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Occupational Exposure of Forest Workers to Glyphosate during Brush Saw Spraying Work*

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The purpose of this study was to measure forest workers' exposure to the herbicide glyphosate during silvicultural clearing work done with brush saws equipped with pressurized herbicide sprayers. Both the exposed (study) group and the nonexposed (control) group contained five persons who were medically examined before and after their 1-week working period (including laboratory tests) for possible health effects. In addition, exposure to glyphosate was measured in the study group from samples taken from the workers' breathing zone and from urine samples collected during the afternoons of the workweek. The laboratory tests and urinary glyphosate analyses were repeated for the exposed group 3 weeks later, when the men had entirely stopped their work with the herbicide. Exposure to glyphosate through the workers' breathing zone was low. The highest value found was 15.7 μg/m³. In this study, a biological monitoring method was also developed to monitor the workers' exposure to glyphosate. Urine concentrations were under the gas chromatographic detection level of <0.1 ng/μL (<1.0 μmol/L). No major differences were noted, either in medical examinations or in the laboratory tests performed, between the exposed and control groups before and after the work period.

Most of the studies on the toxicity of glyphosate have been published by the manufacturer of Roundup® (Monsanto Co., St. Louis, Mo.). For rodents receiving glyphosate orally, the LD_{50} value is between 4–6 g/kg. The substance has been shown to cause moderate (commercial formula, Roundup) or slight (pure glyphosate) irritation to the eyes and skin of rodents. In long-term toxicity tests it has not been carcinogenic.[1] In mutagenicity tests (based on the sister-chromatid exchange test), glyphosate has been at most only weakly mutagenic.[2] There have been no mutagenic effects with the Ames test,[3] nor more recently in in vitro and in vivo assays.[4] Embryotoxicity has been tested in chicken eggs[5] and in mallard eggs[6]; no embryotoxicity was found. There are variations in the toxicity of pure glyphosate, the commercial product, and the surface-active agent MON 0818, of which the commercial product contains 15%. The surfactant was most toxic when tested on salmonids,[7] compared with glyphosate and Roundup.

Some studies have been done on the toxicity of glyphosate to man. The phototoxic effect of glyphosate was reported[8] but was later explained to be caused by the preservative benzisothiazolone in the commercial product Tumbleweed (Murphys Ltd., Wheathamstead, U.K.).[9] When Roundup was tested in 346 volunteers for irritative and allergic reactions, the results were negative, and its irritant potential was comparable to a baby shampoo.[10]

Roundup has been reported to have caused several poisonings, even some fatal ones, when taken orally. The toxic effect was presumably caused by the surface-active agent (polyoxyethyleneamine or POEA).[11] The manufacturer disputed the report because there was no mention of the possible ingestion of medicines and alcohol at the same time. The manufacturer also stated that it had never claimed Roundup to be nontoxic, but to be safe if stored, handled, and used in accordance with the manufacturer's instructions.[12] Recently, the possible toxicity of glyphosate has aroused public interest, because it is being used increasingly in home gardens.[13]

The aim of the study was to measure the workers' exposure to the herbicide glyphosate when they used sprayers connected to brush saws. Exposure was determined from the breathing zone and from urine samples. The possible health effects of glyphosate at the measured exposure levels were also examined.

EXPERIMENTAL MATERIALS AND METHODS

Workers and Exposure

The field studies were performed in northeastern Finland during one week in August 1988. The test group consisted of five forest workers who sprayed glyphosate (Roundup) with a brush

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saw. The control group consisted of five forest workers who planted young trees.

The mean age of the exposed group was 46 (range 44–49 yr) and that of the control group 47 (range 36–54 yr). Two men in both groups were smokers, but none took any medication nor had been exposed to known chemicals during the previous year.

The forest workers prepared a fresh spraying solution every day by mixing Roundup (360 g/L glyphosate as isopropylamine salt), water, and a commercial carrier liquid, which contained 40% isopropylamine alcohol (Kantotehoste®, Kemira Inc., Finland). The final spraying solution contained 8% Roundup, 87% water, and 5% carrier liquid. The workers used an average of 9.8 L of this solution per day per man, and they worked effectively about 6 hr/day.

The herbicide mixture for each day was mixed at the field store by the sprayers themselves; they also filled the saw tanks (3.5 L) when necessary. During the fillings and while repairing the brush saws, skin contamination by the glyphosate mixture occurred. There was no possibility for the men to wash their hands in the field. The workers wore cotton overalls, cotton or rubber gloves, a hat or safety helmet, and rubber boots. On 2 days during the study week rain fell for some time. On those days the workers used rain clothes.

The work was physically heavy, and it was done under contract.

Clinical and Laboratory Examination

The medical examination on the first and last workday included a health questionnaire and clinical examination by a doctor. Laboratory tests included erythrocyte sedimentation rate (ESR), hemoglobin (Hb), mean corpuscular hemoglobin concentration (MCHC), white blood cell count (WBC), creatinine (Crea), aspartate aminotransferase (ASAT), alkaline phosphatase (ALP), creatine kinase (CK), γ-glutamyltransferase (GT), C-reactive protein (CRP), hematocrit (Hct), white blood cell differential count, and urine sample with sediment microscopy. Electrocardiogram (ECG) and pulmonary function tests (peak expiratory flow [PEF], forced vital capacity [FVC], and forced expiratory volume in one second [FEV_1]) were also performed.

The health questionnaire was completed with the help of an occupational health nurse. It included questions on the general health condition as well as an evaluation of smoking habits, alcohol consumption, medication, and possible exposure to chemicals (at work or outside work) during the previous year. Specific questions were asked about the following symptoms before and after the workweek: eye irritation, blurred vision, skin symptoms, nausea, fatigue, headache, tremor of the hands, and muscular spasms or twitching.

In the clinical examination, special attention was paid to the possible target organs such as the skin and mucous membranes, respiratory organs, nervous system, muscles, liver, and kidneys, as well as to the cardiovascular organs. Blood pressure and pulse were measured, as well at the pressure craft of the hands (by Martin® Vigorimeter® Tuttlingen, Germany).

The health questionnaire and laboratory tests were repeated 3 weeks later, when the forest workers had stopped their work with the herbicide.

Sample Collection

Air samples were collected with a portable pump (rate 1.5 L/min) onto an absorption liquid (20 mL distilled water in a

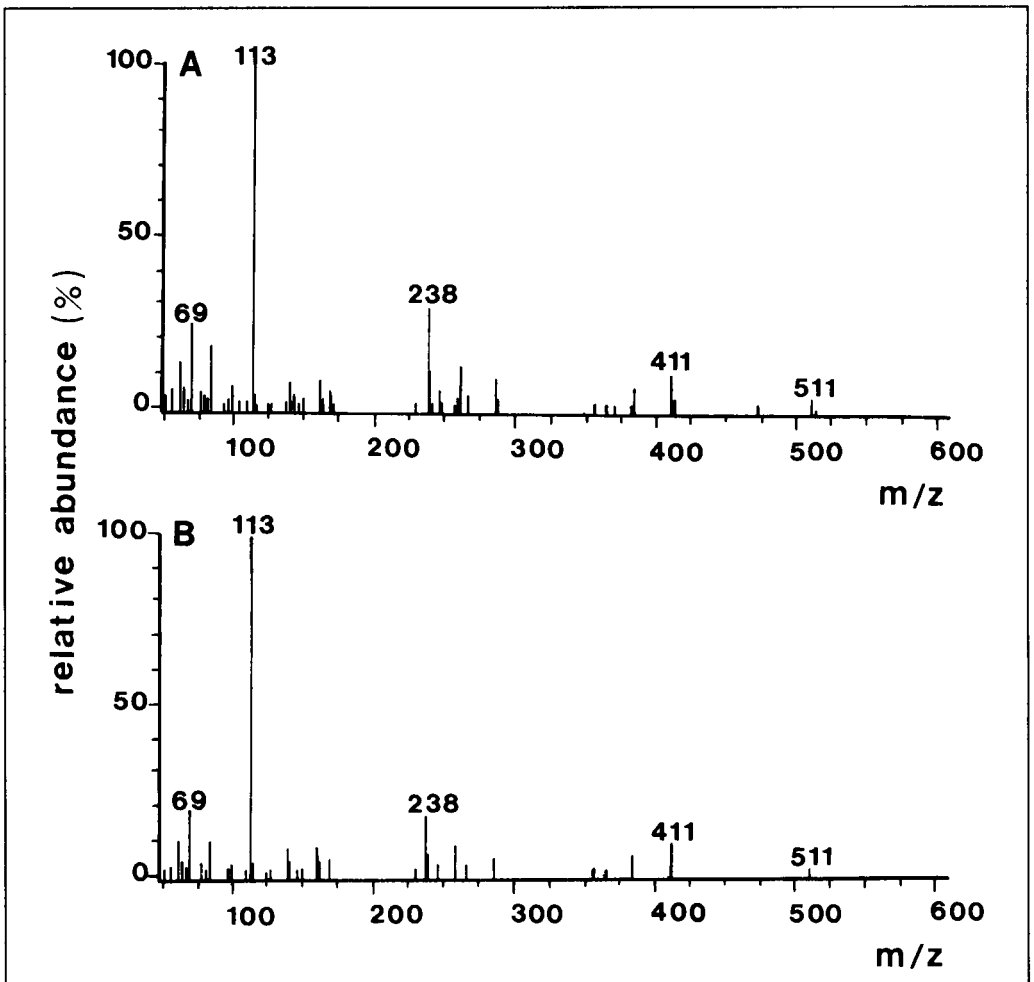

FIGURE 1. Comparison of the electron impact (EI) mass spectrum of derivatized glyphosate (A) in an air sample and (B) in a glyphosate standard

TABLE I. Blood Values, Serum Chemistry, and Lung Function Tests of the Exposed and Control Forest Workers before and after Glyphosate Exposure

Laboratory and Lung Function Tests		Before Glyphosate Exposure				After Glyphosate Exposure			
		Exposed (n = 5)		Control (n = 5)		Exposed (n = 5)		Control (n = 5)	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
Blood values									
ESR	(mm/hr)	4.6	1.9	3.8	1.5	5.4	2.7	3.8	1.5
Hb	(g/L)	153	6.9	154	8.1	151	6.9	165	11.7
Hct	(%)	46	1.8	47	2.1	46	2.1	49	3.3
MCHC	(g/L)	335	15.0	330	9.1	334	13.4	334	7.2
WBC		4.4	0.3	6.6	2.7	3.9	0.3	5.9	2.1
Serum chemistry									
Crea	(μmol/L)	93	6.1	101	16.6	88	7.4	100	17.3
ASAT	(U/L)	22	3.0	18	2.2	28	9.8	21	3.1
GT	(U/L)	19	5.2	15	6.3	18	4.3	16	7.2
ALP	(U/L)	114	22.3	130	13.9	115	25.1	124	14.4
CK	(U/L)	113	44.1	139	51.5	338	212.8	208	82.4
Lung function tests									
PEF	(L/min)	523	46.8	563	82.4	560	32	603	97.0
FVC	(L)	4.79	0.43	4.18	0.63	4.74	0.38	4.29	0.81
FEV_1	(L/sec)	3.40	0.38	3.16	0.62	3.34	0.37	3.32	0.67
FEV_1/FVC	(%)	71.2	4.8	75.4	9.0	70.4	6.1	77.4	5.1

midget impinger) in the breathing zone of the workers. The sampling time varied from 1 to 6 hr.

Urine samples were taken at the end of each workday during the study week. A follow-up sample was taken from each of the five sprayers after the 3-week work period.

All the samples were kept frozen (–18°C) until analyzed.

Instrumentation

A Hewlett Packard 5880 A gas chromatograph equipped with a ^{63}Ni-electron capture (EC) detector (330°C) was used for all measurements. An HP fused silica capillary column (25 m × 0.20 mm ID) was used. The column temperature was kept at 50°C for 1 min, then raised to 110°C at 5°C/min and held there for 1 min. Next, the temperature was raised 140°C at a rate of 2°C/min and held there for 2 min, after which the temperature was raised quickly to 280°C.

Mass spectra were collected with a VG TRIO-2 quadrupole mass spectrometer (70 eV) interfaced with a Hewlett Packard 5890 gas chromatograph. The column used was the same as with the EC-detector.

Analysis of Glyphosate

The air samples collected from the workers' breathing zone were transferred to 50-mL round-bottomed flasks and rotary evaporated to dryness at about 60°C. The residues were dissolved in 350 μL of trifluoroethanol (TFE) and 700 μL of trifluoroacetic anhydride (TFAA) according to the method of Deyrup et al.[14] and transferred to capped Kimax tubes (Schott Geräte, Mainz, Germany). The tubes were incubated at 100°C for 1 hr, after which they were cooled to room temperature and the reagents removed with a stream of nitrogen at 25°C. The residues were dissolved in 1 mL of ethyl acetate, the tubes were capped, and the next day 1-μL aliquots were injected into the gas chromatograph.

Glyphosate was identified in the air samples by gas chromatography/mass spectrometry (GC/MS) (Figure 1) and quantitated with GC/EC and GC/MS using standards of glyphosate in distilled water.

The detection limit of glyphosate in the gas chromatograph was 0.1 ng/μL (about 0.3 μg/m^3) in the air samples. The average recovery of glyphosate from distilled water samples was 94% at the 1-ppm level.

Urine samples were cleaned up with extraction cartridges containing the strong anion exchanger, SAX (purchased from Analytichem International, Harbor City, Calif.). One mL of each urine sample was diluted to 10 mL with distilled water and passed through a cartridge. The cartridges were washed with 3 mL of distilled water. Glyphosate was then eluted from the anion exchangers with 2 × 0.5 mL of 2 N HCl, and 100 μL of this solution was pipetted into Kimax tubes and evaporated to dryness with a stream of nitrogen. Next, 50 μL of TFE and 100 μL of TFAA were added, and the tubes were capped and incubated at 100°C for 1 hr, after which they were cooled. The solutions were evaporated to dryness and dissolved in 200 μL of ethyl acetate. The next day, 1-μL aliquots were injected into the gas chromatograph. The derivatives were found to be stable for several days. The identification and quantitation of glyphosate in the urine samples was performed as with air samples. Glyphosate standards were prepared in control urine taken from nonexposed persons.

This purification and derivatization method was also used for the determination of (aminomethyl)-phosphonic acid (AMPA), the principal metabolite of glyphosate, in the urine

samples. The detection limits of glyphosate and AMPA in the urine samples were 0.1 ng/μL (1 μmol/L) and 0.05 ng/μL (0.5 μmol/L), respectively. The method was tested to be linear over the range of 0.1–10 ng/μL of glyphosate and AMPA.

RESULTS

Workers' exposure

The glyphosate levels in the air samples taken from the workers' breathing zone were very low. The samples collected at midweek during spraying (Wednesday) contained <1.25 μg glyphosate/m^3 air. At the end of the spraying week (Friday), two air samples were found to have measurable levels of glyphosate; the concentrations were 2.8 μg/m^3 and 15.7 μg/m^3.

In this study, a new biological monitoring method was also developed to monitor the workers' exposure to glyphosate. During the spraying week and also after a 3-week work period, the glyphosate concentration in the urine samples remained below the gas chromatographic detection level of <0.1 ng/μL (<1.0 μmol/L). One urine sample was further quantitated with selective ion monitoring mass spectrometry and was found to contain a glyphosate concentration of 0.085 ng/μL (0.85 μmol/L).

The metabolite AMPA was not detectable in the urine samples.

Clinical and Laboratory Findings

Findings in the medical examinations done before and after the exposure did not differ. In the health questionnaire, two workers in the exposed group reported headache during the workweek, compared to none in the control group. There were no other specified symptoms in either group.

The results of the laboratory and pulmonary function tests are shown in Table I. There were no major differences in these values, except for serum creatine kinase, which seemed to rise more in the exposed than in the control group. This difference was not, however, statistically significant ($p = 0.4$, repeated measures analysis of variance). The values were normalized in the third analysis made for the exposed group after a 3-week working period (mean 189 U/L, SD 61.7, normal value for men <270 U/L).

DISCUSSION

The glyphosate concentrations measured from the forest workers' breathing zone were low. In this study, exposure to glyphosate by inhalation was clearly lower than the corresponding exposure of forestry workers to phenoxy acid herbicides measured earlier in a similar study.[15]

Although the workers' exposure to glyphosate was low with the spraying method used, some exposure may still occur, for example during pesticide dilution and administration and during repairing and servicing of the sprayer in the field.

The exposed and control groups did not differ from each other in the medical examination either before or after the workweek. Two workers in the exposed group experienced mild headache of unknown reason while working. No statistically significant changes were noted in the laboratory or pulmonary function tests. Because of the minimal exposure and small worker groups in this study, remarkable conclusions of the effects of glyphosate cannot be made.

Using basic protective equipment suitable for pesticide work is recommended in order to minimize possible exposure.

The analysis of urine samples can be used to indicate glyphosate exposure.

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