"Extent of Industrial Exposure to Epichlorohydrin, Vinyl Fluoride, Vinyl Bromide and Ethylene Dibromide"

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ABSTRACT

Industrial hygiene studies were conducted in several industries to evaluate present levels of worker exposure to hazardous chemicals and to determine the feasibility of worker mortality studies.

The greatest exposure to epichlorohydrin in the chemical and resin manufacturing processes occurs with chemical operators. In the chemical manufacturing processes, operator exposures were found to be less than 2.1 ppm for 20 TWA personal samples and in the resin manufacturing processes, operator exposures were found to be less than 0.8 ppm for 39 TWA personal samples.

Chemical operator exposure levels for vinyl fluoride in the manufacture and polymer processes were generally found to be less than 5 ppm for 18 TWA personal and general area samples. One sample was found to be 21 ppm during the initial start-up shift. Operator exposure to vinyl bromide in the manufacturing process was found to be less than 0.4 ppm for 4 TWA personal samples. Tank car loading crewmen were found to have the greatest exposure, and 1.2 and 6.3 ppm VBr, respectively for two TWA personal samples taken.

Ethylene dibromide median exposures by similar job types in the manufacturing processes ranged 10 to 500 ppb for 35 TWA personal samples; while, median exposures by similar job types in antiknock blending operations ranged 0.2 - 54 ppb for 39 TWA personal samples. EDB ceiling levels for quality control sampling and for loading or unloading tank cars ranged 0.04 to 23 ppm for 7 samples and 0.09 to 2.4 ppm for 4 samples, respectively.

The reported levels are generally within permissible exposure limits. Many of the processes are closed chemical processing which minimize worker exposure. However, continued attention needs to be given to the open system processes and equipment maintenance to prevent worker exposure.

I. INTRODUCTION

The purpose of this paper is to present and summarize the pertinent findings of industrial hygiene studies of epichlorohydrin, vinyl halides (vinyl fluoride and vinyl bromide) and ethylene dibromide. Study of these substance agents have been conducted by NIOSH under contract with Tracor Jitco, Inc. and SRI, International. More complete information concerning the findings of these studies is contained in the published NIOSH technical reports: (1) Epichlorohydrin Manufacture and Use - Industrial Hygiene Survey, (2) Vinyl Fluoride and Vinyl Bromide Industrial Hygiene Survey Report, and (3) An Industrywide Industrial Hygiene Study of Ethylene Dibromide.

II. STUDY PROCEDURE

These chemical agents were selected by NIOSH for industrial hygiene study because they are suspect cancer agents and limited information exists on exposure levels in industry.

Walk-through surveys were conducted for each facility to obtain preliminary information about the process, production activity, potential exposures and worker exposure. Based on the walk-through survey, in-depth survey requirements are defined for each plant. Essentially, the in-depth survey procedures are directed toward determining 8-hour TWA personal exposure and characterizing work practices and controls. Sampling is conducted for all shifts (usually three) and potentially exposed workers. Job descriptions are characterized for potentially exposed workers. Appropriate survey sampling and analytical methods are determined. The NIOSH recommended sampling and analytical methods formed the basis for methodologies employed in the study. Recommended vinyl fluoride sampling methods were not available at the time of the study. Personal sampling for vinyl fluoride was conducted using a 7.7 liter Teflon bag attached to the sampling pump operating at flow rates of 14 to 100 cc/min. The samples were analyzed by gas chromatography within two days after sampling. Laboratory decay tests indica-

ted 10% loss in four days and 50% loss in two weeks for the Teflon bag used.

All other samples were collected on the standard 150 mg charcoal tube and analyzed by gas chromatographic techniques.

III. Epichlorohydrin

Studies were conducted at two epichlorohydrin manufacturing and user facilities and at three additional resin manufacturing facilities in 1976.

A. HEALTH CONCERN

The current Federal Standard for epichlorohydrin is 5 ppm (19 mg/m^3) as an 8-hour time-weighted average permissible exposure level. This is based on the known acute (short term) health effects to humans from over exposure, i.e., respiratory tract irritation and systemic poisoning. Skin contact can result in dermatosis and systemic effects. Exposures occur principally by inhalation and direct skin contact and to a less extent by ingestion. After a comprehensive literature review, NIOSH concluded that exposure risks may include carcinogenesis, mutagenesis, and sterility in humans and recommended a time-weighted average occupational exposure limit of 2 mg/m^3 of air (0.5 ppm) and a ceiling limit of 19 mg/m^3 (5 ppm) as reported in the Criteria Document (Sept. 1976). More recent information on human exposure data has prompted NIOSH to issue a Current Intelligence Bulletin-30 recommending exposures be reduced to the extent feasible. (Oct. 1978) The 1978 TLV for epichlorohydrin has been revised downward to 2 ppm for the 8-hour TWA and a short term exposure level of 5 ppm.

B. SURVEY RESULTS

1. Epichlorohydrin Manufacturing Process

Manufacturing plants for epichlorohydrin are of conventional open structure chemical process design. Both facilities manufacture epichlorohydrin for shipment and for use in their own manufacture of glycerine and epoxy resins.

Epichlorohydrin is made by the chlorination of allyl chloride yielding a mixture of dichlorohydrin. These products are washed with a cold dilute alkali solution to remove hydrochloric acid and yield impure epichlorohydrin. The epichlorohydrin is then further refined by distillation processes.

Survey Data

Plant A

The results of 8-hour TWA personal sampling at the first facility ranged from not detected (less than 0.05 ppm) to 0.4 ppm for 8 chemical operator samples (median of 0.3 ppm). Lower exposures were reported for the shift foreman, drumming operator and maintenance personnel. An exposure level of 0.3 ppm was reported for the tank car loader.

Plant B

Similar survey data was collected at the second manufacturing facility. Personal samples ranged from not detected (less than 0.05 ppm) to 2.1 ppm for 12 chemical operator samples (median of 0.1 ppm). However, two of these samples were considered significant levels, i.e., 1.9 ppm and 2.1 ppm. Lower exposures were reported for the shift and maintenance foreman. An exposure of 0.3 ppm was reported for the tank car loader. No drumming operations were performed during the survey of Plant B.

Controls

The production of epichlorohydrin operations are located out of doors

with automated operations monitored from a control room. For normal operation, operators are seldom in the process areas. They are in the production areas for routine inspection of equipment, product sampling and occasional on-stream maintenance. Maintenance work is supervised by the shift foreman with work practices in effect. Pumps and pipe flange seals are maintained to prevent leakage.

Tank cars and trucks are cleaned and inspected before loading by outside contractors. Loading involves connecting supply lines and vent lines, loading tank car in which the operator is located at a control site some distance away and the disconnecting once the tanks are filled. The most significant exposure to the loader is during the disconnect procedure. The loading operation takes about 2½ hours with about ½ hour for connect and disconnect time. Drumming operations employ local exhaust ventilation systems.

2. Epoxy Resin Manufacture

Five epoxy resin facilities were surveyed for epichlorohydrin exposure. The first two are also manufacturers of epichlorohydrin.

Process

The basic process involves reacting epichlorohydrin with bisphenol A under alkaline conditions. Depending on the particular resin being produced, reactants and/or solvents may be introduced to modify the resin properties and the viscosity of the liquified resin products. The reactants, depending on resin specifications may include bisphenol A, tetrabromo bisphenol A, o-cresol, paraformaldehyde, caustic soda, oxalic acid, and p-tertiary butyl phenol. The solvents may include methyl ethyl ketone, methyl isobutyl ketone, acetone, toluene, and xylene. Epoxy resins may be produced as thermoset (cured) resins, containing no free or unreacted epichlorohydrin.
Survey Data

Plant A & B

Exposure to epichlorohydrin were found to be less for the epoxy resin

production operations. Resin process chemical operator exposures were found to be less than 0.8 ppm for 22 TWA personal samples taken in the two epichlorohydrin manufacturing facilities. Lower exposures are reported for foremen and maintenance.

Plants C, D and E

At three additional epoxy manufacturing facilities, chemical operator exposures were less than 0.2 for 17 TWA samples. The highest level reported (1.5 ppm) is that for a general area sample taken in the isolated pump room at Plant C. This demonstrates the potential for exposure from equipment leakage.

Control

Epoxy resin processes are located both indoors and outside, with a variety of natural, general and local types of ventilation control. Reaction kettles are equipped with vent lines. Epichlorohydrin was fully reacted in processes studied so that packaging and shipping of finished products did not present epichlorohydrin exposure to workers.

IV. Vinyl Halides

Studies were conducted for vinyl fluoride exposure at a manufacturing facility and at a polymerization facility. Vinyl bromide was studied at a manufacturing facility.

Health Concern

The vinyl halides including vinyl fluoride and vinyl bromide have gained prominent health concern as a result of the recently reported cancer associated with vinyl chloride workers. In the absence of definitive Federal Standards for vinyl fluoride and vinyl bromide, NIOSH recommends that worker exposures be minimized and that levels be maintained below the established Federal Standard for vinyl

chloride. The Federal Standard for vinyl chloride is one ppm as an 8-hour TWA exposure limit. The 1978 ACGIH Threshold Limit Value for vinyl bromide has been revised downward for the 8-hour TWA exposure limit.

B. Survey Results

1. Vinyl Fluoride Manufacturer Process

Plant A is a manufacturer of vinyl fluoride. The basic process involves a pressurized reaction of hydrofluoric acid and acetylene. Difluoroethane is formed as an intermediate product that is cracked to yield ethylene fluoride and hydrogen fluoride. The reaction products are refined by distillation and the off-products are recycled to the process stream. Liquid vinyl fluoride is piped to insulated storage tanks and from there to insulated tank cars for shipment.

Survey Results and Control

Sampling results at Plant A were generally not detected due to interference from difluoroethane. Difluoroethane was measured to be 5 ppm and less. The vinyl fluoride was estimated to be less than 2 ppm for 7 personal and general area samples. One TWA personal operator sample was 21 ppm for the start-up process on the first shift. This demonstrates that during a work shift with abnormal or unusual work operations, the potential for greater exposure exists. With continued caution on the part of the operators and the use of respiratory protection, protective clothing and face shields during these operations, the actual worker exposure is minimized.

2. Vinyl Fluoride Polmerization Process

Plant B is a manufacturer of polyvinyl fluoride. Vinyl fluoride is received, transferred to storage tanks and piped to the vinyl fluoride polymerization building. The monomer is continuously pumped to a

supply tank at the process building and from there injected into water and pumped to the reactor to which an aqueous solution of the reaction initiator is simultaneously added. The reactor is barricaded from the remainder of the processing area since the reaction is conducted under high pressure carefully controlled. The reacted vinyl fluoride, a finely divided precipitate, is separated from the reactor aqueous liquor. Unreacted vinyl fluoride is recycled to the supply tank. The polymer is stored as a 5% aqueous slurry and fed to a rotary filter. The process is a closed system until the slurry (completely polymerized) is fed to the rotary filter.

The resulting filter cake (white odorless product) is further dried, collected in bag filters, classified and stored for further processing.

Survey Results

Sampling results for the vinyl fluoride polymer plant, Plant B, indicate TWA exposure levels below 5 ppm vinyl fluoride. Samples ranged from 1 to 4 ppm with a median of 2 ppm for 7 TWA personal samples of polymer operators. One general area sample in the pump room was 5 ppm vinyl fluoride.

3. Vinyl Bromide Manufacturing Process

Plant C is a manufacturer of vinyl bromide. Ethylene dibromide is continuously fed to a reactor where caustic reacts with the dibromide and yields vinyl bromide. The unreacted ethylene dibromide is removed by distillation and recycled. The vinyl bromide is pumped to storage tanks and transferred to railroad cars for shipment. The process is enclosed and the plant structure is open and outside. The process control instrumentation is located in a separate building, where operators monitor the process.

Survey Data

Operator exposures to vinyl bromide in Plant C range from 0.1 to 0.4 ppm with a median of 0.3 for 4 samples. Higher levels were associated with the laboratory analysis and loading operations. A level of 6.3 ppm for a one hour sample during loading vinyl bromide tank cars was reported. A level of 1.2 ppm was reported for the 8-hour TWA sample.

Control

The primary control in these processes involve enclosed processes and process equipment located out of doors. The integrity of process systems are maintained to present minimal exposure hazards to plant workers.

Certain potential exposure situations such as sample collection, coupling or decoupling tank lines and maintenance of process equipment require specialized controls and work practices.

V. Ethylene Dibromide

Two manufacturing and two blending operations were studied for exposure to ethylene dibromide.

A. Health Concern

The current Federal Standard for ethylene dibromide is 20 ppm determined as an 8-hour time-weighted average occupational exposure level. This is based on the acute health effects to the respiratory tract and systemic poisoning. Skin contact may result in skin irritation and systemic effects. Exposures occur principally by inhalation and skin contact and to a less extent by ingestion. A comprehensive review of the literature by NIOSH of animal studies indicate reproductive effects, carcinogenicity, mutagenicity and teratogenicity. Based on these adverse effects possibly associated with human exposure,

NIOSH recommended an employee exposure ceiling of 0.13 ppm (1.0 mg/m^3) in the Criteria Document of August, 1977. Based on the preliminary results of animal studies of a toxic interaction between disulfiram and ethylene dibromide, NIOSH published a Current Intelligence Bulletin-23, April 11, 1978. NIOSH recommended that workers should not be exposed to ethylene dibromide during the course of disulfiram therapy, used for treatment of alcoholics in industry. The TLV committee recognizes the carcinogenic potential of EDB as reported in animal studies and is considering a reduced recommended exposure limit.

B. Survey Results

1. EDB Manufacturing Process

Plants A and B are manufacturers of ethylene dibromide in continuous flow, closed system operations. Ethylene dibromide is produced by the exothermic reaction of bromine and ethylene in a countercurrent flow reactor. With bromine entering the top and gaseous ethylene entering the bottom, the reaction occurs in the upper portion of a column packed with ceramic chips. The crude liquid product is further refined, stored and loaded into tank cars for shipment.

Survey Data

The results of 8-hour TWA personal sampling at two manufacturing operations range from not detected (less than 0.02 PPB) to 1600 PPB. The higher levels were reported for the surveillance technician and lab technician at Plant A and the crew leader and product leader at Plant B. For normal plant operations these higher levels of exposure to ethylene dibromide result from open system operations.

Control

These processes are operated and monitored from remote control rooms.

Potential exposures to operating personnel occur for quality control sampling and analysis, loading and maintenance operations.

2. EDB Blending Process

Plants C and D produce antiknock blends using ethylene dibromide. Antiknock blends consist of homogeneous mixtures of ethylene dibromide, ethylene dichloride, tetraalkyl lead and may contain toluene and a dye. Raw materials are generally received by tank car, stored and pumped to the process area. Blending is performed in outdoor blending tanks. The batch-type process is activated and monitored from a control room. Manual operations involving potential worker exposure include loading and unloading tank cars, quality control sampling and analysis and drum loading.

Survey Data

The results of sampling for various operations for antiknock blending operations indicate lower overall exposures than occur in manufacturing. The results of 8-hour TWA personal sampling for two plants ranged from 0.1 to 8.2 ppb for 39 samples. Again, the higher exposures result from open system operations such as loading and unloading, drum cleaning, and sample collection. Drum loading is conducted using enclosed local exhaust hoods. The drums are reusable and undergo cleaning prior to reuse. Drum cleaning is conducted in an enclosed exhaust hood and the operators wearing respiratory protection.

3. Ceiling Levels

The final slide presents short term exposure data for quality control sampling, loading and unloading of tank cars at the various plants. These levels are expressed in ppm rather than ppb for the two prior slides.

The sample collection process varies from 5 to 30 minutes depending

on the number and location of sampling. Process stream sampling is conducted generally at the beginning of each shift. Tank car loading requires about one to two hours and may be sporadic depending on shipment schedule.

Most of these levels are above the NIOSH Criteria Document recommended ceiling level of 0.13 ppm for a 15 minute period.

VI. Conclusion

The reported exposure levels are considered typical of the industries represented. The primary control for each operation is the closed processing of chemicals. This is typical of the chemical processing industry in general. Particular attention needs to be given to those operation and processes which can not be conducted as closed processes and routine maintenance to detect and correct leaks or accidental chemical releases. The potential for health effects associated with the materials studied is significant to warrant application of ventilation systems, personal protection and safe work practices.

No discussion followed this paper.

EPICHLOROHYDRIN

	TWA	CEILING
FEDERAL STANDARD	5 PPM	-
NIOSH CRITERIA DOC.	0.5 PPM	5 PPM
1978 T.L.V.	2 PPM	5 PPM

TABLE 1.1

EPICHLOROHYDRIN SAMPLING DATA SUMMARY
EPICHLOROHYDRIN MANUFACTURERS
8-HR. TWA PERSONAL SAMPLES

PLANT / JOB	SAMPLE SIZE	RANGE (PPM)	MEDIAN (PPM)
PLANT A:			
CHEMICAL OPERATORS	8	N.D. - 0.4	0.3
SHIFT FOREMEN	3	0.1 - 0.2	0.1
DRUMMING OPERATOR	2	0.06 - 0.08	0.07
TANK CAR LOADER	1	0.3	0.3
PIPEFITTER (MAINTENANCE)	3	N.D.	N.D.

N.D. - NOT DETECTED BASED ON THE SAMPLING AND ANALYTICAL METHOD.

TABLE 1.2

EPICHLOROHYDRIN SAMPLING DATA SUMMARY
EPICHLOROHYDRIN MANUFACTURERS
8-HR. TWA PERSONAL SAMPLES

PLANT / JOB	SAMPLE SIZE	RANGE (PPM)	MEDIAN (PPM)
PLANT B:			
CHEMICAL OPERATORS	12	N.D. - 2.1	0.1
SHIFT FOREMEN	3	N.D - 0.3	N.D.
TANK CAR LOADER	1	0.3	0.3
MAINTENANCE FOREMAN	1	0.08	0.08

N.D. - NOT DETECTED BASED ON THE SAMPLING AND ANALYTICAL METHOD.

TABLE 2.1

EPICHLOROHYDRIN SAMPLING DATA SUMMARY

RESIN MANUFACTURERS

8-HR. TWA PERSONAL SAMPLES

PLANT / JOB	SAMPLE SIZE	RANGE (PPM)	MEDIAN (PPM)
PLANT A:			
CHEMICAL OPERATORS	6	N.D. - 0.4	N.D.
PLANT B:			
CHEMICAL OPERATORS	16	N.D. - 0.8	0.04
OPERATING FOREMEN	5	N.D - 0.6	N.D.
MAINTENANCE FOREMEN	1	N.D.	N.D.

N.D. - NOT DETECTED BASED ON THE SAMPLING AND ANALYTICAL METHOD.

TABLE 2.2

EPICHLOROHYDRIN SAMPLING DATA SUMMARY
RESIN MANUFACTURERS
8-HR. TWA PERSONAL SAMPLES

PLANT / JOB	SAMPLE SIZE	RANGE (PPM)	MEDIAN (PPM)
PLANT C:			
CHEMICAL OPERATORS	1	0.09	0.09
G.A. IN PUMP ROOM	1	1.5	1.5
PLANT D:			
CHEMICAL OPERATORS	2	0.05 - 0.15	0.1
PLANT E:			
CHEMICAL OPERATORS	14	N.D.	N.D.
OPERATING FOREMEN	1	N.D.	N.D.
RESIN FINISHING FLAKER	2	N.D.	N.D.

N.D. - NOT DETECTED BASED ON THE SAMPLING AND ANALYTICAL METHOD.

VINYL HALIDES

	TWA
VINYL FLUORIDE:	
FEDERAL STANDARD	-
NIOSH CRITERIA DOC.	1 PPM
1978 T.L.V.	-
VINYL BROMIDE:	
FEDERAL STANDARD	-
NIOSH CRITERIA DOC.	1 PPM
1978 T.L.V.	5 PPM
VINYL CHLORIDE:	
FEDERAL STANDARD	1 PPM
NIOSH CRITERIA DOC.	1 PPM
1978 T.L.V.	5 PPM

TABLE 3.1

VINYL HALIDE SAMPLING DATA SUMMARY

VINYL FLUORIDE

8 HR. TWA SAMPLES

PLANT/JOB/LOCATION	SAMPLE SIZE	RANGE (PPM)	MEDIAN (PPM)
PLANT A:			
PLANT OPERATOR	4	N.D.	N.D.
PLANT OPERATOR (START-UP PROCESS)	1	21	21
G.A. IN CONTROL ROOM	3	N.D.	N.D.
PLANT B:			
POLYMER OPERATOR	7	1-4	2
G.A. IN SUPERVISOR'S OFFICE	3	1-2	2
G.A. IN PUMP ROOM	1	5	5

G.A. - GENERAL AREA SAMPLES.

N.D. - NOT DETECTED BASED ON SAMPLING AND ANALYTICAL METHOD.

TABLE 3.2

VINYL HALIDE SAMPLING DATA SUMMARY

VINYL BROMIDE

PLANT/JOB/LOCATION	SAMPLE SIZE	RANGE (PPM)	MEDIAN (PPM)
PLANT C:			
PLANT OPERATOR	4	0.1 – 0.4	0.3
LAB. TECHNICIAN	2	0.3 – 0.5	0.4
LOADING CREWMAN	1	1.2	1.2
LOADING CREWMAN	1	6.3	6.3

RESULTS ARE 8-HR. SAMPLES UNLESS INDICATED.

ETHYLENE DIBROMIDE

	TWA	CEILING
FEDERAL STANDARD	20 PPM	—
NIOSH CRITERIA DOC.	—	0.13 PPM
1978 T.L.V.	-(20) * PPM	

* SUSPECT CARCINOGEN AWAITING REASSIGNMENT

TABLE 4.1

ETHYLENE DIBROMIDE SAMPLING DATA SUMMARY
8 HR. TWA PERSONAL SAMPLE RESULTS
EDB MANUFACTURING

PLANT / JOB	SAMPLE SIZE	RANGE (PPB)	MEDIAN (PPB)
PLANT A:			
CONTROL ROOM OPERATOR	4	20 - 140	80
SURVEILLANCE TECHNICIAN	8	N.D. - 1600	370
LAB. TECHNICIAN	4	N.D. - 570	140
BRINE FIELD TECHNICIAN	4	N.D. - 30	10
PLANT B:			
CONTROL ROOM OPERATOR	7	3 - 160	40
CREW LEADER	2	40 - 950	495
PRODUCT LOADER	4	50 - 620	360
LABORATORY TECHNICIAN	2	10 - 80	45

N.D. - NOT DETECTED BASED ON SAMPLING AND ANALYTICAL METHOD.

TABLE 4.2

ETHYLENE DIBROMIDE SAMPLING DATA SUMMARY
8 HR. TWA PERSONAL SAMPLE RESULTS
EDB BLENDING OPERATIONS

PLANT / JOB	SAMPLE SIZE	RANGE (PPB)	MEDIAN (PPB)
PLANT C:			
BLEND OPERATOR	5	4 - 58	22
LAB. TECHNICIAN	6	0.2 - 12	4
SHIFT SUPERINTENDANT	3	0.1 - 0.4	0.2
PLANT D:			
BLEND OPERATOR	3	1 - 9	6
RELIEF OPERATOR	2	0.5 - 7	4
REACTOR OPERATOR	2	1 - 3	2
DRUM LOADER	4	8 - 18	14
DRUM PROCESSING	3	12 - 36	16
RAW MATERIAL HANDLER	2	27 - 82	54
LAB. TECHNICIAN	4	0.1 - 0.5	0.4
COMPOUND BULK OPERATOR	2	1 - 8	4

N.D. - NOT DETECTED BASED ON SAMPLING AND ANALYTICAL METHOD.

TABLE 5

EDB CEILING LEVEL PERSONAL SAMPLING

(SAMPLE TIME VARIES 1 TO 20 MIN.)

TASK/PLANT	SAMPLE SIZE	RANGE (PPM)	MEDIAM (PPM)
QUALITY CONTROL SAMPLING:			
PLANT A	3	5.3 - 23.4	12.0
PLANT B	2	0.3 - 0.5	0.4
PLANT C	1	1.5	1.5
PLANT D	2	0.04 - 0.7	0.4
LOADING TANK CAR:			
PLANT D	1	0.1	0.1
UNLOADING TANK CAR:			
PLANT D	1	1.6	1.6

NIOSH

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The papers included in these Proceedings were printed as they were submitted to this office.

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