp., Matthews, NC). Two replicates, 1−1.2 g each, were weighed (±0.1 mg) into digestion vessels; 7 mL of concentrated nitric acid was added to each vessel, and the samples were allowed to react at room temperature for 2 h. Prior to sealing the vessels, 3 mL of distilled deionized (DDI) water was added. The oven program was 20% power for 5 min followed by 30% power for 10 min and 35% for 20 min. The digests were transferred to 25 mL volumetric flasks, brought up to volume with DDI water, and stored in 125 mL poly(ethylene) containers until analysis. This relatively mild digestion procedure was used, as the syrup was found to be quite reactive. By using these steps, pressure overruns in the sealed microwave vessels were avoided.

For control purposes the lead content in procedure blanks and spiked samples was determined. The lead concentrations in the procedure blanks ($n = 4$) were below the solution detection limit of 0.5 μg/L. The average spike recoveries (10 μg/L added lead) were 102 ± 9% (samples, $n = 6$) and 91 ± 4% (spiked blanks, $n = 4$). The average percent relative standard deviation (%RSD) of the lead content between the replicate syrup samples was 2.4%.

To facilitate the analysis, the 1995 syrup and syrup-processing samples were digested by an alternative method first described by Miller-Ihli (1994) and employed here with only minor modifications. Briefly, 0.5 g (±0.1 mg) syrup samples were weighed into 10 mL volumetric flasks, 1 mL of concentrated nitric acid was added, and the mixture was heated in a water bath (50 ± 10 °C) for 30 min. After cooling, 0.5 mL of 30% H_2O_2 was added, and the samples were returned to the water bath for 15 min. The peroxide step was then repeated prior to bringing the samples to volume. The average %RSD of the lead content between the replicate syrup samples was 2.5%. For quality control purposes, one or two procedure blanks and either spikes (10 μg/L added lead) or a control syrup were run with each batch. The lead in the procedure blanks ($n = 20$) was below the solution detection limit of 0.5 μg/L. The average recovery for the spiked procedure blanks was 101 ± 3% ($n = 12$), and for the spiked samples, it was 103 ± 10% ($n = 8$). The average amount of Pb in the control syrup was 265 ± 13 μg/kg ($n = 13$), compared to an average of 251 ± 54 μg/kg obtained in a round robin study with 13 participating laboratories (organized by N. Shambaugh, Vermont Department of Agriculture, Waterbury, VT 05671).

The lead content (μg/kg) of one of the syrups was compared using the two methods described above, and the results were within 5% of each other (microwave digestion, 288 and 286; water bath digestion, 300 and 303).

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Collection and Analysis of Maple Sap Samples. In an effort to determine the potential sources of lead in the final maple syrup product, maple sap samples were taken representative of the various stages of sap collection, storage, and processing. Maple sap only flows in harvestable amounts when, in late winter to early spring, below freezing nighttime temperatures are followed by above freezing daytime temperatures. When such limited conditions prevailed, maple sap samples were taken from two producers on a total of seven occasions.

The first samples in the sequence, denoted as grove sap samples, were taken from enclosed containers located at the maple groves. At the grove, the sap from the trees flows into these containers via a network of taps and plastic tubing. Producer 1 employs a combination of plastic and galvanized (Zn-coated) grove tanks. In addition, 5−10% of the sap is collected directly from tree taps into covered galvanized buckets. On five occasions (February 22, March 6, 7, 8, and 13), one sample was collected from each of the six to seven grove tanks, along with composite samples from the buckets, for a total of 39 samples. Producer 2 uses only plastic tanks, from which, on two occasions (March 6 and 7), one sample was collected from tanks in each of the six groves.

The next processing step is that the sap in the grove tanks is pumped into large plastic transport tanks situated on a truck bed. Producer 1 uses a bronze gear pump, whereas producer 2 employs an aluminum centrifugal pump. Sap samples from these truck tanks, denoted as truck tank samples, were taken in duplicate on three occasions (March 6, 7, and 13) from producer 1 and on one occasion (March 7) from producer 2. The sap in the truck tank is then clarified and filtered by recirculation through a diatomaceous earth (DE) filter, with both producers using aluminum centrifugal pumps. Sap samples taken after this step, denoted as DE-filtered sap, were taken in duplicate on two occasions (March 6 and 13) from producer 1 and on one occasion (March 6) from producer 2. Finally, the sap is pumped through an ultraviolet treatment unit and into a stainless steel (SS) holding tank. Sap samples taken at this point were denoted as SS-tank samples. Producer 1 employs an aluminum centrifugal pump, while producer 2 uses a bronze gear pump. Sap samples were taken in duplicate after this step on five occasions (February 22, March 6, 7, 8, and 13) from producer 1 and on two occasions (March 6 and 7) from producer 2. Logistical problems prevented us from acquiring samples reflecting all intermediate steps for the five producer 1 sampling events and the two producer 2 sampling events during which beginning samples (grove sap) and final samples (SS-tank) were taken.

The sap and syrup-processing samples were collected in 125 mL poly(ethylene) containers. The containers were washed with 2% nitric acid, rinsed three times with DDI water, and then dried. Field blanks, obtained by adding DDI water to containers at the site, were all below the detection limit of <0.5 μg/L for lead.

The sap samples were prepared for lead analysis by adding 1.8 mL of sap and 0.2 mL of concentrated nitric acid directly into the 2.5 mL poly(ethylene) autosampler cups. The average spike recovery (5 μg/L added lead) was 99 ± 4% (n = 6), and the detection limit was 0.5 μg/L (1 pg) lead. The sap samples were prepared for copper, phosphorus, and zinc analysis by acidifying with 1 part concentrated nitric acid to 9 parts of sap.

Collection and Analysis of Maple Syrup-Processing Samples. Sap is processed into maple syrup by vigorous boiling in a series of metal pans, as described elsewhere (Nearing and Nearing, 1970; USDA, 1982). Many of the evaporators and finishing pans currently in use were fabricated using lead solder, and thus, these processing steps have the potential to add lead to the syrup. Moreover, lead in the sap could be concentrated into the syrup, as the concentration factor for sap to syrup is around 40:1 (USDA, 1982).

Process batch samples were collected on two occasions during the 1995 season (March 6 and 13). The first samples in the sequence were taken from the incoming sap stored in the stainless steel holding tank. The next samples were taken after the sap had been gravity fed through a copper tubing preheater, at the point where a float valve is used to regulate the sap flow into the stainless steel evaporator (denoted as post-preheater samples, ppreht). Samples from the evaporator and the triple-partition stainless steel finishing pan are denoted as evaporator (evap) and pan 1A−C. At point pan 1C, the syrup is tested for specific gravity, and after the appropriate value is obtained, a quantity is withdrawn (termed a draw). Prior to bottling in plastic containers, the syrup is filtered through a rayon cellulose filter. These samples are denoted final and filtered final (ffinal).

In order to obtain representative, steady-state conditions, the process batch samples were taken midway in the runs. The initial amounts of sap processed were 850 L (batch 3-6-95) and 1550 L (batch 3-13-95). Duplicate batch samples (50−100 mL total for each sample), taken 0.5 h apart using disposable 5 mL poly(ethylene) plastic pipets, were placed into 125 mL poly(ethylene) containers.

During processing, a precipitate invariably forms during the evaporation step (Winton and Winton, 1939; Nearing and Nearing, 1970). This solid matter, commonly referred to as sugar sand, is dragged along by the solution phase until it is filtered out at the final step. To distinguish between analytes in the solution and solid phases, the following protocol was developed. First, the process batch sample was vigorously shaken, and two replicates were weighed out for digestion. These samples, containing both the solid and solution phases, were denoted as "mixed phase." Separation of the phases was accomplished by centrifuging (4000 RPM, 10 min). Two replicates of the solution phase were withdrawn and weighed for digestion. These samples were denoted as "solution phase." All of the process batch samples were prepared for lead and phosphorus analysis by acid digestion in the water bath, as described above. To obtain sufficient volume for the determination of phosphorus, the replicate digests were combined after they were analyzed for lead.

To express the results on a dry weight basis, 5 mL portions of the phase samples were dried in an oven (100 °C) overnight followed by drying in a vacuum oven (60 °C) for 1 h. As a percentage of their original mass, these values ranged from 2−2.7% (incoming sap) to 65−70% (final syrup). Scrapings of the solder used in the fabrication of the preheater, the evaporator, and the pans were taken and prepared for lead analysis by dissolution in aqua regia.

Instrumentation and Reagents. The lead (Pb) content was determined using a PE 5100PC graphite furnace atomic absorption spectrophotometer (GFAAS) (Perkin Elmer Corp., Norwalk, CT) following the manufacturer's guidelines regarding modifiers ($NH_4H_2PO_4$, $Mg(NO_3)_2$), sample aliquots (20 μL), furnace time, and temperature profiles. An additional air-ashing step was employed to reduce the amount of organic residues (Maeda *et al.*, 1989; Stilwell and Musante, 1994). The copper (Cu), phosphorus (P), and zinc (Zn) content of the sap and digested syrup samples, as well as the lead content in solder scrapings, were determined using an Atom Scan 16 inductively coupled plasma atomic emission spectrometer (ICP) (Thermo Jarrell Ash, Franklin, MA). Check standards, either AA-1 or QC-19 (Spex, Edison, NJ), were run after every five determinations, and the agreement was within 10% of the certified value.

All chemicals used were reagent grade for trace metal analysis. All aqueous solutions were prepared using DDI water. All of the plastic containers and pipets were metal free grade for trace analysis.

RESULTS AND DISCUSSION

Lead Concentrations in the Maple Syrup. Table 1 summarizes our findings for the lead content in maple syrup. The number of samples exceeded the number of producers since replicate samples were occasionally taken (two producers in 1994, three producers in 1995), and, in those cases, the results were averaged. The range and average lead content in the syrups were higher in the 1995 samples. Five of the samples in 1995

Table 1. Lead Content (μg/kg) in Maple Syrup

	year		
	1994	1995	overall
range	46−469	38−948	38−948
average	199	378	291
no. of samples[a]	23	21	44
no. of producers[b]	17	18	23
producers with samples in the range:			
<20	0	0	
20−100	4	4	
101−200	7	2	
201−350	3	3	
351−500	3	4	
>500	0	5	

[a] An average result was used when multiple samples were obtained from the same producer. [b] Twelve producers were represented in both years.

Table 2. Lead Content (μg/kg) in Other Syrup Products

product	samples	brands	range	average
corn syrup	8	8	<20−32	<20
molasses	7	7	<20−255	68
blackstrap molasses	3	2	106−490	238

(14%) exceeded the 500 μg/kg lead advisory limit recently established by the State of Vermont. Statistical analysis by matched pair *t*-test (Ott, 1984) was carried out on the results from 12 of the producers sampled in both the 1994 and 1995 seasons. The average lead level in these samples was significantly higher ($P < 0.05$) in the 1995 samples (351 μg/kg) than in the 1994 samples (193 μg/kg). Explanations for this increase are not apparent. Nonetheless, the overall average of 291 μg/kg lead reported here is lower than that obtained from 27 producers in Canada (Robinson *et al.*, 1989). In that study, an average and range for lead (μg/kg) in maple syrup of 490 and 310−860, respectively, were reported in Nova Scotia, 610 and 290−2010 in New Brunswick, and 360 and 250−650 in Quebec. The data in Robinson *et al.* (1989) were converted from m/v (μg/mL) to m/m (μg/kg) using 1.33 g/mL (Winton and Winton, 1939) for the density of maple syrup.

The results for the lead levels in commercially available table (corn) syrups and molasses products are given in Table 2. The lead content in only one of the 8 corn syrup samples was above the detection limit, while lead was detected in 6 of the 10 molasses samples. In this limited survey, the lead content of the blackstrap molasses was similar to that found in the maple syrups. Only minor amounts of lead in table syrup were also reported by Dabeka and McKenzie (1995). In a sample size of five, they found the lead level (μg/kg) ranged from 1.8 to 14.9 and averaged 5.3.

Lead in Maple Sap Samples. A summary of the results obtained from producer 1 on five sampling occasions is shown in Figure 1. The lead values for the grove samples represent an average of all samples (n = 6−10) taken at the various grove sites at a given date. The lead level of all grove site samples (n = 39) taken from producer 1 averaged 3.3 μg/L. The results show a marked, and reproducible, increase in the lead content in the sap after the point where the bronze gear pump was used to transfer the sap into the truck tank. Subsequently, the lead concentration in the sap decreased somewhat after passing through the DE filter and then remained constant after transfer into the SS-holding tank. For example, the average lead content (μg/L) of saps collected for batch 3-6-95 was 4.3 (groves), 31 (truck tank), 19 (DE filtered), and 20 (SS-tank) and for batch 3-13-95 was 4.2 (groves), 14 (truck tank), 13 (DE filtered), and 13 (SS-tank).

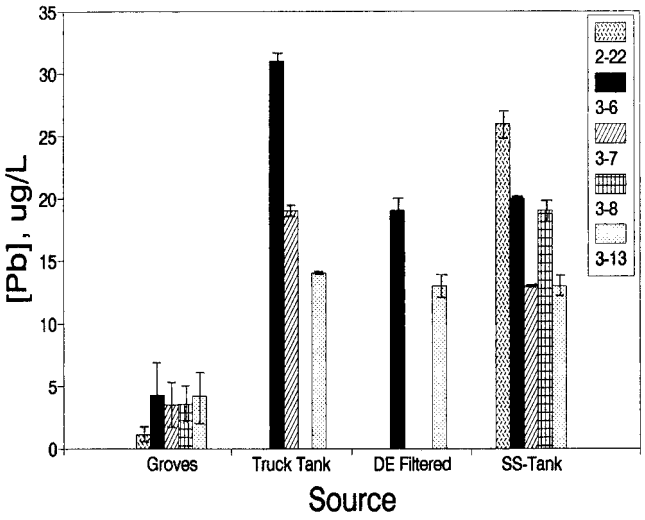

Figure 1. Lead concentration in sap samples from producer 1. The sampling date (1995) is given in the legend.

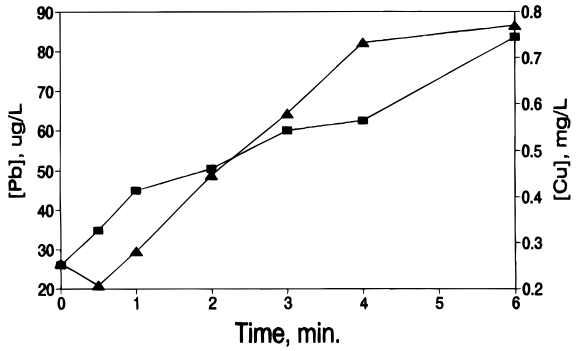

Figure 2. Lead and copper concentrations in sap samples versus time. Samples were taken from sap that was recirculated by a bronze gear pump (■, Pb; ▲, Cu).

For producer 2, the average lead content (μg/L) of sap samples taken on the two occasions was 2.4 (groves), 0.9 (DE filtered), and 28 (SS-tank) for sample set 3-6-95 and 1.7 (groves), 2.0 (truck tank), and 14 (SS-tank) for sample set 3-7-95. The lead level of all grove site samples (n = 12) taken from producer 2 averaged 2.0 μg/L. Note that the lead content of the sap from producer 2 also increased significantly after it was passed through a bronze gear pump; however, for producer 2, this type of pump was used to transfer the sap into the SS-tank.

Since both producers employed a bronze gear pump (5% Pb in the bronze) to transfer the sap just prior to the process step for which the lead content of the sap samples markedly increased, leaching of lead from the bronze seemed to be a likely explanation. To test this hypothesis, approximately 20 L of sap was placed into a plastic container, recirculated by the pump, and periodically sampled (100 mL) for 6 min. Both the copper and lead contents increased by more than 300% over their original values within 6 min (Figure 2). Thus, if the gear pump was responsible for leaching lead into the sap, increases in copper should also be observed. The results for the copper content in sap samples from producer 1, shown in Figure 3, confirm the increase in copper at the same processing stage where the lead was observed to increase (Figure 1). For example, the average copper level of the sap samples from the grove

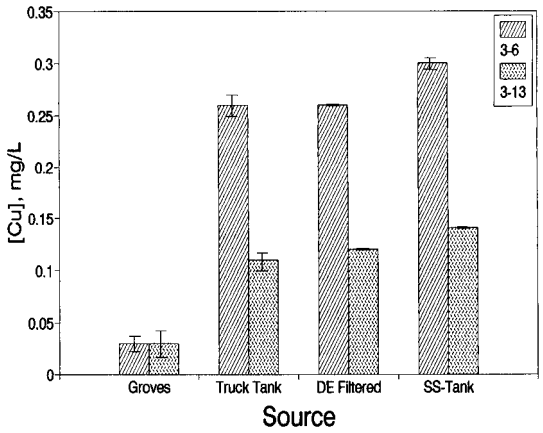

Figure 3. Copper concentration in sap samples from producer 1. The sampling date (1995) is given in the legend.

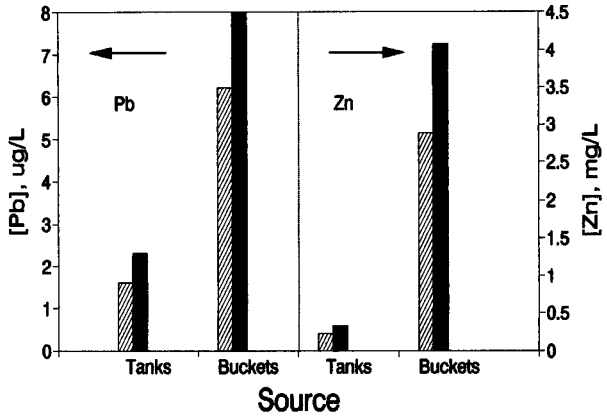

Figure 4. Comparison of lead and zinc concentrations in sap samples from plastic tanks and galvanized buckets (hashed bars, grove CR1; solid bars, grove S1).

tanks compared to the truck tanks increased from an average of 0.03 to 0.26 mg/L in sample set 3-6-95 and from 0.03 to 0.12 mg/L in sample set 3-13-95. Similarly, for producer 2, the copper content increased from 0.04 to 0.13 mg/kg in sample set 3-6-95 after the stage (described above) where the sap passed through the gear pump. These results demonstrate that the bronze gear pump can contaminate the sap with lead and copper.

Another potential source of lead contamination was the galvanized buckets and tanks used to collect the sap in the groves. A comparison of the lead and zinc concentrations in sap samples from two plastic grove tanks to those of composite sap samples taken from five metal buckets located in the same grove is shown in Figure 4. Both lead and zinc levels in the sap from the bucket samples were much higher than those from the tanks. To a lesser extent, there tended to be higher levels of lead in sap collected in galvanized tanks than in plastic tanks. For example, the average lead level in sap from a galvanized tank was 2.4 ± 0.3 μg/kg ($n = 5$) compared to <0.5 μg/kg ($n = 5$) in samples collected from a plastic tank at a nearby location.

In summary, sap from maple trees contained very little lead, as reflected by the values found in the grove site collection vessels. The lead content in all sap samples collected from plastic grove site tanks ($n = 35$) ranged from <0.5 to 5.4 μg/L and averaged 1.1 μg/L, while the lead level in all sap samples collected in the groves ($n = 48$) ranged from <0.5 to 19 μg/kg and averaged 2.6 μg/kg. The higher values were associated with samples taken from metal containers. Further contamination of the sap occurred after use of a bronze

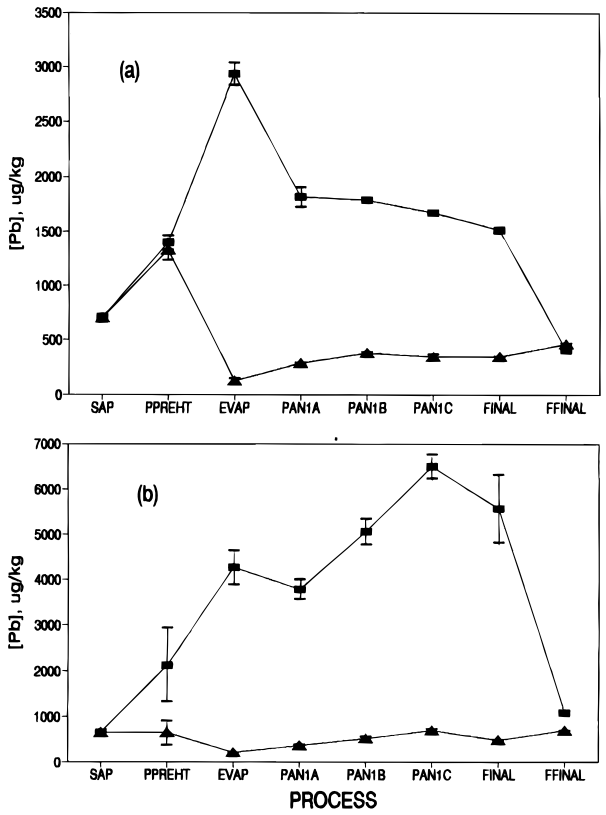

Figure 5. Lead concentration, on a dry weight basis, in process batch samples from producer 1: (a) process batch series 3-6-95 and (b) series 3-13-95 (■, mixed phase; ▲, solution phase).

gear pump, resulting in an average lead concentration of 19 μg/L ($n = 7$) in sap samples taken from the SS-holding tanks. The lead content (μg/L) of sap samples from the final stainless steel holding tanks reported here (13−28) is similar to data reported by Robinson *et al.* (1989) for sap samples collected from SS-holding tanks in Nova Scotia (3−18) and Quebec (4−42) but much lower than those for New Brunswick (3−2090). Since maple sap is acidic (pH 3.4−6.6; Robinson *et al.*, 1989), and in the presence of oxygen (Stilwell and Musante, 1994) has the potential to react with many metal surfaces, contact with any metal containing lead should be avoided when sap is collected and processed.

Lead in Maple Syrup-Processing Samples. The lead contents in the syrup-processing samples, expressed on a dry weight basis, are shown in Figure 5. The standard deviations about the average for each duplicate sample set are graphically represented in the figures as error bars. In relative terms (%RSD), they ranged from 0.01% to 40% and averaged 5.5%.

Analyses indicate that much of the lead precipitates out of solution during processing, as shown by the higher lead levels in the mixed phase samples up until the final product is filtered. For example, the average lead concentration in the evaporator samples (3-13-95 series) averaged 4270 μg/kg in the mixed phase samples and only 221 in the solution phase samples. Also, a sample of the filter residue (3-13-95 series) collected prior to bottling the final syrup contained 33 000 μg/kg lead, while the final syrup contained 575 μg/kg.

The solution phase data show that the lead content reached a minimum at the evaporator stage and then slowly increased. The reasons for the gradual increase are not known, but decreasing pH may be one factor (shown below). Another factor could be leaching of lead

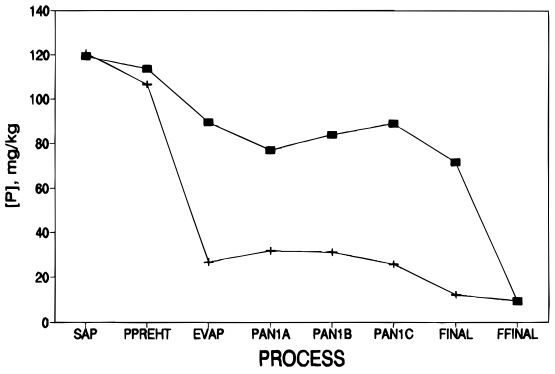

Figure 6. Phosphorus concentration in process batch series 3-13-95 (■, mixed phase; +, solution phase).

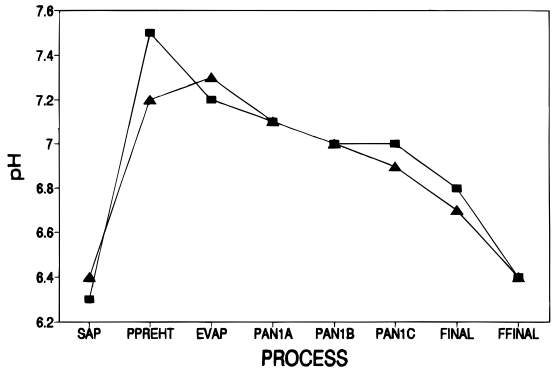

Figure 7. pH in duplicate process batch samples (series 3-13-95), after dilution to 2.3% solids (■, sample 1; ▲, sample 2).

from the solder used in fabricating the evaporator and pans (the solder in the preheater was lead free, <0.5% Pb, while the lead content of the solder used in the evaporator and pans ranged from 50% to 60%). However, with only one exception, the solution phase lead concentration is higher in the incoming sap than the process batch samples, including the final filtered product. Due to this, any extent of lead leached from the solder remains uncertain.

The amounts of lead with process stage differ in the mixed phase results between the two sample sets, while in the solution phase samples sets they were similar. During processing, the buildup of lead laden sugar sand is likely to account for the observed increase in mixed phase lead from the second sample set.

The precipitation of lead from solution could have been due to the formation of lead phosphate. Lead phosphate is highly insoluble, with a reported solubility product constant (K_{sp}) of 1.0×10^{-54} (Wallace, 1984). To test this hypothesis, we determined the phosphorus content for process batch samples 3-13-95, before and after centrifugation. The results in Figure 6 show that phosphorus compounds precipitate from solution. Commencing with the evaporator stage, the phosphorus content was much lower in the solution phase samples than in the mixed phase samples. For corroboration, the filter residues from this batch were tested for lead and phosphorus and found to be 33 mg/kg (Pb) and 732 mg/kg (P). To form lead phosphate (taken as $Pb_3(PO_4)_2$) requires a Pb/P ratio by weight of 10. Thus, there is ample excess of phosphorus, as only 3.3 mg/kg P is required to form the lead phosphate.

The pH of the process batch series 3-13-95 was measured, after dilution to equal the 2.3% solids in the sap, and is shown in Figure 7. The increased pH, compared to the incoming sap, could be one explanation for the formation of the precipitate. Increased pH is well known to decrease the solubility of the salts of acids, including lead phosphate (Kolthoff, 1969). Moreover, the gradual decrease in pH observed in the latter processing steps would be expected to increase the lead phosphate solubility and could be one explanation for the gradual increase in the solution phase lead content shown earlier (Figure 5).

Conclusions. The lead content of 44 maple syrup samples from 23 Connecticut producers ranged from 38 to 948 μg/kg and averaged 291 μg/kg. The lead levels in sap samples ($n = 35$) from plastic grove site tanks ranged from <0.5 to 5.4 μg/L and averaged 1.1 μg/L, while the lead content in all grove site sap samples ($n = 48$) ranged from <0.5 to 19 μg/L and averaged 2.6 μg/L. The higher values were associated with samples taken from galvanized containers. Further contamination of the sap, shown to be caused by the use of a bronze gear pump employed in sap transfer steps, resulted in an average lead concentration of 19 μg/L in sap samples ($n = 7$) from the final SS-storage tanks. Contact of any lead-bearing metal with the acidic sap should be avoided.

In addition, the lead level of syrup-processing samples was determined. It was found that significant amounts of lead precipitate out of solution in a form consistent with lead phosphate. Due to the formation of the precipitate, the importance of distinguishing between mixed phase and solution phase samples was demonstrated. Finally, on a dry weight basis, the final syrup was no higher in lead content than the incoming sap.

ABBREVIATIONS USED

Sap Samples. Grove, sap in enclosed containers located at the maple groves; truck tank, sap in plastic transport tanks on a truck bed; DE filtered, sap after recirculation through a diatomaceous earth filter; SS-tank, sap in stainless steel tanks.

Syrup Processing Samples. ppreht, in-process syrup after passing through a preheater; evap, in-process syrup in the evaporator; pan, in-process syrup in pans A−C; final, final unfiltered syrup; ffinal, final filtered syrup; mixed phase, process samples containing both solution and solid phases; solution phase, process samples containg only the solution phase.

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