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Assessing Exposure to 4,4'-Methylene bis(2-chloroaniline) (MBOCA) in the Workplace

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The chemical 4,4'-methylenebis(2-chloroaniline) (MBOCA) is used commercially as a curing agent for cast polyurethane products. While MBOCA has a low vapor pressure suggesting little likelihood of airborne vapor concentrations, worker exposure to MBOCA is indicated by its presence in urine, suggesting that MBOCA is readily absorbed through the skin. This study was conducted to assess various environmental exposure measurements (air samples, surface wipes, and skin pads) with comparison to biomonitoring (urine samples) results. The inhalation exposures to MBOCA measured by personal sampling were below $1 \mu\text{g}/\text{m}^3$; however, the area sampling indicated air concentrations up to $92 \mu\text{g}/\text{m}^3$. Wipe sampling indicated moderate contamination of the workplace by MBOCA dust (up to $19 \mu\text{g}/100 \text{cm}^2$). The average MBOCA concentration found on skin pads worn on workers' hands was generally less than $10 \mu\text{g}/\text{set}$, with a high of $25 \mu\text{g}/\text{set}$. Sixty-six percent of urine samples had detectable concentrations of MBOCA (up to $159 \mu\text{g}/\text{L}$). The urinary MBOCA results were generally consistent with the workers' potential for exposure as predicted by their proximity to MBOCA sources in their duties, the use of personal protective equipment, and general work practices. Clapp, D.E.; Piacitelli, G.M.; Zaebs, D.D.; Ward, E.: Assessing Exposure to 4,4'-Methylenebis(2-chloroaniline) (MBOCA) in the Workplace. *Appl. Occup. Environ. Hyg.* 6:125-130; 1991.

Introduction

4,4'-methylenebis(2-chloroaniline) (MBOCA) is a curing agent which is reacted with isocyanate-based polyurethane polymers to create a urethane material with a wide variety of uses ranging from industrial products such as gears, gaskets, belts, and rollers to consumer products such as sport boots and skate board wheels. Because of MBOCA's structural resemblance to benzidine, a known human bladder carcinogen, and its recognized carcinogenicity in animals, MBOCA is classified as a potential human carcinogen, e.g., by the International Agency for Research on Cancer (IARC) and the National Institute for Occupational Safety and Health (NIOSH).⁽¹⁾ The detection of three noninvasive papillary tumors of the bladder in a

study of MBOCA production workers is consistent with the hypothesis that MBOCA induces bladder neoplasms in humans.⁽²⁾

The federal Occupational Safety and Health Administration (OSHA) permissible exposure limit (PEL) for MBOCA of $220 \mu\text{g}/\text{m}^3$ (0.020 ppm) became effective September 1, 1989, and includes a "skin" notation which requires that an employee's skin exposure to MBOCA "shall be prevented or reduced to the extent necessary in the circumstances through the use of gloves, coverall, goggles, or other appropriate protective equipment, engineering controls, or work practices."⁽³⁾ For reference purposes, California/OSHA currently enforces exposure limits for air sampling ($10 \mu\text{g}/\text{m}^3$), wipe sampling ($100 \mu\text{g}/100 \text{cm}^2$ of surface area), and urinary monitoring ($100 \mu\text{g}/\text{L}$ urine).⁽⁴⁾ The American Conference of Governmental Industrial Hygienists (ACGIH)⁽⁵⁾ recommends an ambient air Threshold Limit Value (TLV) of $220 \mu\text{g}/\text{m}^3$, while NIOSH⁽⁶⁾ recommends that no worker be exposed to air concentrations exceeding $3 \mu\text{g}/\text{m}^3$. ACGIH and NIOSH do not currently recommend any standards for wipe sampling or urinary MBOCA levels.

The assessment of worker exposure to MBOCA is complicated by the physical properties of this chemical as well as the lack of a single effective environmental sampling methodology. MBOCA is normally a solid (pelletized) at room temperature and must be melted at 50°C for blending with polyurethane polymers (which are liquids at room temperature). Molten MBOCA has a low vapor pressure (i.e., $< 0.000036 \text{ mmHg}$ at 100°C) and is, therefore, unlikely to become airborne in the breathing zone of most workers.

Linch *et al.*, in one of the earliest studies of MBOCA exposure, demonstrated levels of urinary MBOCA among

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workers with no measurable airborne exposure, suggesting that most MBOCA exposure results from skin contact and could be indirectly measured by urinary monitoring.⁽⁷⁾ The MBOCA-cured urethane industry has, therefore, turned to urinary MBOCA measurement for assessing worker exposures to MBOCA. However, problems and limitations with urinary monitoring for MBOCA have been previously identified.⁽⁸⁾ This study was conducted to assess various environmental exposure measurements with biomonitoring results and to make recommendations to minimize worker exposures to MBOCA.

Plant Description

In May 1986, a study of occupational exposures to MBOCA was conducted at a Midwestern company that manufactures commercial polyurethane products and is a large user of MBOCA.⁽⁹⁾ Process operations are separated within a single building into three major departments: Urethane, Machine Shop, and Wood Model. The process studied was the manufacture of MBOCA-cured urethane elastomer and its subsequent molding in the molding area of the Urethane department.

During the survey period, the production of MBOCA-cured urethane began with the receipt of drums of dry MBOCA pellets which were handled and opened by a worker called a "mixer." The mixer transferred MBOCA pellets by hand from the shipping containers to a melting pot, melted the chemical, and dispensed molten MBOCA to "molders" who mixed hot MBOCA (either manually in a disposable container with a hand stirrer or using an automatic machine) with an isocyanate prepolymer to form a moldable urethane prior to pouring into molds. The urethane mixture was then poured into a heated metal mold and cured at either room temperature or in an oven, depending on product specifications. "Trimmers" removed the finished urethane part from the molds and then inspected, trimmed, and prepared the products for shipment. A "cleanup man," responsible for sweeping and housekeeping, and "supervisors" also intermittently move through the Urethane department.

All employees in the Urethane department wore disposable latex "surgical" gloves; in addition, the mixers wore Tyvek[™] coveralls and were observed to sometimes wear disposable dust masks.

Methods

Environmental Sampling

NIOSH industrial hygienists collected air, surface wipe, and skin pad samples from workers in the Urethane department. Both area and personal, full-shift air samples were collected. Personal air samples for MBOCA dust were collected in the workers' breathing zones using sampling pumps calibrated at 2 L/min and worn by the workers; a filter cassette (with a 37-mm Gelman Type AE acid-coated glass fiber filter) attached at the shirt collar was connected by Tygon[™] tubing to the pump. Area samples for MBOCA

dust were collected using an identical sampling train at some representative locations throughout the department to investigate background concentration of MBOCA.

To assess potential skin contamination, each worker wore gauze pads on his most frequently used hand for a full work shift. These "hand monitors" were prepared by attaching elastic straps to two small (approximately 1-inch square) surgical cotton gauze pads. The monitors were affixed on the outside of latex gloves worn by workers with one pad on the palm and the other pad on the back of the hand. This sampling was chosen to simulate worst case conditions (i.e., *no* glove protection worn). This technique has been previously used for assessing exposures to contaminants readily absorbed through the skin among pesticide workers.⁽¹⁰⁾

Wipe samples were taken to quantify the contamination on surfaces which workers may potentially contact. Samples were collected by wiping a 100-cm² area with dry Whatman filter tabs in a square pattern. Sampling locations were selected from surfaces where workers were predicted to most likely contact settled MBOCA dust or condensed vapor, if such contamination was present. For comparison purposes, some wipe samples were collected in areas infrequently touched (e.g., tops of storage cabinets) to determine background contamination.

The air sample filters, wipe tabs, and hand monitor pads were analyzed using a two-detector approach developed by the NIOSH Division of Physical Sciences and Engineering specifically for this study to maximize analytical sensitivity.⁽¹¹⁾ Filter samples were desorbed in 4 ml, gauze pads in 10 ml, and wipe tabs in 2 ml of 0.1N KOH in methanol. Samples were analyzed using a Waters high-performance liquid chromatography (HPLC) system utilizing a C₁₈ column in a radial compression mode, a mobile phase of 50 percent water/50 percent acetonitrile with 0.1N sodium acetate and a flow rate of 1 ml/min for separation. Quantitation was performed by a fixed ultraviolet (UV) detector (set at 250 nm); all samples which gave a response less than 2 µg/ml were reanalyzed using an electrochemical detector (voltage potential set at 0.85 mV in the oxidative mode). The choice of 2 µg/ml of MBOCA per sample as the break between the UV and electrochemical quantitation was arbitrary. The analytical limit of detection (LOD) using this approach was 0.015 µg/filter, while the limit of quantitation (LOQ) was 0.050 µg/filter for the air samples. The LOD for the gauze pads was 0.040 µg/pad (LOQ of 0.12 µg/pad) and the LOD for the wipe samples was 0.008 µg/tab (LOQ of 0.025 µg/tab). The LODs and LOQs were calculated based on a linear regression analysis of calibration curves. The LOD is that level at which an instrument response can be attributed to the presence of MBOCA, and the LOQ indicates the point at which an indicated response is within acceptable confidence limits and the concentration can be quantified.

Biomonitoring

NIOSH researchers collected urine samples from consenting workers as an index of MBOCA exposure. The large

number of participants and the logistics of urine collection prevented the collection of all voids during a work shift; therefore, all of the samples collected were spot voids collected at the end of the work shift (except for mixers, who also submitted urine samples pre- and postshift). Workers also submitted a urine sample before and after the two-day weekend without occupational exposure to provide information concerning the clearance of urinary MBOCA.

To investigate possible MBOCA contamination outside of the Urethane department, urine samples were also obtained from seven office employees ("controls"). To provide a set of known blank samples, five aliquots of urine were obtained from a bulk supply of MBOCA-free urine at the NIOSH laboratories. All participants were requested to wash their hands before voiding to minimize direct contamination of the urine samples.

Each participating worker was provided 250-ml Nalgene bottles for collection of urine samples. All urine collected was preserved in 30 percent citric acid (w/w) and was stored frozen until analyzed. The specific gravity of each urine sample was determined using a urinometer, and samples were subsequently analyzed for MBOCA content by HPLC (LOD = 5 µg/L).⁽¹²⁾

Results and Discussion

Air Sampling

Six of ten area samples were collected at fixed, nonwork area locations to investigate general background airborne levels. The remaining four area samples were collected directly adjacent to the MBOCA melting pot (where large quantities of pure MBOCA are handled) to estimate the "worst case" concentration. This location was chosen primarily to determine if air sampling would produce detectable (i.e., > LOD) results where the highest MBOCA air concentrations would be expected.

The results of the personal and area air sampling are summarized in Table I. Not surprising, since previous research^(7,13) has suggested that MBOCA is not readily va-

TABLE II. Summary of Wipe Sampling Results

Location	Number of Samples	MBOCA Concentration (µg/100 cm ²)	
		Avg.	SD*
MBOCA room near melting pot	3	19.1	29.4
Top of standing cabinet, Molding Dept.	2	4.7	5.4
MBOCA room dispensing counter	3	1.4	0.2
Trimmer work table	1	0.1	—
Molder work table	14	0.5	0.8

*SD = standard deviation.

porized, very little MBOCA was detected in the majority of either area or personal air samples. Of the 35 personal samples, 9 were above the LOD, with only 5 above the LOQ. The five detectable results from personal samples were very low—all below 1 µg/m³. Of the ten area samples, only the four "worst case" samples from the MBOCA melting area had detectable results. It should be remembered, however, that these "worst case" results did not represent actual worker exposures.

Wipe Sampling

All 23 wipe samples collected indicated low (when compared to the California OSHA [Cal/OSHA] standard of 100 µg/100 cm²) but detectable MBOCA contamination throughout the Urethane department as shown in Table II. As with the highest area air samples, the highest MBOCA contamination, averaging 19.1 µg/100 cm², was found in the areas immediately adjacent to the MBOCA melting pot. The average contamination level found on work tables of molders and trimmers was about 0.5 µg/100 cm². Two samples collected on the top of storage cabinets (covered with accumulated dust) did reveal MBOCA contamination (4.7 µg/100 cm²) several times higher than on work tables (0.5 µg/100 cm²) which were regularly wiped clean. Overall, these results indicate the presence of MBOCA contamination throughout the Urethane department on most horizontal surfaces where dust could settle.

Hand Monitors

Twenty-four of the 25 sets of hand monitors in the Urethane department contained detectable MBOCA contamination as shown in Table III. In calculating the results presented here, the total MBOCA detected on both the palm and back-of-hand pads were summed to yield a single value for each set of pads. The averaged data indicate that the day mixer, who most directly handled MBOCA, had the highest contamination on his gloves (average 24.6 µg/set of pads), which confirmed expectations.

Biomonitoring

Twenty-four of 34 (71%) exposed employees in the Urethane department consented to provide spot urine voids. A total of 72 spot urine voids were collected from the participating exposed and nonexposed (control) workers during the three-day survey. Thirty-eight of 58 samples

TABLE I. Summary of Air Sampling Results

Type	Number of Samples	MBOCA Concentration (µg/m ³)
Area		
General work area	6	ND*
MBOCA melting pot (day 1)	1	61
MBOCA melting pot (day 2)	1	92
MBOCA melting pot (day 3)	1	58
MBOCA melting pot (night 1)	1	12
Personal		
Molder A (day)	1	0.70
Molder B (day)	1	0.13
Molder C (day)	1	0.13
Mixer A (night)	1	0.06
Mixer B (day)	1	0.34
All other samples	35	ND

*ND = Not detected (limit of detection = 0.015 µg/liter).

TABLE III. Summary of Hand Monitor Sampling Results

Job	Number of Samples		MBOCA Concentration ($\mu\text{g}/\text{set}$) ^A	
	Total	< LOD ^B	Avg.	SD ^C
Mixer (day)	2	0	24.6	16.4
Molder (day)	13	0	7.3	9.5
Mixer (night)	2	0	4.7	2.4
Molder (night)	6	1	3.0	2.4
Trimmer (day)	2	0	2.3	1.5

^ASum of both palm and back-of-hand pads.

^BLOD = limit of detection.

^CSD = standard deviation.

(66%) submitted by exposed workers contained detectable urinary MBOCA concentrations. The distribution of urinary MBOCA concentrations, by worker category, is shown in Table IV. To conform to current practice by Cal/OSHA in its urinary standard (which is used as an informal guideline by others, including the castable-urethane elastomer industry), the urinary MBOCA results presented here were adjusted for urine dilution by using specific gravity.⁽¹⁴⁾

All ten samples submitted by the two MBOCA mixers, who were the most directly exposed workers, contained detectable MBOCA concentrations; two of these samples exceeded the Cal/OSHA standard of 100 $\mu\text{g}/\text{L}$ urine. Twenty-five of 35 samples (71%) submitted by 16 molders, who were more distant from direct exposure, contained detectable concentrations, but these concentrations were much lower (50 $\mu\text{g}/\text{L}$ or less) than for mixers. None of the samples submitted by trimmers and supervisors, who were expected to have the least exposure, contained detectable MBOCA concentrations. None of the 14 urine samples submitted by controls contained detectable (> 5 $\mu\text{g}/\text{L}$) MBOCA concentrations which indicated the absence of MBOCA exposure outside of the production area.

A more detailed presentation of urinary MBOCA concentrations from molders and mixers is shown in Table V. The overall average urinary concentrations for the two MBOCA mixers (61.9 $\mu\text{g}/\text{L}$) was more than four times higher than the overall average for molders (14.8 $\mu\text{g}/\text{L}$). From these results, it is clear that: 1) workers (particularly mixers) may not be wearing protective gloves correctly (e.g.,

irregular wearing of gloves; wearing damaged gloves, possibly with interior contamination); 2) gloves alone do not provide complete and effective protection from dermal absorption of MBOCA; and 3) dermal exposures are highest for the mixers who come in close contact while transferring dry MBOCA from storage drums, melting MBOCA, and dispensing molten MBOCA.

Three of four urine samples submitted by the cleanup man contained detectable MBOCA concentrations (not shown in Table V) averaging 11.1 $\mu\text{g}/\text{L}$ (SD = 0.5). Although these concentrations were generally less than those of MBOCA mixers or molders, these results suggest that MBOCA exposures probably occur from dermal contact with the contaminated surfaces in the Urethane department.

Thirteen workers provided urine samples on a Monday morning after two days away from work. These results are summarized in Table VI. Only the day mixer, who had the second highest average (29.9 $\mu\text{g}/\text{L}$) and highest peak (56.5 $\mu\text{g}/\text{L}$) urinary MBOCA levels during the prior week, presented a urine sample with a detectable MBOCA concentration on Monday. The other participants had no detectable concentrations. These data suggest that the biological half-life of MBOCA is relatively short, although such a finding was not an objective of this study.

Conclusions

In this study, the large percentage of air samples with nondetectable results confirms prior research which indicated that air sampling inadequately characterizes potential occupational exposures to MBOCA. Since MBOCA dust rapidly settles to surfaces and since very little vapor is generated during melting, air concentrations are expected to be low. Based upon the wipe sampling and hand monitor results, most potential for exposures occur when workers directly contact MBOCA or surfaces contaminated with MBOCA. This generally involves those workers required to work in areas where MBOCA is stored, transferred, mixed, or melted, and who may not regularly wear gloves, and whose gloves are prone to puncture and tearing.

While wipe sampling and hand monitoring sampling consistently revealed contamination on most surfaces, the magnitude of these results was low in comparison with

TABLE IV. Distribution of Urinary MBOCA Results

Job	Number of Workers	Number of Participants	Number of Samples MBOCA Concentration ($\mu\text{g}/\text{L}$ urine) ^A					Total
			ND ^B	< 5	5-50	50-100	> 100	
Mixer	2	2	0	0	5	3	2	10
Molder	24	16	10	3	22	0	0	35
Cleanup Man	1	1	1	0	3	0	0	4
Trimmer	5	3	4	0	0	0	0	4
Supervisor	2	2	5	0	0	0	0	5
Control	NA ^C	7	14	0	0	0	0	14
Blank	NA	NA	5	0	0	0	0	5
Total	34	29	39	3	30	3	2	77

^ANormalized to specific gravity of 1.019 per Cal/OSHA standardized practice (limit of detection = 5 $\mu\text{g}/\text{L}$).

^BND = not detected.

^CNA = not applicable.

TABLE V. Urinary MBOCA Results for Molders and Mixers

Job	Number of Participants	Number of Samples	MBOCA concentration ($\mu\text{g/L}$ urine) ^A		
			Avg.	SD ^B	Maximum
Mixer					
Day	1	5	29.9	14.9	56.5
Night	1	5	94.0	45.5	158.9
Total	2	10	61.9	46.6	158.9
Molder					
Day	11	23	9.5	9.4	39.3
Night	5	12	20.6	11.9	40.0
Total	16	35	14.8	12.0	40.0
Overall Total	18	45	28.8	34.0	158.9

^ANormalized to specific gravity of 1.019 per Cal/OSHA standard practice.

^BSD = standard deviation.

the CAL/OSHA exposure criteria. Despite the low relative surface and skin contamination levels, urinary MBOCA concentrations were well above the limit of detection, with a few samples exceeding the Cal/OSHA standard. Generally, the workers in the closest proximity to MBOCA presented the highest urinary MBOCA levels. Our study results also indicate that even exposure to relatively low levels of MBOCA from all sources can lead to substantial absorbed doses. Therefore, compared to environmental sampling, biological monitoring seems to provide the most useful and sensitive indication of occupational exposure to MBOCA.

Recommendations

1. Exposures to MBOCA must be vigilantly controlled by utilizing effective engineering controls, including local exhaust ventilation, enclosures, automation, and substitution; promoting conscientious work practices and frequent housekeeping; required wearing of company-supplied protective clothing (i.e., impermeable gloves, coveralls, and eye goggles) by any workers who directly handle MBOCA or who may have contact with contaminated surfaces; providing company laundering of work clothes and shower facilities for employees; and educating all workers of the hazards and the proper handling of MBOCA.
2. Environmental surveillance should be regularly performed to assess potential exposures to MBOCA. Because low air concentrations do not indicate low exposure potential, primarily due to the low volatility of MBOCA, methods for evaluating surface contamination (wipe sampling) and skin contact (skin pads) should also be considered to supplement air sampling. For example, an inexpensive, semiquantitative "spot test" is available for assessing surface contamination (a pre-measured area is wiped with a wipe tab, reagent is added to the tab which is viewed under UV light with brightness of fluorescence proportional to MBOCA contamination).⁽¹⁵⁾
3. Biomonitoring of urinary MBOCA concentrations should be available and actively promoted for workers potentially exposed to MBOCA. Adequate consideration should be given to how such data are collected and used. In-

dividual metabolism of the chemical is not well understood as well as the ultimate issue of carcinogenic risk in humans. Despite these uncertainties, an aggressive program to reduce and monitor MBOCA exposures is essential since positive urinary concentrations of MBOCA indisputably indicate an absorbed dose (which should be eliminated or, at least, minimized).

4. A registry of persons exposed to MBOCA should be established for use in future epidemiologic studies. Documentation of employee medical data, work histories, exposure information, and methods of control should be included.

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TABLE VI. Postweekend Urinary MBOCA Results

Job	Urinary MBOCA Concentration ($\mu\text{g/L}$) ^A			
	Prior Week		Postweekend	
	Avg.	Maximum	n	Avg.
Mixer (day)	29.9	56.5	1	8.9
Molder (night)	20.6	40.0	2	ND ^B
Cleanup	11.1	29.9	1	ND
Molder (day)	9.5	39.3	6	ND
Trimmer	ND	ND	1	ND
Supervisor	ND	ND	2	ND

^ANormalized to specific gravity of 1.019 per Cal/OSHA standardized practice (limit of detection = 5 $\mu\text{g/L}$).

^BND = not detected.

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