

How much arsenic is released when CCA treated wood is burned?

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Abstract

Waterborne salts have been used to preserve wood for many years. One of the more common formulations contains copper, chromium, and arsenic salts and is known as chromated copper arsenate, or CCA. Questions have been raised about the amount of arsenic released when CCA treated wood is burned in wood stoves, fireplaces, or boilers. Distribution of the ash which contains residual chemicals also presents a problem. This study was undertaken to clarify the amount of total arsenic released to the atmosphere when CCA treated wood is burned under various combustion conditions and to determine from selected samples the nature of the arsenical chemicals released to the atmosphere. Through the use of a horizontal tube furnace laboratory procedure, we found the amount of arsenic released ranges between 22 and 77 percent, with the higher values occurring when the fuel ash is exposed to high temperatures for long periods. Arsenic that is released is found primarily in condensed (particulate) emissions and consists of arsenates and arsenites. Percentages of copper and chromium volatilized were found to be 11 and 15 percent, respectively.

Waterborne salts have been used to preserve wood from insect and fungal damage for many years. In 1978, approximately 73 million cubic feet of wood was treated with inorganic arsenicals (5). One of the more common formulations contains copper, chromium, and arsenic salts and is known as chromated copper arsenate or CCA. A series of reactions occurs in the wood following treatment with CCA. This results in the formation of water insoluble salts within the wood structure. Substantial amounts of CCA remains in the wood for many years, and the disposal of scrap CCA treated wood by burning could be a serious problem.

In July 1984, the Environmental Protection

Agency issued new restrictions on the handling and use of wood preservatives. For disposal, they stated...

"Dispose of treated wood by ordinary trash collection or burial. Treated wood should not be burned in open fires or in stoves or fireplaces because toxic chemicals may be produced as part of the smoke and ashes. Large quantities of treated wood from commercial or industrial use (e.g., construction sites) may be burned in commercial or industrial incinerators in accordance with state and Federal regulations." (5)

There have been a few reports in the literature concerning burning CCA treated wood. Amounts of arsenic volatilized have been reported to vary between 8 and 95 percent (3, 4, 11, 12). The reasons for this variability and the nature of the arsenic species released have not been established.

This study was undertaken to meet two objectives: 1) to clarify the amount of total arsenic released to the atmosphere when CCA treated wood is burned under various combustion conditions, and 2) to determine from selected samples the nature of the arsenical chemicals released to the atmosphere.

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Forest Prod. J. 36(11/12):45-50.

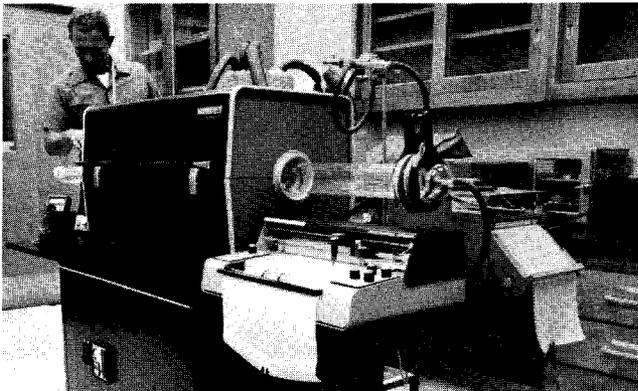


Figure 1. — Overall view of horizontal combustion tube furnace and associated equipment.

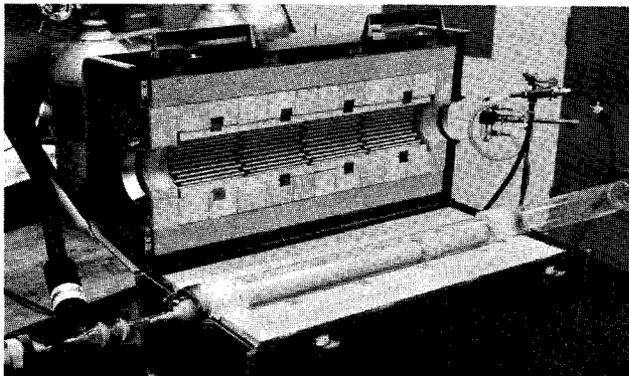


Figure 2. — Close-up view of quartz chamber and combustion tube in furnace. Air flow moves from right to left into glass transfer tube and filter sampling systems.

Materials and methods

CCA treated wood

A sample of CCA treated wood was obtained from the Forest Products Laboratory in Madison, Wis. A portion of a southern yellow pine 2 by 4 treated with 0.60 lb./ft.³ of Type III CCA was used as the starting material. According to Federal specifications, Type III is a CCA formulation containing 47 ± 3 parts by weight of hexavalent chromium as CrO_3 , 19 ± 2 parts by weight of divalent copper as CuO , and 34 ± 4 parts by weight of pentavalent arsenic as As_2O_5 (9). Wood is treated with CCA in the range 0.25 to 2.5 lb./ft.³ depending on end use. Most lumber is treated to 0.25 or 0.4 lb./ft.³. The recommended retention for foundation grade lumber and poles is 0.6 lb./ft.³ and this was the level chosen for this study. In earlier work, Dobbs and Grant (4) reported that CCA treatment level was not an important factor in terms of the amount of arsenic released.

The sawdust from several cross-sectional cuts of the 2 by 4 was well mixed and stored in a dessicator for later use. The amount used for each combustion sample was approximately 850 mg.

Combustion procedures

A key factor in conducting this study was to select a combustion procedure that was reproducible and that would simulate the wide range of conditions under

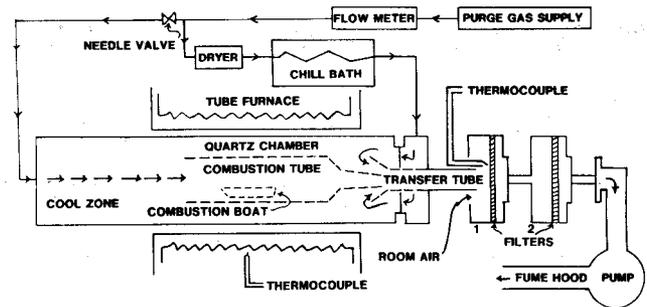


Figure 3. — Schematic drawing of horizontal combustion tube furnace and associated sampling systems (not to scale).

which CCA treated wood could be burned. It is well known, for example, that wood burns differently in fireplaces than in wood stoves, or under conditions of flaming and smoldering combustion. A horizontal tube furnace procedure developed in an earlier study (7) for burning pesticide-treated wood was selected for this study. The tube furnace allows small fuel samples to be subjected to various time/temperature conditions under dynamic flow conditions. It also permits quantitative sampling procedures to be applied to the combustion gases and particles generated during each run. As shown in the photographs (Figs. 1 and 2) and the line drawing (Fig. 3), the furnace is a 3-inch-diameter, 2-foot-long open tube electric furnace which can accommodate a 4-foot-long quartz tube. One-half of the quartz tube sits in the heated zone while the remainder lies outside in a cool (unheated) zone. A 1.5-inch-diameter, 30-inch-long ceramic combustion tube containing a small ceramic sample boat is placed inside the quartz chamber. The furnace can be heated up to 1,000°C, which far exceeds the conditions necessary for the complete combustion of wood. A heat controller can cause the furnace to heat the sample boat at a slow rate (0 to 15°C/min.). To attain rapid heating, the sample boat can be inserted from the cool zone into a preheated furnace. Thus, the furnace allows a range of heating rates and temperatures to be applied to the fuel and ash residue, with a high degree of precision in setting and reproducing the test conditions. For this study, we used the rapid heating or insertion method to combust the fuel in the furnace.

When defining and describing the conditions under which woody fuels are normally burned, it is convenient to use the terms "flaming" and "smoldering" combustion. Flaming combustion is an efficient burning process in which the fuel and emissions are rapidly oxidized and only a small amount, approximately 1 percent of black, sooty visible smoke is released. Smoldering combustion is an inefficient, slow-burning process in which a copious amount, approximately 10 percent of gray, tarry visible smoke is released. In most wood-burning situations, both processes take place with one usually dominant depending on the conditions of the fuel and the surrounding environment.

Our initial plan was to conduct the furnace experiments at 400° and 800°C to simulate the smoldering and flaming conditions just described. From previous ex-

periments confirmed in this study, we found that at 400°C we obtain smoke (particulate matter) similar in physical appearance and yield to that obtained from smoldering wood combustion. Similarly, at 800°C we obtain smoke characteristics and yield equivalent to flaming wood combustion.

It should be noted here that exothermic reactions in wood usually begin at temperatures around 320°C. Thus, we recognize that in the microenvironment surrounding the fuel particles in the furnace, temperatures higher than the furnace setting can occur. In fact, a small flame envelope can be observed over the fuel during the 800°C experiments. Even though exothermic reactions occur, the furnace operating characteristics permit a much higher degree of process control and experimental reproducibility than normally obtained in open combustion experiments. In addition, our furnace procedures permitted continued heating of the fuel residue and ash for well-defined time periods. This was important because we suspected that additional loss of arsenic would occur during these "ash-roasting" periods.

Sampling procedures

For idealized stoichiometric conditions, 1 g of wood requires a minimum of 6 g of air for complete combustion. Under actual burning conditions, both pyrolytic and oxidative decomposition occurs with the resulting release of smoke gases and particles. For this study, an air flow of 2.8/liters per minute, supplied from compressed air cylinders, was maintained through the furnace. This provided both excess combustion air and a means to sweep the combustion products out of the furnace and into the samplers.

We recognize that these flow conditions could result in an underventilated, fuel-rich condition during rapid exothermic reactions. However, we believe this simulates many of the conditions in which wood is normally burned, as discussed earlier.

The hot combustion gases exiting from the furnace are diluted and cooled with 26/liters per minute of air circulated through a cooling apparatus. The condensing particulate matter and gases are then passed through a glass transfer tube (TT) and a series of 47-mm filters. The first filter (which is partially open to room air) is a Type A, glass fiber filter (GFF) used to trap the condensing smoke as particulate matter. The second filter was employed to trap arsenic compounds in the gas or vapor state. It consisted of a Whatman No. 41 filter treated with a strong base, tetra-n-butylammonium hydroxide (TBAH) as suggested by Appel (1) and Walsh (10) for the collection of arsenic (III) oxides. Air flow through the filters was maintained above 30 liters per minute by a vacuum pump located downstream from the filter.

This system of sampling was selected after a series of trial-and-error attempts with various forms of open, closed, and manifold sampling procedures. We fixed on the flow and samplers described after preliminary runs at 400°C showed >95 percent recovery of arsenic. No attempt was made to optimize the sampling procedures

for copper and chromium even though these elements were included in subsequent analyses.

For each run, the following samples were collected for chemical analysis:

1. Glass transfer tube;
2. Glass fiber filter (Filter No. 1);
3. TBAH treated filter, (Filter No. 2);
4. Fuel residue (ash).

In addition, a sample of the powdered-wood starting material was analyzed for total arsenic, copper, and chromium several times throughout the course of experiments.

Analytical methods

Total CCA analysis. Samples were analyzed by the Cooperative Extension Service Laboratory, University of Georgia, for total copper, chromium, and arsenic by Atomic Absorption (AA) methods. Copper and chromium were analyzed by graphite furnace procedures and had a detectability limit of 0.2 µg/sample. Arsenic was analyzed by a hydride generation procedure and had a detectability limit of 0.1 µg/sample.

Background analyses run on filter blanks and untreated wood gave negligible elemental residues. In addition, a number of spiking experiments were conducted with a 2.2 percent Type III CCA stock solution. Quantitative recovery was achieved for copper, chromium, and arsenic from all filter and ash samples fortified with the stock solution at low (≈ 5 µg) and high (≈ 50 µg) levels.

Arsenic speciation. Several experiments at 400°, 800°, and 1,000°C were conducted for the purpose of arsenic speciation. The samples were analyzed at the USDA Pesticide Degradation Laboratory in Beltsville, Md. Repeat runs at 400° and 800°C incorporated a third filter in the sampling train to see if additional gaseous forms of arsenic could be trapped. It consisted of a 37mm in-line filter holder containing five layers of Whatman 541 filter paper treated with mercuric nitrate (6). The TBAH samples were extracted with 1:1 methanol, 3.2 moles of ammonium hydroxide solution while filter one, ash, and starting material were shaken overnight in 5N HCl. An aliquot of each extract was diluted to an appropriate volume and injected onto 2 Brownlee Labs PPAGU high-pressure liquid chromatography (HPLC) guard columns connected in series. The sample was separated via HPLC with a water/0.2 mole of ammonium carbonate gradient. The water contained 0.001 mole of quinolinol sulfate and the ammonium carbonate contained 0.001 mole of citric acid. Arsenite eluted at fractions 4-5 and arsenate eluted at fraction 15-16 and were analyzed with a graphite furnace Atomic Absorption Spectrophotometer.

Results and discussion

The CCA treated wood used as starting fuel was analyzed at various times throughout the study. The following results expressed as parts per million (\pm standard deviation) were obtained:

Copper	1,642 \pm 28	n = 9
Chromium	3,159 \pm 310	n = 8
Arsenic	2,324 \pm 175	n = 13

These values are consistent with the elemental proportions in the Type III CCA specification and were used as the basis for calculating the copper, chromium, and arsenic percentages shown in Tables 1 to 3.

In addition to several preliminary runs, a series of 26 furnace experiments was conducted between 400° and 1,000°C. Our initial plan called for replicated experiments at 400°, 600°, and 800°C for time periods of 10, 20, and 30 minutes. After obtaining some early results, we dropped the 600°C experiments and added experiments at 1,000°C and longer time periods (60 and 360 minutes).

The percentage of copper, chromium, and arsenic volatilized (lost) from the treated wood for each of the furnace treatments is shown in Table 1. Three replicates were run for all treatment levels except for duplicate runs for the 360-minute, 1,000°C treatment.

Except for one case, all fuel was reduced to a finely divided black residue that looked similar to small pieces of steel wool. The 10-minute treatments at 400°C left approximately 10 percent of the original fuel as a black char residue. As shown in Table 1, the various furnace treatments did not have a dramatic impact on the loss of copper and chromium. An average of 11 percent copper and 15 percent chromium was lost for all runs. The loss of arsenic, on the other hand, increased with increasing

furnace temperature and residence time. At 400°C, an average of 22 percent arsenic was lost. This result is consistent with the findings of Watson who examined the smoke from burning CCA treated sawdust (11) and wood shavings (12) which are used to smoke fish and bacon. Watson reported a temperature of 415°C in the smoldering sawdust and losses of arsenic between 10 and 22 percent.

We also experienced a loss of 22 percent arsenic in our 10-minute runs at 800°C. This result is consistent with the findings of Dobbs and Grant (4) who reported a loss of 24 percent when CCA treated wood particles were burned with a Bunsen burner. Their burn times were short (6 to 9 min.) with reported flame temperatures of approximately 900°C.

In our experiments, the amount of arsenic released increased dramatically when the ash residue was heated for extended periods. An average of 40 percent arsenic was lost for the longer (20 to 60 min.) runs at 800°C and this jumped to 70 and 77 percent for extended heating periods at 1,000°C. These results can be compared with a 42 percent loss when the CCA treated wood was burned over a domestic grate and a 90 percent loss when burned in a commercial incinerator. These latter two findings cited by Dobbs and Grant (3, 8) are not well documented, but clearly are situations where ash resi-

TABLE 1. — Percentage of fuel consumed and CCA components volatilized upon burning treated wood in a tube furnace.

Exposure time (min.)	Furnace temp. (°C)	Fuel consumed Mean (SD) ^a	Percent volatilized		
			Copper Mean (SD)	Chromium Mean (SD)	Arsenic Mean (SD)
10	400	89.4 (1.7)	10 (2.7)	9 (1.8)	27 (6.1)
20	400	98.6 (0.1)	13 (1.9)	11 (6.3)	13 (1.4)
30	400	98.8 (0.1)	20 (6.0)	21 (3.7)	27 (6.2)
10	800	98.9 (0.1)	6 (2.9)	15 (2.6)	22 (6.3)
20	800	98.9 (0.1)	7 (7.8)	17 (4.9)	42 (4.6)
30	800	98.9 (0.0)	7 (1.9)	19 (2.4)	34 (4.0)
60	800	98.7 (0.0)	8 (6.6)	6 (4.8)	44 (3.6)
60	1,000	99.0 (0.1)	16 (7.9)	22 (8.3)	70 (1.9)
360	1,000	99.1	No Data	No Data	77

^aSD is standard deviation, $n = 3$ in all cases except 360-min. runs where $n = 2$.

TABLE 2. — Percentage of arsenic recovered in sampling train components and ash for various time/temperature treatments.

Exposure time (min.)	Furnace temp. (°C)	Percent recovered				
		TT ^a Mean (SD) ^d	GFF ^b Mean (SD)	TBAH ^c Mean (SD)	Ash Mean (SD)	Total Mean (SD)
10	400	0.9 (0.8)	15.5 (1.0)	0.26 (0.08)	73 (6.2)	89 (5.3)
20	400	0.7 (0.4)	15.4 (1.6)	0.34 (0.05)	87 (1.8)	104 (2.9)
30	400	1.6 (1.6)	15.1 (1.3)	0.42 (0.12)	73 (6.2)	90 (6.4)
10	800	4.1 (0.2)	15 (1.4)	0.20 (0.07)	78 (6.3)	98 (6.3)
20	800	5.8 (0.5)	21 (0.9)	0.43 (0.30)	58 (4.6)	86 (4.8)
30	800	4.8 (0.9)	26 (1.1)	0.31 (0.11)	66 (4.0)	97 (1.9)
60	800	3.8 (0.7)	12 (1.6)	2.5 (2.3)	56 (3.6)	74 (4.4)
60	1,000	8.0 (1.5)	20 (7.6)	2.2 (2.3)	31 (1.9)	61 (2.5)
360	1,000	No Data	No Data	No Data	23	No Data
					Average recovery (all treatments)	87 (14)

^aTT = glass transfer tube.

^bGFF = glass fiber filter.

^cTBAH = Tetra-n-butylammonium hydroxide treated filter.

^dSD = standard deviation, $n = 3$ in all cases except 360-min. runs where $n = 2$.

TABLE 3. — Percentage of arsenite/arsenate in sampling train components for various furnace temperatures (average of two 30-min. runs).

Furnace temp. (°C)	Arsenic form ^a	Percent recovered					Ash	Total
		TT ^b	GFF ^c	TBAH ^d	Hg(NO ₃) ₂ ^e			
400	+3	1.1	7.0	ND ^f	0.1	ND	8.2	
	MAA	tr ^g	0.3	ND	tr	ND	0.3	
	+5	0.3	7.6	0.5	0.1	70.6	79.1	
						Sum	87.6	
800	+3	6.9	18.3	ND	tr	ND	25.2	
	MAA	0.1	0.2	ND	tr	ND	0.3	
	+5	1.5	3.1	1.4	tr	69.2	75.2	
						Sum	100.7	
1,000	+3	NA ^h	12.6	tr	NA	0.1	12.7	
	+5	NA	11.1	2.4	NA	46.2	59.7	
							Sum	72.4

^aArsenic form: +3 is arsenite; +5 is arsenate; MAA is methanearsonate.

^bTT = glass transfer tube.

^cGFF = glass fiber filter.

^dTBAH = tetra-n-butylammonium hydroxide treated filter.

^eHg(NO₃)₂ = mercury nitrate treated filter.

^fND = not detected.

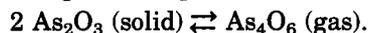
^gtr = trace less than 0.1 percent.

^hNA = not analyzed.

due could have been subjected to extended periods of heating.

In summary, our furnace experiments explain the source of variability that has been reported in the literature concerning how much arsenic is released when CCA treated wood is burned. It appears that arsenic release is not so much a function of how the fuel is burned, but rather how long the residual ash is exposed to high temperatures. Thus, approximately 22 percent of the arsenic will be released in situations without extended heating, whereas between 40 to 77 percent will be released in cases where there is the opportunity for prolonged high temperature ash heating (up to 6 hr.).

The distribution of arsenic in the sampling train components are given in Table 2. For all experiments run for 30 minutes or less, we recovered an average of 95 percent of the original arsenic. Sampling efficiency decreased at furnace times above 30 minutes where we were able to recover only 74 and 61 percent of the arsenic. In those cases, it is likely that some of the collected arsenic desorbed from the samplers during the extended sampling periods. Of the arsenic that was volatilized and recovered, essentially all was trapped in the condensed (particulate) form on the glass fiber filter and to a lesser extent on the glass transfer tube. Only trace amounts of arsenic were found on the TBAH filter, which has been reported to be 90 percent effective for trapping volatile arsenic trioxide (As₂O₃). As₂O₃ volatilizes according to the equilibrium:



In the experiments where heating exceeded 30 minutes, a significant but small amount of arsenic was trapped on the TBAH filter, which supports the speculation that arsenic desorption was probably occurring during these runs.

For copper and chromium, the sampling procedures proved to be unreliable for trapping volatilized copper and chromium compounds. In all our experiments, only

trace amounts of copper and chromium were found in the glass transfer tube, glass fiber filter, and TBAH filter. Yet, based on analysis of residual ash, we see an average of 11 percent loss of copper and 15 percent loss of chromium. Based on our quantitative recovery of copper and chromium from ash samples fortified with a CCA solution, we are confident that these losses are real. Early work by Dobbs and Grant (3) reported negligible losses of copper and chromium. Their findings, however, were based on an analysis for copper and chromium in the sampling train and not based on a material balance and ash analysis. Other work cited by Dobbs and Grant (3, 8), but unavailable for review, reported a loss of 13 percent chromium, similar to our findings.

Arsenic speciation

The speciation of the volatile arsenic compounds from CCA treated wood combustion has not been widely studied other than to show that negligible amounts of gaseous arsine (H₃As) are formed (3). One would expect thermal decomposition of the pentavalent arsenic in the wood to yield trivalent arsenic trioxide (As₂O₃ or As₄O₆) as well as various trivalent arsenites (AsO₃⁻³) and pentavalent arsenates (AsO₄⁻³). Arsenic trioxide begins to sublime at 135°C and it is generally agreed that trivalent arsenic is considerably more toxic than pentavalent arsenic (2), so the question of whether volatilized arsenic exists in the form of arsenite or arsenate is very important.

We ran duplicate 30-minute experiments at 400°, 800°, and 1,000°C for our arsenic speciation work. Results of the replicates were in close agreement and the averaged results are tabulated in Table 3. The percentages of arsenic volatilized (100-ash residue level) are similar to results shown in Table 1. Total arsenic recoveries are similar to those seen in Table 2. However, the arsenite/arsenate recovery ratio on the glass fiber filters was 6 to 1 at 800°C and only 1 to 1 at 400° and

1,000°C. It is not clear from these experiments whether there is an arsenite maxima at 800°C or simply that our sampling procedures were not quantitative at 1,000°C. Arsenic trioxide, which is somewhat volatile, is difficult to trap efficiently. The lower overall recovery at 1,000°C indicates that trapping with TBAH is probably not quantitative under these conditions. The addition of the mercury nitrate trap did not improve arsenite/arsenate recoveries. Essentially, all the arsenic recovered in the residue (ash) was in the pentavalent (arsenate) form.

Conclusions

1. Results of this study indicate that the amounts of arsenic volatilized when CCA treated wood is burned will vary according to the temperature and duration of the combustion process;

2. Approximately 22 percent arsenic is evolved when CCA treated wood is burned under low temperature (400°C) conditions;

3. Up to 77 percent arsenic was evolved under burning conditions in which the CCA treated wood and resultant ash were subjected to prolonged high temperature (800 to 1,000°C) heating;

4. Of the volatilized arsenic recovered, essentially all was found in the condensed (particulate) form and consisted of both trivalent arsenites and pentavalent arsenates;

5. An average of 11 percent copper and 15 percent chromium were also evolved under the combustion conditions studied.

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