

Industrial exposure to 1,3-butadiene in monomer, polymer and end-user industries

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Summary. Researchers from the National Institute for Occupational Safety and Health (NIOSH) of the Centers for Disease Control and Prevention conducted an extent-of-exposure study of 1,3-butadiene monomer, polymer and end-user industries to assess occupational exposure to butadiene and to evaluate control technologies. The findings of the exposure assessment are reported here. Walk-through surveys were conducted in 11 monomer, 17 polymer and two end-user plants; in-depth industrial hygiene surveys were conducted at four monomer, five polymer and two end-user plants. Airborne exposure concentrations of butadiene were determined for various job categories by personal sampling. The samples were analysed by a new method developed at NIOSH that is sensitive to 0.2 µg per sample. A total of 687 personal (full-shift and short-term) and 232 area samples were taken. The results indicate that all exposures were well below the current permissible exposure limit of the US Occupational Safety and Health Administration of 1000 ppm. The US Occupational Safety and Health Administration (1990) has proposed a new standard that would reduce exposure to 2 ppm. Exposures ranged from less than 0.005 ppm to 374 ppm, and 3.7% of the samples contained more than 10 ppm, 7.8% more than 2 ppm but less than 10 ppm and 88.5% less than 2 ppm. We recommend means for reducing exposure by the use of engineering controls.

Introduction

In the USA, all of the 2500 million pounds (1135 million kg) of 1,3-butadiene produced is as a co-product of ethylene manufacture. Styrene-butadiene rubber and latex and polybutadiene rubber production are the two largest uses of butadiene in the USA, accounting for approximately 1600 million pounds (726 million kg), primarily for use in the tyre industry; polychloroprene (neoprene) rubber production ranks third, with 200 million pounds (91 million kg) (Anon., 1986).

The National Institute for Occupational Safety and Health (NIOSH) estimated that 9500 workers in the USA are potentially exposed to butadiene (D.S. Sundin, unpublished data). Recent studies of exposure of rats (P.E. Owen, unpublished data) and mice (Powers, 1983) to butadiene by inhalation showed that a carcinogenic response was induced at multiple sites in response to levels of exposure below the permissible exposure limit of the US Occupational Safety and Health Administration, 1000 ppm. On the basis of the findings of the experimental studies and owing to deficiencies in the available data on exposure levels, NIOSH, through an Interagency Agreement with the US Environmental Protection Agency Office of Toxic Substances, conducted an 'extent-of-exposure' study of the butadiene industry. Between 1984 and 1987, NIOSH surveyed 30 butadiene monomer, polymer and end-user plants (Fajen *et al.*, 1990). The data generated from the study were to be used by the US Occupational Safety and Health Administration in developing a new health standard for butadiene. The project included development of a new analytical method, determination of occupational exposure to butadiene, and documentation of effective control techniques and personal protective equipment. This report presents data on extent-of-exposure from the industrial hygiene evaluation and on the engineering control programmes of the monomer, polymer and end-user industries.

Study design

The exposure assessment reported here was done in two phases and involved detailed evaluations of the three industries. The first phase involved walk-through surveys at 11 monomer production plants, 17 polymer plants and two end-user plants. The surveys were undertaken to define production methods, work practices, numbers of workers potentially exposed, personnel records and engineering controls.

At the time of our study (1984-87), 11 companies in the USA were producing butadiene monomer at 16 locations; walk-through industrial hygiene surveys were conducted for all 11 producers. In order to study the polymer industry, it was first necessary to identify the different kinds of butadiene polymers and products produced. We identified 24 polymers and products containing butadiene and randomly selected 17 representative production facilities for walk-through surveys. A bulk sample of each of the 24 polymer products was collected and subjected to head-space analysis to determine the release of butadiene monomer at temperatures typical of various fabrication processes used in the manufacture of the finished products containing the polymer. The data generated from the evaluation enabled us to select plants within the end-user industry.

Two plants of the end-user industry were randomly selected from among hundreds of potential candidates. Because styrene-butadiene rubber and polybutadiene rubber were

detected in trace amounts in the bulk samples and represent the two largest uses of butadiene, a rubber tyre plant and an industrial hose plant were selected. Walk-through and in-depth surveys were combined in view of the limited potential for exposure to butadiene in the end-user industry.

The second phase of the study was in-depth industrial hygiene surveys. This phase was similar to the first, except that air samples were collected and engineering controls were evaluated. In-depth surveys were conducted at four monomer plants, five polymer plants and the two end-user plants. These facilities were chosen on the basis of a matrix that was a representation of the monomer, polymer and end-user group as a whole.

Air sampling and analytical method

The major limitation at the start of the exposure assessment was the sensitivity and selectivity of the existing analytical method for butadiene (NIOSH Method S91; Taylor, 1977). In order to resolve this problem, a new method (NIOSH Method 1024) was developed, which is sensitive to 0.2 µg per sample or 0.005 ppm for 25-L samples (Lunsford *et al.*, 1987).

During the in-depth surveys, both personal and area samples were taken. The samples were collected with SKC Model 224 and Gillian Model HFS-113A-UT portable low-flow air-sampling pumps on tandem coconut-shell charcoal tubes at a flow rate of 0.05–0.5 L/min. The forward tube contained 400 mg of coconut charcoal and acted as the primary collection medium. The back-up tube contained 200 mg of coconut charcoal and was used to quantify the level of breakthrough. The charcoal tubes were connected to the pumps with plastic Tygon® tubing. Sample air volumes were limited to a minimum of 1 L and a maximum of 25 L. The samples were desorbed in dichloromethane and analysed by means of high-resolution gas chromatography using a back-flushable pre-column and an aluminium oxide fused-silica capillary analytical column and flame-ionization detection.

In order to ensure the quality of the results, blank samples and quality control spikes were generated, analysed and reported, in accordance with NIOSH quality assurance and quality control procedures (US National Institute for Occupational Safety and Health, 1984). Field samples were refrigerated during shipment and storage and were found to remain stable for at least 21 days when kept at – 4 °C.

Industrial hygiene sampling strategy

The 11 in-depth surveys were planned and scheduled so as to obtain personal and area air samples under normal production conditions. The sites for the in-depth industrial hygiene surveys were selected as a representative subset of the butadiene industry for use in characterizing exposures by job title. The final selection was based on a matrix developed from the 28 walk-through surveys of the monomer and polymer industries.

Exposure concentrations of butadiene were determined for each potentially exposed job category by personal sampling for at least one worker over a full shift. During each in-depth survey, three shifts were monitored over the course of three work days. Auxiliary jobs in the production area, in which exposure to butadiene was intermittent, were also monitored. Non-production jobs, involving maintenance and laboratory workers, were also evaluated to determine potential exposure. Area samples were collected to determine the butadiene

concentrations in work place air in the general process area. Short-term samples (1–90 min) were obtained in relation to jobs and tasks in order to evaluate peak exposure during performance of the task. Such short-term samples were taken during quality control sampling, cylinder voiding or whenever a job required a worker to open a butadiene line (i.e., maintenance). Samples in which butadiene was reported to be undetectable were given values, for statistical calculations, corresponding to one-half the limit of detection.

Results and discussion

Monomer industry

A total of 111 personal samples (comprising 88 full-shift and 23 short-term samples) and 100 area samples (comprising 97 full-shift and three short-term samples) were collected during the four in-depth surveys of the monomer industry. Seven job descriptions were monitored: four involved tasks that required workers to spend the majority of their time in the process area, where the production, handling and storage of butadiene take place; two of the titles describe jobs performed in the quality control laboratories at each plant. In addition to personal sampling of employees with specific job responsibilities, five work areas and the general ambient air at the perimeter of the process were monitored for concentrations of butadiene.

The butadiene concentrations in personal full-shift air samples are presented in Table 1 by job title. Table 2 presents similar information for the short-term personal samples (< 90 min). The concentrations in area full-shift air samples are presented in Table 3.

Table 1. Butadiene concentrations in personal full-shift air samples in the monomer industry

Job	Butadiene concentration (ppm)					
	No. of samples	Range	Arithmetic		Geometric	
			Mean	SD	Mean	SD
Laboratory technician	29	0.03–6.31	1.06	1.61	0.40	4.36
bomb voiding	3	0.42–374	126	215	7.43	33.60
Process technician						
control	10	< 0.02–1.87	0.45	0.72	0.09	7.93
loading	12	0.08–124	11.60	35.30	1.00	7.82
production	29	< 0.06–34.9	2.16	6.38	0.60	4.46
storage	5	< 0.05–1.53	0.44	0.62	0.21	4.26
Total	88	< 0.02–374	7.00	41.8	0.47	6.49

A review of personal exposures by job title (Table 1) suggested that those jobs that require workers to handle or transport containers of butadiene present the greatest potential exposure. Laboratory technicians who void sample cylinders, process technicians who load or unload tank trucks or rail cars and technicians in process areas had arithmetic mean exposures of 126, 11.6 and 2.2 ppm and geometric mean exposures of 7.43, 1.00 and

0.60 ppm, respectively. All other job titles resulted in arithmetic mean exposures of less than 1.06 ppm and geometric mean exposures of less than 0.5 ppm. Maximal exposures for two job titles exceeded 100 ppm: 8-h time-weighted average values of 124 ppm for a process technician in the loading area and 374 ppm for a laboratory technician. These two exposures were associated with poor work practices or uncontrolled emissions. In both cases, there was a poor connection of threaded fittings which permitted escape of butadiene into the work environment.

Table 2 shows that exposure can exceed 10 ppm for short periods during all three types of periodic activities in the plants: cylinder sampling, cylinder voiding and maintenance. Short-term monitoring is used to evaluate peak exposures during a job with a definite potential exposure. The highest short-term butadiene concentrations were associated with open-loop sampling (147 ppm) and cylinder voiding (108 ppm).

A review of the results for 97 area samples in Table 3 indicates that arithmetic mean exposures in rail-car terminals and tank storage farms were 10.5 ppm and 7.8 ppm and the

Table 2. Butadiene concentrations in personal short-term air samples in the monomer industry

Job	Butadiene concentration (ppm)					
	No. of samples	Range	Arithmetic		Geometric	
			Mean	SD	Mean	SD
Bomb sampling	8	< 0.7–147	21.5	50.7	3.41	6.86
Bomb voiding	6	< 0.1–108	22.9	42.8	2.25	18.3
Maintenance	7	0.05–16.8	5.17	6.83	0.99	11.6
Process	2	< 0.3–0.36	0.26	0.14	0.25	1.72
Total	23	< 0.05–147	15.1	36.5	1.67	10.2

Table 3. Butadiene concentrations in area full-shift air samples in the monomer industry

Job	Butadiene concentration (ppm)					
	No. of samples	Range	Arithmetic		Geometric	
			Mean	SD	Mean	SD
Production	25	< 0.04–2.13	0.57	0.57	0.34	3.13
Railcar, loading	20	0.09–64.3	10.5	18.4	1.85	8.56
Semi-trailer, loading	4	0.10–1.92	0.57	0.90	0.24	4.01
Laboratory	18	0.03–5.88	1.04	1.92	0.22	5.82
Tank farm	5	0.15–23.8	7.80	10.4	2.12	8.36
Subtotal	72	0.03–64.3	3.94	10.9	0.54	6.73
Perimeter	25	< 0.02–0.50	0.08	0.12	0.04	2.97
Total	97	< 0.02–64.3	2.94	9.49	0.28	7.95

geometric mean concentrations, 1.85 ppm and 2.12 ppm, respectively. The arithmetic mean concentrations in other work areas were less than 1.04 ppm and the geometric means less than 0.4 ppm. In no full-shift area sample did the concentration exceed 70 ppm. In the 25 samples taken at locations on the plant perimeter, the arithmetic mean was 0.08 ppm and the geometric mean was 0.04 ppm.

For the monomer industry as a whole, 7.1% (15/211) of the samples contained levels greater than 10 ppm, 12.8% (27/211) contained 2–10 ppm, 12.3% (26/211) contained 1–2 ppm, and the remaining 67.8% (143/211) had less than 1 ppm. The four monomer plants studied were built in 1944, 1951, 1975 and 1981. The arithmetic mean concentration of butadiene in these four plants was 5.9 ppm and the geometric mean, 0.42 ppm.

Polymer industry

During the five in-depth surveys of butadiene polymer facilities, 452 personal samples (comprising 438 full-shift and 14 short-term samples) and 132 area air samples were collected. Tables 4 and 5 provide a breakdown by job category or work activity of the results of full-shift and short-term personal monitoring, respectively. Full-shift exposures in the different job categories ranged from undetected (< 0.005 ppm) to a high of 42.9 ppm (Table 4), whereas the short-term exposures range from undetected (< 0.1 ppm) to a high of 280 ppm (Table 5).

Table 4. Butadiene concentrations in personal full-shift air samples in the polymer industry

Job	Butadiene concentration (ppm)					
	No. of samples	Range	Arithmetic		Geometric	
			Mean	SD	Mean	SD
Laboratory technician	49	< 0.006–37.9	3.09	6.91	0.332	12.1
Tank farm operator	23	0.009–24.0	1.97	5.01	0.260	9.59
Front end (reaction)	108	< 0.006–24.7	1.80	4.02	0.151	12.1
Maintenance technician	42	0.012–42.9	1.84	6.85	0.144	7.44
Back end (finishing)	79	< 0.005–7.12	0.351	1.07	0.037	7.13
Other	137	< 0.005–0.167	0.035	0.032	0.022	3.03
Total	438	< 0.005–42.9	1.14	4.02	0.072	9.27

Table 5. Butadiene concentrations in personal short-term air samples in the polymer industry

Job	Butadiene concentration (ppm)					
	No. of samples	Range	Arithmetic		Geometric	
			Mean	SD	Mean	SD
Quality control sampling	10	< 0.1–280	48.7	86.4	9.37	11.8
Maintenance	4	0.087–14.4	4.50	6.77	1.05	9.95
Total	14	0.087–280	36.1	74.9	5.02	12.8

The highest full-shift personal exposure was 42.9 ppm for a maintenance technician working on a butadiene compressor. The highest short-term exposure was 280 ppm for a process technician (unloading area) sampling a barge for butadiene. The short-term personal monitoring was conducted with the intention of identifying peak exposures during operations or activities that were considered to involve potential exposure to butadiene. The results given in Table 5 show at least one short-term exposure to butadiene that was greater than 10 ppm in both periodic job categories in the plant.

Table 4 clearly shows the four job categories in which full-shift (personal) exposures to butadiene were greater than 10 ppm in at least one sample. Those were laboratory technicians, process technicians in tank farm and front end (purification, polymerization and reaction) areas and maintenance technicians. Geometric mean exposures for all other job categories were less than 0.1 ppm. Maximal full-shift exposures for laboratory and maintenance technicians exceeded 35 ppm in at least one sample.

A total of 132 area samples were obtained during the five in-depth industrial hygiene surveys of the polymer industry. Table 6 provides a breakdown by work environment of the results of full-shift area monitoring. The butadiene concentrations in the work areas ranged from undetected (< 0.006 ppm) to 9.01 ppm. The maximal concentration of 9.01 ppm was observed in a quality control laboratory near a gas chromatograph and was caused by a faulty threaded connection of the cylinder to the gas chromatograph. The cylinder was not contained in a ventilated exhaust hood. In the 51 samples taken at locations near the plant perimeter, an arithmetic mean of 0.03 ppm and a geometric mean of 0.01 ppm were calculated.

Table 6. Butadiene concentrations in area full-shift air samples in the polymer industry

Job	Butadiene concentration (ppm)					
	No. of samples	Range	Arithmetic		Geometric	
			Mean	SD	Mean	SD
Barge	1	0.474–0.474	0.474	–	0.474	–
Tank farm	8	0.077–1.75	0.668	0.670	0.382	3.32
Laboratory	3	0.025–9.01	3.02	5.18	0.197	27.5
Front end (reaction)	50	< 0.006–2.82	0.285	0.486	0.090	5.40
Back end (finishing)	19	< 0.006–0.026	0.009	0.008	0.006	2.18
Subtotal	81	< 0.006–9.01	0.362	1.08	0.058	7.45
Perimeter	51	< 0.006–0.170	0.027	0.036	0.013	3.37
Total	132	< 0.006–9.01	0.232	0.859	0.033	6.58

For the polymer industry as a whole, 3.3% (19/584) of the samples indicated exposure levels greater than or equal to 10 ppm, 7.7% (45/584) contained butadiene at 2–10 ppm, 3.3% (19/584) contained 1–2 ppm and the remaining 85.8% (501/584), less than 1 ppm. Of the five polymer plants in the study, one was built in 1946 and the remainder in 1943. The arithmetic mean for personal full-shift exposures (n = 438) in the five polymer plants was 1.14 ppm; the geometric mean was 0.072 ppm.

End-user industry

A rubber tyre plant and an industrial hose plant were selected to represent the end-user industry. The plants used styrene-butadiene, polybutadiene and acrylonitrile-butadiene rubber. A total of 124 personal samples were collected over three shifts, with 34 in the hose plant and 90 in the tyre factory. The concentrations of butadiene were below the limit of detection in all samples.

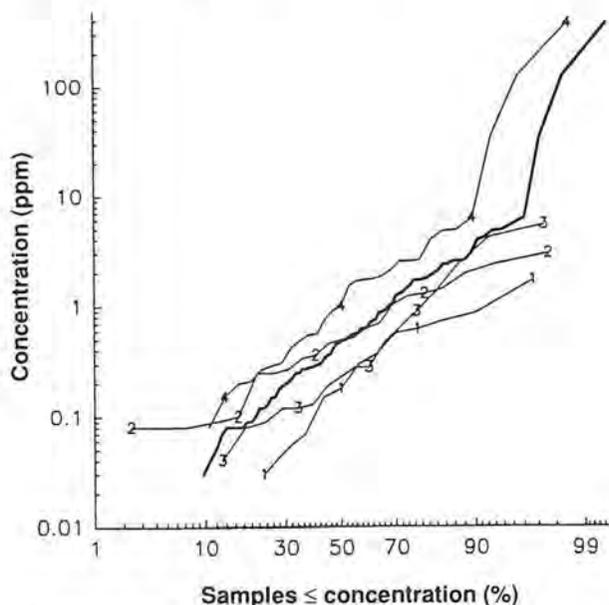
Conclusions

The butadiene monomer and polymer processes are highly automated, and operators do not routinely spend much time in the process area. Few opportunities are therefore available throughout most of the process for occupational exposures to butadiene of greater than 10 ppm. Three categories of ancillary operations associated with the process, however, result in potential exposure to butadiene: decontamination and maintenance of process equipment; sampling and analysis of samples for quality control; and loading and unloading of crude feed and butadiene product. Five distinct job categories have been identified in these operations in which there is potential occupational exposure to butadiene: in loading (0.08–124 ppm); tank farm (0.009–24.0 ppm); process, i.e., purification, polymerization and reaction (< 0.005–34.9 ppm); and laboratory (< 0.006–374 ppm) areas. Exposures of maintenance technicians ranged from 0.012 to 42.9 ppm.

The data on exposure show a number of relatively high levels resulting from either poor work practices, ineffective controls or failure of the control system. They show that properly implemented and maintained up-to-date controls would reduce the probability of such excursions; inclusion of these high numbers results in an upward bias of the estimated feasibility of control. Figures 1 and 2 show the percentile distributions of the concentrations of butadiene in full-shift personal air samples taken at individual plant sites and composite percentile distributions for the monomer and polymer industries. Figure 1 shows that in three of the four monomer plants there was no exposure above 10 ppm. The high exposures in the polymer industry (Figure 2) were slightly higher than those in the monomer industry, but overall exposures were lower: The geometric mean concentrations of butadiene were 0.42 ppm in the monomer industry and 0.072 ppm in the polymer industry. At the polymer plant with the highest exposures, plant 5, approximately 90% of the exposures were below 7 ppm. In Figure 1, two samples from plant 4 were associated with poor work practices or uncontrolled emissions. The arithmetic mean concentration of butadiene in the 88 personal full-shift air samples taken in the monomer industry is 7.0 ppm and the geometric mean, 0.47 ppm. When the outlying results for plant 4 are removed, the arithmetic mean is reduced to 1.4 ppm and the geometric mean to 0.41 ppm. Exclusion of the outliers is appropriate in evaluating properly functioning engineering controls.

In summary, the results of this exposure assessment in the butadiene industry (monomer and polymer) show that full-shift exposures in all job categories are below 10 ppm. Of the 526 personal full-shift air samples collected during the study, 2.7% (14) contained more than 10 ppm, 10.5% (55) more than 2 ppm but less than 10 ppm and 86.5% (457) less than 2 ppm. For all job categories, the geometric mean concentration for a full-shift exposure was 0.098 ppm and the arithmetic mean was 2.12 ppm.

Figure 1. Percentile distribution of concentrations of butadiene in personal full-shift samples taken at four monomer plants



Concentration is given on a log scale, and the fraction of samples containing that concentration or less is given on a probability scale. Distributions for individual plants are identified by number. The bold line depicts the combined distribution for all four plants.

The results obtained in the end-user industry document, on a very limited basis, that workers in this industry do not currently have measurable exposure to butadiene. The results were expected on the basis of information obtained by head-space analysis of the bulk polymer samples.

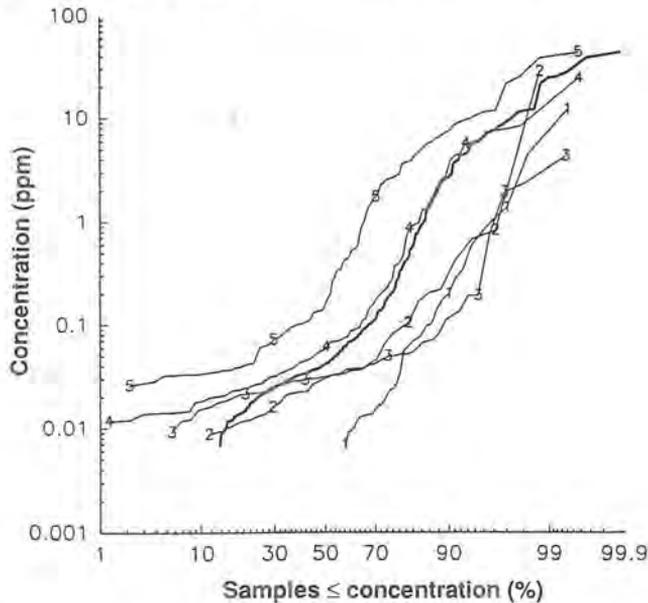
The engineering controls and work practices that have been developed are effective in minimizing exposures of personnel if they are adhered to. Short-term exposure will occur if the fittings on a closed-loop system are worn or improperly connected. The data thus show that personal full-shift exposures in all job categories can be maintained below 10 ppm by application of effective engineering controls. Additional studies would have to be conducted to determine the feasibility of achieving lower concentrations of butadiene (e.g., 1 ppm) in all job categories in the industry.

The new NIOSH Analytical Method 1024 is the preferred method for use. The enhanced sensitivity provided by high-resolution gas chromatography should enable detection down to 0.005 ppm in a 25-L sample.

Recommendations

In order to reduce potential exposure to butadiene, engineering controls and work practices will have to be modified. As exposures do occur in certain jobs and because of concern that butadiene may present both carcinogenic and teratogenic risks, the following

Figure 2. Percentile distribution of concentrations of butadiene in personal full-shift samples taken at five polymer plants



Concentration is given on a log scale, and the fraction of samples containing that concentration or less is given on a probability scale. Distributions for individual plants are identified by number. The bold line depicts the combined distribution for all four plants.

additional control measures are recommended for use in production plants where such controls are not already being implemented.

1. Plants should consider converting to a closed-loop sampling system for obtaining cylinder samples for quality control, in order to lower the mean exposures of laboratory technicians and technicians working in process areas.
2. Leaking pumps can result in exposure of technicians in process areas. The release of butadiene from such equipment can be controlled by the use of dual mechanical seals. Plants should consider retro-fitting pumps that have single mechanical seals with the more effective dual mechanical seals.
3. Because magnetic gauges are known to limit the release of butadiene (and hence exposure of process technicians in the loading area) while rail cars are being loaded, plants should consider a programme to convert to 100% magnetic gauges for monitoring rail-car filling operations.
4. As is evident from the monitoring results for laboratory technicians involved in voiding cylinders, workers assigned to this task may be exposed to butadiene.

Consideration should be given to using a laboratory hood or a vacuum exhaust with an enclosure for cylinder voiding. Furthermore, workers should be trained in the proper conduct of tasks such as cylinder voiding and cylinder sampling.

5. The new NIOSH sampling and analytical method for butadiene is recommended in areas of potentially low exposure and where there is potential interference from other C₄ compounds.

The results of this NIOSH monitoring study demonstrate that use of analytical methods specific to butadiene is preferable for assessing exposures to butadiene.

Acknowledgements

The work was supported by an Interagency Agreement with the US Environmental Protection Agency (DW75930917-01-2). Without collaboration with the US Environmental Protection Agency with respect to study design and critical review, this study would not have been possible.

We also are grateful to the employees and management of those companies that participated in the study. We specifically acknowledge the diligent efforts of Yvonne Gagnon and John Palassis of the Division of Physical Sciences and Engineering at NIOSH for developing a specific analytical method for butadiene.

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WORLD HEALTH ORGANIZATION



INTERNATIONAL AGENCY FOR RESEARCH ON CANCER

Butadiene and Styrene: Assessment of Health Hazards

Edited by

M. Sorsa, K. Peltonen, H. Vainio and K. Hemminki

IARC Scientific Publications No. 127

International Agency for Research on Cancer
Lyon
1993

Published by the International Agency for Research on Cancer,
150 cours Albert Thomas, 69372 Lyon Cedex 08, France

© International Agency for Research on Cancer, 1993

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Distributed in the USA by Oxford University Press, New York

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ISBN 92 832 2127 3
ISSN 0300-5085

Printed in the United Kingdom