Paraquat Exposure of Knapsack Spray Operators on Banana Plantations in Costa Rica

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A study of occupational exposure to paraquat was performed among 11 knapsack spray operators at banana plantations in Costa Rica. External and internal exposures were quantified and determinants of exposure identified by measurements, observations, and interviews. Dermal exposure was measured with skin pads, respiratory exposure by personal air sampling, and internal exposure by urine sampling. The wrists, back, and legs were the areas with the highest levels of dermal exposure. Respiratory exposures appeared to be strongly influenced by differences between days, while dermal exposures varied mostly due to differences between plantations. The use of protective clothing did not effectively protect against dermal exposures. Both respiratory and dermal exposures were significantly related to internal exposures, and both should be considered possible routes for systemic absorption of paraquat. It cannot be excluded that measurable levels of exposure can lead to acute as well as chronic health effects. Furthermore, due to poor conditions within the working environment, the spray operators are continuously at risk for high exposures that could lead to severe intoxication, and therefore a strategy for control of exposure is necessary. *Key words:* dermal exposure; respiratory exposure; biological monitoring; knapsack spray operators; herbicides; paraquat.

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Paraquat (1,1'dimethyl-4,4'-bipyridinium dichloride) is used as a contact herbicide and is commonly sold as Gramoxone, a solution containing 20% of the paraquat ion. It is used intensively on banana plantations, as well as elsewhere, in Costa Rica, where it is a frequent cause of occupational accidents, including topical injuries as well as systemic poisonings leading to hospitalization and death.[1] Banana companies, which employ 50,000 people, represent a major part of the export industry of Costa Rica.[2,3] Depending on the plantation's size, 40 to 200 people work on each plantation, and three to eight of those people apply paraquat by means of spraying it from knapsack containers.

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Exposure to paraquat may cause systemic poisoning characterized by dysfunction of the liver, kidney, myocardium, central nervous system, and lungs, with progressive respiratory failure ultimately leading to death.[4] Topical effects include inflammation and burns of skin and eyes, nail damage, and epistaxis.[5,6] Whether chronic exposure to low levels of paraquat results in adverse health effects is controversial.[7–9]

Paraquat can enter the body by means of ingestion, inhalation, and dermal contact. Oral exposure may occur when sucking or blowing blocked spray nozzles, by eating food that has been in contact with contaminated hands, or by ingestion of inhaled spray droplets. Paraquat is poorly absorbed through the intact skin, but when the skin is damaged paraquat can enter the body more easily, and may cause systemic intoxication.[10] Occupational systemic intoxication has been reported, caused by oral intake of small amounts and dermal exposure to concentrated or diluted paraquat solution.[1,11–15] Respiratory exposure to paraquat is not thought to contribute significantly to the internal exposure.[16] However, some studies report systemic paraquat poisoning possibly due to respiratory exposure.[14,15]

Several studies of plantation workers' exposures to paraquat in developing countries such as Malaysia and Sri Lanka have been performed. In these studies, dermal, respiratory, and internal exposures have been assessed, but the relationship between external and internal exposures has not been discussed. The authors of

Scooping diluted paraquat solution from the 200-liter tank.

Measuring the 20% concentrated paraquat solution.

these studies concluded that measurable levels of exposure to paraquat do not have adverse health effects.[17–20]

There is a seeming discrepancy between the occurrence of systemic poisonings and fatalities in people using paraquat in occupational settings and the conclusion that occupational exposure to paraquat does not affect health. More research is needed to clarify the risks of occupational paraquat exposure, and therefore we performed a study on knapsack spray operators' exposures to paraquat at banana plantations to: (1) quantify dermal and respiratory exposures to paraquat; (2) identify determinants of exposure routes in order to be able to control exposures; (3) evaluate urinary levels of paraquat; and (4) study the relationships between external and internal exposures.

MATERIALS AND METHODS

Study Population and Measurement Strategy

Eleven paraquat spray operators working at four banana plantations (plantations A, B, C, and D) situated in the Atlantic zone of Costa Rica were studied. At each plantation, dermal, respiratory, and urinary paraquat were measured in two or three operators. The operators were also observed during their work and interviewed.

At each plantation, paraquat measurements were performed during one week. Paraquat levels were measured throughout each operator's working day of paraquat application, on Monday, Wednesday, and Friday at plantations A, B, and C, and on Tuesday and Wednesday at plantation D. At plantations A and C, one worker was not available for paraquat measurement on the second sampling day, because he performed other tasks on that day.

Skin pads of α-cellulose (Schleicher and Schuell, diameter 3.5 cm) were used to determine dermal exposures to paraquat. The pads were placed between two circular Fixomul stretch bandage plasters (BDF), 5 cm, with a circular opening 2.5 cm in diameter in the upper plaster layer.[21] The pads were located directly on the skin in body areas subject to high levels of exposure[17,18,22] and/or where high permeability of the skin to paraquat was expected.[23] These locations were the middle of the forehead, the nape of the neck, the middle of the back, the inner sides of the right and left wrists, the front of the left upper leg, and the outer side of the left ankle. Paraquat in the breathing zone was measured by collecting inhalable aerosols by means of a Gilair pump (2 L/min) on a PTFE filter mounted in a PAS-6 sampler according to a personal-air-sampling method.[24,25]

To determine internal exposure to paraquat, the operator's urine was collected just before and at the end of the working day on the same day as external exposure measurements were carried out. The operators were asked to wash their hands before urinating to prevent contamination of the samples. Urine samples were taken before work started in order to detect possible paraquat residues due to exposure during previous working days. Because clearance of small amounts of paraquat was thought to be rapid (80% within three hours according to a study in dogs[26]), the concentrations of paraquat measured in the urine samples collected after work were assumed to reflect external exposures during the working day. The concentration of paraquat found in urine was adjusted for creatinine concentration.

The spray operators were interviewed to collect in-

Applying the spraying solution.

The application site is often rough.

formation about work practices, personal hygiene during work, the use of protective clothing, and the occurrence of paraquat-exposure-related health complaints. Activities associated with high risks of paraquat exposure were defined using the outcome of the questionnaires and by observing the operators during work.

Extraction and Analysis

The α-cellulose filters were taken out of the skin pads using a punch (22 mm) and dissolved in 5 ml of deionized water in a centrifuge tube. After 5 minutes of ultrasonification, the filter was taken out of the solution. The Teflon filters were extracted according to NIOSH-method 5003.[27] Urine was extracted according to an ion-pair extraction method.[28]

After extraction, the solutions were stored at −20 °C until further analysis. Before analysis, the solutions were thawed and 3-mL volumes were pipetted in a centrifuge tube. To reduce the paraquat, 0.5 mL sodium dithionite (Merck, Darmstadt) was added. Absorption was measured by spectrophotometry (uv/vis spectrophotometer lambda 28, Perkin Elmer), wavelength between 350 and 450 nm, measuring the second derivative.[28]

Estimation of Total Dermal Exposure

A measure for total dermal exposure was developed to relate dermal exposures of different spray operators to their internal exposures to paraquat. The paraquat concentration on a skin pad was thought to be representative for the exposure of a certain body area.[29] The representative areas were based on the anatomic model suggested by Popendorf and Leffingwell[30] and modified on the basis of the locations of the pads. This means that the values obtained from the pads were extrapolated to representative areas only. Total dermal exposure calculated with our measure according to equation 1 represented 42% of the total body area.

$$E_d = \Sigma_{i=1}^{n} A_i \cdot C_i \quad (1)$$

where E_d = dermal exposure of 42% of body area; A = representative area of a skin pad; C = paraquat concentration measured on the skin pad (μg/cm^2).

When one of the operators was missing, for example, a neck pad, the mean relative contribution of the neck to the total dermal body exposure of the other operators was calculated. This value was used to estimate the paraquat concentration on the missing pad.

Fumbling with the nozzle.

Data Analysis

Data were imported in SAS 6.04 for statistical analyses. Values under the limit of detection were given two-thirds of the limit of detection. First, variables were tested on lognormality and descriptive statistics were calculated. When comparing dermal and respiratory results from different spray operators and plantations, values were adjusted for amount of paraquat sprayed per operator, in order to exclude differences in the amounts of paraquat applied as a factor explaining differences in average exposures between operators and plantations. Analyses of variance (ANOVAs) of total dermal exposures and total inhalational exposures were performed by using a two-way ANOVA random-effects model, resulting in estimates of geometric standard deviations (GSDs) and variance ratios (R95s) between companies, workers, and days. The variance ratio is the quotient of the 97.5th and 2.5th percentiles of the log-normally distributed mean exposures, and provides information regarding the ranges of measured exposures.[31,32] In order to find differences in dermal exposures related to the use of protective equipment, Student's two-sample *t*-test and, if values were not lognormally distributed, the Wilcoxon rank-sum test were used. Spearman's rank-correlation was calculated to estimate correlations between dermal and respiratory exposures. Relationships between external and internal exposures were studied by using the Wilcoxon rank-sum test, after internal exposures were divided into two groups based on paraquat-positive and -negative urine samples.

RESULTS

Observations and Questionnaires

Banana plantations. The four banana plantations differed in working circumstances and work practices (Table 1). At plantation B, a more concentrated spray solution was used but less paraquat was applied per hour. At plantations A, B, and C, hardly any protective equipment was used. During work, knapsack spray operators wore rubber boots, long socks, shorts or long trousers, and a singlet or a long-sleeved shirt, and some operators wore headgear. At plantation D, protective equipment was provided and used. This included rubber boots, a vinyl apron to protect the back, a cotton overall, gloves, a face mask, and headgear.

Work practices. The application of paraquat entailed several activities. First, a 20% paraquat solution was diluted with water into a spray solution of 0.1 to 0.2% paraquat and a surface-active agent was added. The solution was made in an open 200-liter tank, hitched on a rail, which was pushed to the place of application. During transport, the solution frequently splashed out of the tank, and on one occasion during the measurement

Spilled paraquat foam after filling the knapsack tank.

period three tanks slipped off the rail after crashing with bananas, which were transported on the same rail. At the place of application, the solution was scooped from the tank into the knapsack tank, using an open jerry-can. During this activity contact of the hands with the solution occurred. While the knapsack tank was being filled, foam frequently spilled over the edges of the tank.

The paraquat solution was applied by right-hand pumping and left-hand spraying. Spray operators sometimes sprayed uphill or walked through high weeds. It was observed and reported in response to the questionnaire that the spraying equipment leaked occasionally. The application sites were often uneven and muddy. Some operators rushed through the fields and sometimes fell while spraying. The operators often fumbled with the equipment with their bare hands to repair small defects. Occasionally, blockage in the spray nozzle was cleared by blowing it out. In their responses to the questionnaire, seven workers reported eating, drinking, or smoking during working time without washing their hands or biting their nails during work. Most of the operators did not shower immediately after work (on average 1.5 hours later). Four operators asked to be informed about how to use paraquat, and eight said that they would like to have more information.

Health complaints. Of the 11 paraquat spray operators under study, seven reported having had one or more health problems in the preceding 12 months that were thought to have been related to paraquat exposure. Three reported skin problems involving blistering and burns of the hands, thighs, back, testicles, and legs. The

TABLE 1 External exposures of paraquat spray operators and plantation spraying conditions

Paraquat Exposure	% Paraquat in Spray*	Amount of Spray† (L/hr)	Spray Solution Made by Operator	Protective Clothing Used	No. of Samples	Arithmetic Mean	Geometric Mean	Geometric Standard Deviation	Range Made by Operator
Respiratory (μg/m³•kg)									
Plantation A	0.1	40	−	−	8	36.7	9.7	3.4	2.0–240
Plantation B	0.2	22	+	−	6	0.6	0.5	1.7	0.3–1.0
Plantation C	0.1	41	+	−	8	4.4	2.1	4.3	0.2–11.3
Plantation D	0.1	42	−	+	6	8.5	5.4	3.5	1.2–18.2
Dermal (mg/kg)									
Plantation A	0.1	40	−	−	8	41.5	27.1	2.6	7.6–113.0
Plantation B	0.2	22	+	−	6	11.3	10.9	1.4	6.3–15.5
Plantation C	0.1	41	+	−	8	6.6	6.1	1.5	3.5–11.3
Plantation D	0.1	42	−	+	6	20.4	18.2	1.6	10.4–40.6

*Percentage of paraquat (w/w) in the spray solution.
†Average amount of solution sprayed in one working hour.

TABLE 2 Descriptive statistics of respiratory and dermal exposures

	No. of Samples	Arithmetic Mean	Gemetric Mean	Geometric Standard Deviation	Range
Respiratory exposure (µg/m³)	28	1.8	0.6	3.9	0.1–24.0
Dermal exposure					
Individual pads (µg/cm²•h)					
Head	28	0.4	0.3	1.6	0.1–1.1
Neck	27	0.5	0.4	1.8	0.1–1.3
Left wrist	27	1.5	0.7	2.7	0.2–8.7
Right wrist	25	1.4	0.8	2.7	0.1–9.2
Back	26	1.1	0.7	2.7	0.1–9.6
Leg	27	1.3	0.8	2.7	0.2–6.2
Ankle	28	0.6	0.3	2.2	0.1–6.8
Total dermal exposure (mg/h)	28	0.8	0.5	2.2	0.2–5.7

injuries had been caused by defective spraying equipment or by contact of the legs with recently-sprayed weeds. Eye problems (n = 2) of redness, irritation, and burning sensations had been caused by paraquat solution splashing in the eyes while the tank was being pushed or by wind rebounding the spray solution. Nail problems were reported by three workers, and three workers had experienced epistaxis, one of them frequently. One worker mentioned an occasional burning sensation in the nose cavity. Five also reported nonspecific systemic complaints in relation to paraquat exposure, including headache, stomachache, nausea, and blurred vision. Two of these workers said that they experienced these problems every time they applied paraquat.

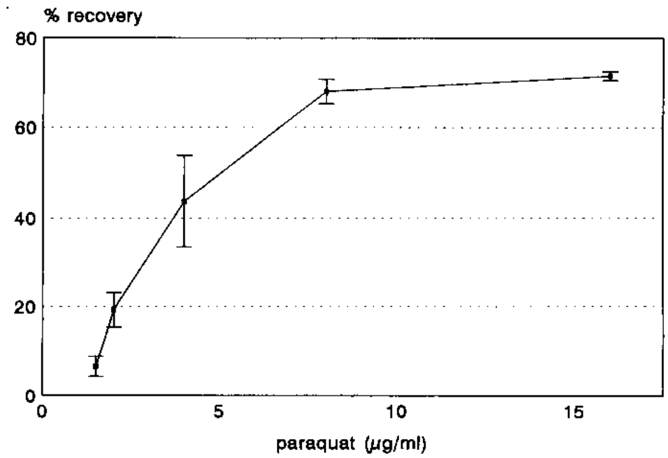

Figure 1—Recovery of paraquat from skin pad.

External and Internal Exposure Measurements

Recoveries and limits of detection. Recoveries of paraquat were between 70% and 95% from PTFE filters (tested concentrations ranged from 0.05 to 1.0 µg/mL), and 79% and 84% in urine samples (tested concentrations 0.15 and 0.30 µg/mL). Coefficients of variation of analysis (CV_a) ranged from 5% to 26% for PTFE filters and from 7% to 20% for urine. Recoveries with standard deviations (SD) for the α-cellulose pads are summarized in Figure 1. Because of the low values, all skin-pad samples were corrected for recovery using Figure 1.

Limits of detection were defined by analyzing blank α-cellulose and PTFE filters, and 12 urine samples of six unexposed men. The mean of the concentrations measured on the blanks was defined as background noise. Background noise plus three times the SD of the background noise was taken to be the limit of detection. Limits of detection for the PTFE filters, α-cellulose filters, and urine were 0.03, 1.5, and 0.03 µg/mL, respectively.

Dermal and respiratory exposures. Table 2 shows descriptive statistics of the respiratory and dermal exposure measurements. The table shows that the wrists, back, and legs were the most exposed body areas. The relative contribution of each exposed area to total dermal exposure is shown in Figure 2. The exposures of the wrists, legs, and back was almost equal and together contributed 87% of the total exposure.

Table 1 shows the results of respiratory and dermal exposure measurements summarized per company. Geometrical means of both dermal and respiratory exposures were higher at plantation A than at the other plantations (t-tests, p-values 0.02 and 0.01, respectively). At plantation B a lower respiratory, and at plantation C a lower dermal, exposure was present (t-tests, p-values < 0.001).

Table 3 shows the geometric standard deviations (GSDs) and variance ratios (R95s) of total dermal exposure (mg/kg) and total respiratory exposure (mg/m³•kg). Differences in average dermal exposures between plantations were slightly larger than those between spray operators and between days for the same operator, which were almost the same. Differences in average exposure levels were larger for respiratory than for dermal exposures. Respiratory exposure levels showed less variation between workers than between companies and from day to day. The highest value of respiratory exposure per kg was over ten times higher

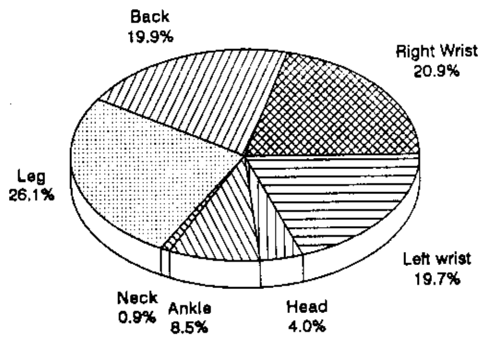

Figure 2—Relative contributions of paraquat recovered from individual skin pads to total dermal exposure.

TABLE 3 Variations in dermal and respiratory exposures between plantations, between workers, and from day to day

	Dermal Exposures		Respiratory Exposures	
	Geometric Standard Deviation	Variance Ratio (R95)	Geometric Standard Deviation	Variance Ratio (R95)
Total	2.45	33.3	5.67	898.7
Between plantations	1.82	10.5	3.00	74.6
Between workers	1.62	6.6	2.16	20.3
Between days	1.60	6.3	3.00	74.6

than the next highest value, 0.240 and 0.018 mg/m^3 • kg, respectively. Excluding this value from the analysis decreased the total variation (GSD 4.51, R95 367.3), but the relationship remained the same. Total respiratory exposure did not correlate with total dermal exposure ($r = 0.07$, $p = 0.74$) or with paraquat levels on individual pads ($r = -0.21$–0.26, $p = 0.16$–0.84), and only the left-wrist-pad value correlated almost significantly with respiratory exposure ($r = 0.36$, $p = 0.06$).

Although all the operators wore boots, remarkably high exposures of the ankles were sometimes present, which could have been the result of indirect exposure due to run-off of paraquat down the leg. Exposures of the ankles and legs were correlated ($r = 0.55$, $p = 0.002$). No difference in the exposures of wrists and legs was detected in comparing workers using gloves or overalls with workers not wearing them. Spray operators using an apron at the back all had relatively low dermal exposures on the back, but no lower exposure was found after performing a Wilcoxon rank-sum test ($p = 0.93$). Workers wearing long trousers had significantly less leg exposures (t-test, $p = 0.002$) than did workers wearing shorts or overalls. To exclude the possibility that this effect was due to an overall lower level of dermal exposure, the relative contribution of leg exposure to total skin exposure per worker was taken into account, which also turned out to be lower for workers wearing long trousers (t-test, $p = 0.001$).

Internal exposure. Paraquat was not detected in urine samples collected before work started. Paraquat was detected in two of 28 urine samples taken after work, and contained 11 and 22 μg paraquat per mmol creatinine. The positive samples were those of one of the workers of plantation A on the third sampling day (worker 1), and one of the workers of plantation D on the first sampling day (worker 9), respectively.

Relationship of external–internal exposures. Table 4 shows external and internal exposures of the two workers with detectable paraquat in their urine samples and summarizes the data of all other workers. Worker 1 had relatively high dermal exposures on all sampling days, while Worker 9 had a high dermal exposure only on the day that paraquat was detected in his urine. Both worker 1 and worker 9 had relatively high respiratory exposures on the days that paraquat was detected in urine, but the respiratory exposure of worker 1 was also high on another sampling day. Generally, a positive urine sample was associated with a higher total dermal exposure (Wilcoxon, $p = 0.04$) as well as a higher respiratory exposure (Wilcoxon, $p = 0.04$) on the same measuring day.

DISCUSSION

Limitations of the Study

This study had several limitations. First, skin-pad exposure values had to be adjusted for low recovery. This correction was justified because a linear relationship ($r = 1.0$) existed between spiked samples and their measured concentrations, and the coefficients of variation of recoveries were relatively low. Second, the numbers of knapsack spray operators and measurement days were relatively small, resulting in only two positive urine

TABLE 4 Internal and external exposures of workers 1 and 9 to paraquat

	Dermal Exposure (mg)	Respiratory Exposure* (mg)	Paraquat in Urine (μg/mmol) Creatinine
Worker 1			
Day 1	11.3	0.086	—†
Day 2	8.4	0.004	—
Day 3	9.4	0.055	11
Worker 9			
Day 1	6.9	0.035‡	22
Day 2	1.7	0.007‡	—
Other workers			
Arithmetic mean	2.4	0.007	—
Range	(1.1–5.3)	(0.001–0.032)§	—

*Respiratory ventilation of 1.8 m^{3}h is assumed, according to an O_2 uptake of 1.5 L/min and pulmonary ventilation of 20 L O_2 during manual labor.[45]
†Non-detectable after correction for creatinine level.
‡A face mask was worn.
§Twelve values were below the detectable limit.

TABLE 5 Dermal, respiratory, and internal exposures of knapsack spray operators in various studies

	Dermal Exposure	Respiratoryl Exposure (mg/h)	Internal Exposure (mg/L Urine)	Spray Dilution (w/w %)
Present study	0.2–5.7 mg/h* 3.5–113.0 mg/kg*	$(0–43) \cdot 10^{-3}$	< 0.03–0.24	0.1–0.2
Sri Lanka[17]	940–2,710 mg/kg†‡	N.A.	< 0.03	0.03–0.04
Malaysia[18]	0.3–12.4 mg/h*§ 12.1–169.9 mg/h†	$(0–5) \cdot 10^{-3}$	< 0.05–0.69	0.1–0.2
United States[20]	0.01–0.57 mg/h*§	$<1 \cdot 10^{-3}$	N.A.	0.2
Malaysia[19]	N.A.	N.A.	<0.1–0.32	0.05

*Direct dermal exposure, exposure of the (uncovered) skin.
†Potential dermal exposure, exposure of the clothing (and uncovered skin).
‡Measured by using a Tyvek coverall.
§Measured according to WHO standard protocol (1975).
N.A. = not assessed.

samples. Nevertheless, the design allowed the identification of determinants of exposure, and the relationship between external and internal exposures could be explored because the measurements were performed simultaneously.

Exposure Measurements

Table 5 compares the ranges of exposures in our study with those found in other studies. The values of our study are of the same order of magnitude as those found in previous studies. Dermal exposure compares well with those in other studies that measured direct dermal exposures, except that of Staiff et al.[20] who reported exposures more than tenfold lower. The reason for the discrepancy could not be deduced from their paper. Respiratory exposure in our study was higher, but when the highest value is not taken into account, the ranges compare well. Also, the amounts of paraquat found in urine compare well with those in the other studies, except the Sri Lankan study,[17] where paraquat was not detected in urine despite present dermal exposure. However, because external and internal exposures were not measured simultaneously in that study, dermal exposures may have been lower at the days urine was sampled.

Regarding the variability of the dermal exposures measured in our study, it seemed that the level of dermal exposure was explained mostly by complaint factors (company and operator). Therefore, improvement of working circumstances at plantations and modification of the working habits of paraquat spray operators would be likely to result in reduction of exposure. Regarding respiratory exposures, the variability in the measured concentrations of paraquat was mainly determined by variations between companies and days. The latter could be due to variable wind speeds and other weather-related factors. Differences in dermal and inhalational variability could be due to the fact that dermal exposure is also determined by direct contact with paraquat, such as splashes, as opposed to contact with spray mist.[33] Besides, wind speed is likely to have more effect on the small droplets of the inhalable fraction than on total spray mist, which determines dermal exposure and also contains larger droplets.

Hazardous Activities

Table 6 summarizes the hazardous activities and possibly exposed body parts that have been defined using observations of work practices and information from the questionnaires. The activities do not necessarily explain the measured exposures but do reflect situations where exposures are likely to occur.

The preparation of the spraying solution could be a serious source of exposure for the spray operators of plantations B and C, because high concentrated solutions were diluted by the operators themselves. However, because average exposures were lower at these two plantations, this activity did not seem to contribute to measured dermal exposure. The operators probably handled the solution without spilling it. Nevertheless, the risk of high exposure while diluting paraquat is evident. Exposure due to walking in the spray mist can be worsened when application takes place against the wind or when spraying is not carried out at the right angle, for example, when spraying uphill. Indirect exposure of the legs can be high when contact with recently sprayed weeds occurs and results in exposure of the feet when the spraying solution leaks into the worker's boots. Falling of workers in the rough and muddy field was associated with rushing through the plantation. Spray operators may be motivated to run because they are payed per sprayed area.

Protective Clothing

The protective equipment used did not effectively control dermal exposure, for no lower exposure was measured. Overalls, vinyl back aprons, and gloves were clean

TABLE 6 Hazardous activities with regard to high incidental exposures to paraquat

Activity	Risk	Exposure*			
		Dermal	Respiratory	Oral	Eyes
Dilution	Spilling	Hands	–	–	+/–
Transport	Splashing	Face, hands	–	+/–	+
	Tank skipping off	Whole body	–	+/–	+
Filling	Hands in solution	Honds, wrists	–	–	–
	Spilling foam	Back	–	–	–
Application	Walking in spray mist	Hands, arms, legs	+	+/–	+
	Contacting sprayed weeds	Legs, ankles	–	–	–
	Leaking equipment	Back, legs, testicles	–	–	–
	Folling with tonk	Whole body	–	–	–
Repoiring	Fumbling without tools	Hands	–	+	–
	Blowing out nozzle	—	–	–	–
After work	Not showering	Whole body	–	–	–
	Contaminated clothing	Whole body	–	+	+
	Eating food contaminated by hands	—	–	+	–

*+ = exposure likely to occur; +/– = exposure could occur occasionally; – = exposure not likely to occur.

at the beginning of the working day, so contamination cannot be explained by the wearing of contaminated protective equipment. Use of protective clothing does not necessarily result in adequate protection, since the herbicide may get under clothing and gloves, resulting in an increased penetration through the covered skin.[34,35] Also, perspiration resulting from the use of protective equipment can increase dermal absorption.[34] When a worker is wearing gloves, liquid can enter easily at the top of the gloves, or exposure can occur when reusing the gloves during the day. Vinyl back protection is likely to reduce dermal exposure on the back, because the use of this protection was consistently associated with low levels of exposure of the back, while without it high-level exposures of the back sometimes occurred. The fact that the overalls were very thin and the operators' thicker trousers provided more protection might explain why the wearing of long trousers led to significantly less leg exposure compared with the wearing of shorts, while the wearing of overalls did not. The overalls, originally provided to protect against dermal exposure to granulated nematocides, were made of cotton, instead of linen, which is required by the Ministry of Health for the use of paraquat.[36] Whether or not the respiratory protection used at plantation D functioned well could not be assessed since respiratory exposures were measured outside the masks.

Relation of External and Internal Exposures

Paraquat was detected only in urine samples collected after work, suggesting that measured internal exposure is determined by external exposure during the working day. This agrees with the elimination time for paraquat in dogs, which after absorption of small amounts of paraquat excrete approximately 80% within three hours and almost 100% within 24 hours.[26] The kinetics of paraquat in dogs were found to be comparable to those in humans.[37] Although a statistically significant relationship was found between both dermal and respiratory exposures and internal exposure to paraquat, dermal exposure is more likely to have resulted in internal exposure in the case of worker 9, who used a face mask. Assuming that this operator wore his mask throughout the working day and it was effective, the respiratory exposure route did not contribute to his internal exposure.

A relatively high level of dermal exposure to paraquat did not always result in internal exposure, but seems to be essential for it. This can be explained by individual differences in paraquat absorption[30] and kinetics, but also by the fact that skin damage can enhance absorption 5 to 20 times.[10] Worker 9 did in fact have skin aberrations on his wrist in combination with a high paraquat exposure. Worker 1 did not have visible skin damage but was exposed to relatively high levels of paraquat during all three sampling days. Since paraquat itself can make the skin more permeable,[38–40] and because it is able to accumulate in the skin,[41] previous dermal exposure could have led indirectly to the measured internal exposure on the third day.

The respiratory exposure of worker 1 was equivalent to only 0.6% of the total dermal exposure, as can be calculated from Table 4. Half of the paraquat in the breathing zone reaches the alveoli,[42] so the respiratory dose constitutes only 0.3% of the total dermal exposure. Yet, respiratory exposure should not be excluded as a possible route of uptake regarding this worker. Assuming that inhaled aerosols that do not reach the alveoli are being swallowed and that 56% intestinal absorption takes place,[4] the dose of paraqnat absorbed in the intestines of the spray operator can be assnmed to be equivalent to

0.17% of the dermal exposure. The percentage of paraquat absorbed through intact skin after direct exposure has been found to be 0.23–0.29%.[40] Therefore, the respiratory route may be at least as significant as the dermal route in the exposure of this worker.

Measured Exposures Related to Health Effects

Assessing the relationship between the reported local and systemic health complaints and the paraquat exposure levels measured in our study is complicated, since the health complaints reflected circumstances in the preceding 12 months and the exposure levels could have been different. Two workers complained of nausea, stomachache, and headache during the sampling period, which therefore may have been caused by exposure to paraquat. These effects are nonspecific for paraquat poisoning, so it cannot be excluded that they were caused by exposure to the emetic and stenching additives of the paraquat concentrate. Systemic health complaints such as headache and nausea were also mentioned by Weinbaum et al.,[6] and Swan found local effects during his study of paraquat spray operators.[19] Measured inhalable paraquat concentrations (0.1–24.0 $\mu g/m^3$) are well below the threshold limit value (500 $\mu g/m^3$) for inhalable paraquat,[44] which is based-on animal studies, and it is claimed that no adverse health effect due to long-term exposure occurs below this value.[45] It should be noted that this value does not take dermal uptake into account. Two epidemiologic studies did not reveal adverse effects of chronic exposure to diluted paraquat on the liver and lungs.[7,8] Another study reported systemic disease in men and rats after chronic exposures, but the paraquat solution in that study was more concentrated than that used by the spray operators in the present study (2.8 and 0.8% vs 0.1–0.2%).[9]

Although it is not clear whether acute and long-term exposure to the paraquat concentrations measured in this study leads to (adverse) health effects, it should be emphasized that the risk of a high and therefore hazardous exposure is continuously present. Wesseling et al. reported several fatal cases in Costa Rica due to the spillage of concentrated paraquat on the legs, eating food that had been in contact with contaminated hands, spraying into the wind, spraying with a leaking knapsack tank, and spilling of diluted spray solution on the face and mouth after slipping.[15] The occurrence of such calamities is particularly likely when spray operators apply paraquat while their skin is damaged, as is illustrated by a fatal case reported by Fitzgerald of a man who applied paraquat with a leaking knapsack tank while suffering from dermatitis,[14] or when prolonged contact with paraquat spray solution takes place.[15] We found that similar situations regularly arose during normal work practices at the banana plantations where this study was performed. Therefore, the use of paraquat on banana plantations cannot be considered safe.

RECOMMENDATIONS

A strategy to control exposure is necessary, and should focus first on reduction of the risk of high-level exposure. Measures that can be taken include elimination of the exposure source, i.e., replacement of paraquat by, for example, mechanical weed control. The potential for exposure can be reduced by adjusting spraying equipment (closing the tank and providing it with a tap; replacement and better maintenance of old and defective spraying devices), by improvement of field infrastructure (site of application, rail system), and by eliminating risky activities such as mixing of spray solution. Reducing working pressure by changing the salary system, as well as instruction of the workers, could reduce the risk of exposure that is due to carelessness in paraquat application. The implementation of use of personal protective equipment has the least priority, for its effectiveness is questionable and the use of such equipment is often found strenuous and uncomfortable in a tropical climate.

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References

1. Wesseling C, Castillo L, Elinder CG. Pesticide poisonings in Costa Rica. Scand J Work Environ Health. 1993;19:227–35.
2. Unión Mundial para la Naturaleza, Oficina Regional para Centroamérica, Oficina Nacional de Costa Rica. Evaluación del Impacto Socio Amhiental de la Expansión Bananera en Sarapiqui, Torteguero y Talamanca, informe al Ministerio de Recursos Naturales, Energía y Minas (MIRENEM), 1992.
3. CORBANA. Costa Rica, Estadísticas de Exportación Bananera 1994. 1995;ISSN 1409–0058.
4. Hayes WJ, Lawes ER. Handbook of Pesticide Toxicology. San Diego, California: Academic Press, 1991.
5. Haley TJ. Review of the toxicology of paraquat. Clin Toxicol. 1979;14:1–46.
6. Weinbaum Z, Samuels SJ, Schenker B. Risk factors for occupational illnesses associated with the use of paraquat in California. Arch Environ Health. 1996;50:341–8.
7. Senanayake N, Gurunathan G, Hart TB, et al. An epidemiological study of the health of Sri Lankan tea planatation workers associated with long term exposure to paraquat. Br J Ind Med. 1993; 50:257–63.
8. Howard JK, Sabapathy NN, Whitehead PA. A study of the health of Malaysian plantation workers with particular reference to paraquat spraymen. Br J Ind Med. 1981;38:110–6.
9. Levin PJ, Klaff LJ, Rose AG, Ferguson AD. Pulmonary effects of contact exposure to paraquat: a clinical and experimental study. Thorax. 1979;34:150–60.
10. Tabak A, Taitelman U, Hoffer E. Percutaneous permeability to paraquat: in vitro experiments with human skin. J Toxicol Cutan Ocular Toxicol. 1990;9:301–11.
11. California Department of Food and Agriculture. Human health problems associated with the herbicide paraquat in California 1965–1976. Sacramento, California: CDFA, 1977.

12. Sullivan CM. Accidental poisoning by paraquat: a report of two cases in man. Br Med J. 1966;i:1272–3.
13. Fernando R, Harendra de Silva DG, Amarasena TSD. An unusual case of fatal accidental paraquat poisoning. For Science Intern. 1990;44:23–6.
14. Fitzgerald GR, Barniville G, Black J, Silke B, Carmody M, O'Dwyer WF. Paraquat poisoning in agricultural workers. J Irish Med Assoc. 1978;10:336–42.
15. Wesseling C, Hogstedt C, Picado A, Johansson L. Nonintentional fatal paraquat poisonings among agricultural workers in Costa Rica: fifteen case reports. Submitted, 1996.
16. Wojeck GA, Price JF, Nigg HN, Stamper JH. Workers' exposure to paraquat and diquat. Arch Environment Contam Toxicol. 1983; 12:65–70.
17. Chster G, Gurunathan G, Jones N, Woollen BH. Occupational exposure of Sri Lankan tea plantation workers to paraquat. WHO Bull. 1982;71:625–32.
18. Chester G, Woolen BH. Studies of the occupational exposure of Malaysian plantation workers to paraquat. Br J Ind Med. 1981;38: 23–33.
19. Swan AA. Exposure of spray operators to paraquat. Br J Ind Med. 1969;26:322–9.
20. Staiff DC, Comer SW, Armstrong JF, Wolfe HR. Exposure to the herbicide, paraquat. Bull Environ Contam Toxicol. 1975;14:334–40.
21. de Cock J, Heederik D, Hoek F, Boleij J, Kromhout H. Urinary excretion of tetrahydrophthalimide in fruit growers with dermal exposure to captan. Am J Ind Med. 1995;28:245–56.
22. Chester G, Hart TB. Biological monitoring of a herbicide applied through backpack and vehicle sprayers. Toxicol Lett. 1986;33: 137–49.
23. Grandjean P. Skin penetration. Hazardous chemicals at work. London, England: Taylor and Francis, 1990.
24. Ter Kuile WM. Vergleichungsmessungen mit verschiedenen Geraten zur Bestimmung der Gesamtstaubkonzentration am Arbeitsplatz: teil 2. Staub-Reinh Luft. 1984;44:211–6.
25. Kenny LC, Aitken R, Chalmers C, et al. A collaborative European study of personal aerosals sampler performance. Ann Occup Hyg. 1996, in press.
26. Davies DS. Paraquat poisoning: the rationale for current treatment regimes. Hum Toxicol. 1987;6:37–40.
27. NIOSH Manual of Analytical Methods. 1994;2:Method 5003.
28. Åkerblom M. Rapid determination of paraquat in urine with ion pair extraction and spectrophotometry. Bull Environ Contam Toxical. 1990;45:157–64.
29. Durham WF, Wolfe HR. Measurement of the exposure of workers to pesticides. Bull WHO. 1962;26:75–91.
30. Popendorf WJ, Leffingwell JT. Regulating OP pesticide residues for farmworker protection. Residue Rev. 1982;82:125–201.
31. Rappaport SM. Assessment of long-term exposures to toxic substances in air. Ann Occup Hyg. 1991;35:61–121.
32. Rappaport SM. Selection of the measures of exposures for epidemiology studies. Appl Occup Environ Hyg. 1991;6:448–57.
33. Brouwer DH, de Vreede JAF, van Hemmen JJ. Variability and reproducibility of dermal exposure of applicators in greenhouses (abstr). Occup Hyg. April 1996.
34. Cherri JW, Robertson A. Biologically relevant assessment of dermal exposure. Ann Occup Hyg. 1995;39:387–92.
35. Meuling WJA, Bragt PC, Leenheers LH, de Kort WLAM. Prediction of percutaneous penetration. Methods, measurements and modelling: dose–excretion study with the pesticide propoxur in volunteers. Southampton, England: IBC Technical Services Ltd., 1991.
36. Ministerio de Salud, Division Saneamiento Ambiental, Dept. Substancias Toxicas y Medicina del trabajo. Programa Vigilancia y Control de la Actividad Bananera: Normas de Ubicacicion de Condiciones Fisico-Sanatarias y de Procedimientos para el Manejo con Seguridad de Plaguicidas en las fincas bananeras. San Jose, Costa Rica, 1995.
37. Bismuth C, Scherrmann JM, Garnier R, Baud FJ, Pontal PG. Elimination of paraquat. Hum Toxicol. 1987;6:63–7.
38. Athanaselis S, Qammaz S, Alevisopoulos G, Koutselinis A. Percutaneous paraquat intoxication. J. Toxicol Cutan Ocular Toxicol. 1983;2:3–5.
39. Srikrishna V, Edmond Riviere J, Monteiro-Riviere NA. Cutaneous toxicity and absorption of paraquat in porcine skin. Toxicol Appl Pharmacol. 1992;115:89–97.
40. Hoffer E, Taitelman U. Exposure to paraquat through skin absorption: clinical and laboratory observations of accidental splashing on healthy skin of agricultural workers. Hum Toxicol. 1989;8: 483–5.
41. Hoffer E, Tzipiniuk A, Gal M, Taitelman U. Skin versus blood distribution of paraquat in experimental animals during percutaneous absorption. J Toxicol Cutan Ocular Toxicol. 1989;8: 179–82.
42. Hogarty C. Exposure of spray operators to paraquat. Dublin, Ireland: Dublin Institute for Industrial Research and Standards, 1975.
43. Wester RC, Maibach HI, Bucks DAW, Aufrere MB. In vivo percutaneous absorption of paraquat from hand, leg and forearm of humans. J Toxicol Environ Health. 1984;14:759–62.
44. American Conference of Governmental Industrial Hygienists. Threshold limit values for chemical substances and physical agents in the workroom environment with intended changes for 1995–1996. Cincinnati, Ohio, 1995.
45. Boleij J, Buringh E, Heederik D, Kromhout H. Occupational hygiene of chemical and biological agents. Amsterdam, The Netherlands: Elsevier Science B.V., 1995.
46. Astrand PO, Rodahl K. Textbook of Work Physiology: Physiological Bases of Exercise. New York: McGraw–Hill Book Company, 1986.