ESTIMATION OF EXPOSURE OF PERSONS IN CALIFORNIA TO PESTICIDE PRODUCTS THAT CONTAIN PHOSMET AND ESTIMATION OF EFFECTIVENESS OF EXPOSURE REDUCTION MEASURES

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ABSTRACT

Phosmet (N-(mercaptomethyl)phthalimide, S-(O,O-dimethylphosphorodithioate) is a selective organophosphate insecticide/acaricide, available in 5 formulations, for use on field crops, ornamentals, livestock and domestic pets to control several insect pests. This compound is currently in the Risk Assessment Review Process formulated by SB950. The dermal absorption rate for phosmet based upon registrant-supplied data was 0.54%/hr (13.1%/24 hr). Dislodgeable residue data supplied by the registrant was used to calculate dermal exposures to homeowners and workers with the use of appropriate transfer coefficients. Estimated absorbed doses for grape, pear and citrus harvesters entering a field one day after application were 0.1, 0.4 and 1.4 mg/kg/d, respectively. A worst case scenario for homeowners estimated an absorbed dose no greater than 0.005 mg/kg/d.

INTRODUCTION

Phosmet [N-(mercaptomethyl)phthalimide, S-(O,O-dimethylphosphorodithioate)] is a selective organophosphate insecticide/acaricide used on field crops, ornamentals, livestock and domestic pets for the control of several insect pests. Phosmet is labeled for use on 18 agricultural crops, several deciduous and evergreen ornamental trees, cattle and hogs and as a pet dip and flea collar for dogs[1]. Based on the 1985 California Department of Food and Agriculture Annual Pesticide Use Report and the CDFA SB950 Report on Chemical Active Ingredients Sold in 1985, agricultural uses accounted for 90,990 pounds of the reported 239,227 pounds of phosmet sold in California[2,3] in 1985. Phosmet is not a Restricted Use Pesticide and applications do not have to be reported to county personnel. Therefore the amount reported for agricultural use is probably underestimated. Reported use on alfalfa and almonds for 1985 was 78,649 pounds or 86% of the total reported for agricultural use. In 1985 there were 435,264 acres planted to almonds and 1,030,000 acres of alfalfa[4,5]. Phosmet is sold to commercial growers, livestock owners, pet kennels and homeowners.

FORMULATIONS

There are 5 different formulations and 25 products containing phosmet registered in California[1]. Of these formulations, 11 are dog collars, 7 are emulsifiable liquids or concentrates, 4 dusts, 2 wettable powders and 1 liquid. Agricultural formulations include 4 emulsifiable concentrates and 1 liquid for commercial livestock use and 1 wettable powder for crop insect control. All collar, dust and 3 emulsifiable concentrate formulations are available to pet kennels and homeowners for flea control on pets. Some of the pet dips and dusts are available to the homeowner only through a veterinarian. One wettable powder formulation is available to the homeowner for garden insect control. The percent active ingredient ranges from 5-50%. Pet dusts have the lowest % active ingredient at 5%, emulsifiable concentrates range from 11-12%, the garden wettable powder formulation contains 12%, dog collars are 15% and the commercial agricultural wettable powder and liquid formulations contain 50% active ingredient.

PHYSICAL CHARACTERISTICS

Phosmet is a white crystalline solid with a distinctive odor[7,8]. The molecular weight is 317.3 and formula is $C_{11}H_{12}O_4NPS_2$. Phosmet is slightly soluble in water (25 ppm) and moderately soluble in xylene (200 g/L). The vapor pressure is <0.1 µm at 50 ^oC, therefore it is relatively nonvolatile. Phosmet has a melting point of 66.5-69.5 ^oC and a flash point of 180 ^oC. Phosmet is thermally stable with a half-life of 80 days at 65 ^oC but readily photodegrades with a half-life of only 2.5 hours. Hydrolysis is pH dependent: the half-life at pH 5 is 9.4 days whereas at pH 9 it is <1 hour[9].

ILLNESS REPORTS

A review of the pesticide illness reports for the years 1978-1986 revealed there were 16 reported cases of phosmet related illness[10]. Of these cases, 13 were systemic illnesses, 2 were skin irritations and 1 was an eye injury. At least two of the systemic illnesses were of questionable validity. Thus, the number of reported illnesses averaged two cases per year. In 1985 there was approximately 240,000 pounds of phosmet sold in California indicating the relative safety with which this compound has been handled. The California Department of Health Services Hazard Evaluation System and Information Service recently published a report Illness Associated With The Use Of Flea Dip Products in which phosmet related illnesses were documented in pet groomers[11]. The two documented cases presented showed that few if any of the label precautions were followed, which resulted in rather severe organophosphate poisoning.

LABEL PRECAUTIONS

Products which contain phosmet carry either the "Caution" or Warning" signal word. Pet collars and dusts list "Caution" while pet dips, livestock formulations and home or agricultural products have the "Warning" signal word. Mitigation measures listed on the label range from avoid contact to wearing rubber gloves, goggles and protective clothing. Pet dips and all but one of the livestock sprays require rubber gloves, goggles and protective clothing to be worn. All other products warn against contact but have no specific clothing requirements. The current reentry interval for crops treated with phosmet is when the spray has dried unless protective clothing is worn.

RESIDUE AND USE CHARACTERIZATION

Agricultural application rates for phosmet range from 0.25 to 15 pounds active ingredient (A.I.)/acre with reentry allowed after the spray has dried[1]. Multiple applications are allowed for most crops. Preharvest intervals range from 3 to 30 days with the majority set at 7 to 14 days. Pets treated with phosmet sprays or dips may not be retreated for 7 days. Cattle have a 7- to 10-day retreatment and 3-day preslaughter interval while hogs have a 14-ay retreatment and a 1-day preslaughter interval.

The residue tolerances for phosmet range from negligible to 40 ppm[12]. Most crop commodities have tolerances between 2 and 10 ppm. Only two commodities have tolerances >10 ppm: kiwifruit is set at 25 ppm and alfalfa at 40 ppm. The highest residue levels measured during the Apple Maggot Eradication Program by the California Department of Food and Agriculture was 2.20 ppm on fruit and 2.84 $\mu g/cm^2$ on leaves[13]. Registrant-supplied data showed 0-day apple fruit residues <10 ppm with application rates at or above label rates[14]. Monitoring by the FDA over a 4-year period for pesticide residues in foodstuffs resulted in 230 findings of phosmet out of 15,000 samples[15]. All residues were within tolerance limits.

A dislodgeable residue study was performed by the registrant on three crops to estimate possible worker exposure and reentry intervals[16]. Maximum label application rates of 2, 10 and 30 pounds per acre (50% A.I.) were made to grapes, pears and oranges respectively, and samples collected periodically for 27-28 days. The maximum sum of phosmet and phosmet equivalent oxon residue for grapes, pears and oranges was 1.72, 5.95 and 11.8 μg/cm², respectively, with the maximum values recorded on 0 or 1 day post application. Regression equations for the residue decay were calculated for each crop and presented in Table 1.

Table 1. Regression equations for phosmet dislodgeable residue on grape, pear and orange foliage.

Crop	Regression Equation	R Value
Grapes	ln residue = 0.286 - 0.068X*	-0.95
Pears	ln residue - 1.738 - 0.066X	-0.98
Oranges	ln residue - 2.426 - 0.017X	-0.98

*X - Days after application. CDFA WH&S, Blewett. Phosmet. 1987.

ENVIRONMENTAL BEHAVIOR

Phosmet is rapidly broken down in soil by hydrolysis and microbial action[17]. The half-life in soil ranges from 3-12 days with the mean near 3 days. By 14 days, 40% of ^{14}C-phosmet was recovered as $^{14}CO_2$. Phosmet degrades to several intermediates but phthalic acid and phthalamic acid greatly predominate[18]. Phosmet rapidly degrades via photolysis with a half-life of 3-4 hours. However, on plant leaf surfaces 40-50% was unchanged after 96 hours including 25 hours of sunlight[17]. Apparently, phosmet may be slightly soluble in epicuticular wax and thus resistant to hydrolysis. Phosmet does not leach readily even after multiple soil applications. The greatest concentration (0.29 ppm) was found in the 0-3" fraction of soil and minimal amounts (<0.05 ppm) found in the 6-12" fraction[19].

ANIMAL METABOLISM

Phosmet is rapidly metabolized and excreted principally via urine in rats and cattle[20,21,22]. When ^{14}C-phosmet was administered to rats by oral gavage, 98.9% of the ^{14}C was recovered in 120 hours after administration with 78% in the urine, 18% in the feces and 2.6% in the tissues[20]. In this study 68% was found in the urine and 9% in the feces at 24 hours. The major metabolites in a separate rat metabolism study were phthalamic acid at 41%, phthalic acid at 21% and five minor metabolites that totaled 11% of the administered radioactivity[22]. Similar results were observed in cattle when phosmet was dermally applied[21]. Blood, urine and feces were monitored for 168 hours. A total of 9.6% of the applied dose was recovered, indicating a rather low dermal absorption rate. After 24 hours, 5.9% was excreted in the urine and 1.1% excreted in the feces.

At 168 hours, urine accounted for 8% and feces ~1.6% of the applied dose. Tissue residues revealed that the subcutaneous fat had the highest concentration (0.11 ppm) of phosmet metabolites but no tissue selectively retained the compound. The presence of two metabolites (phthalic acid and phthalamic acid) as major urine excretory products provides an excellent mechanism for biological monitoring of this compound.

DERMAL TOXICITY

Phosmet has a dermal toxicity LD_{50} of >3160 mg/kg in the rabbit and 71-300 mg/kg in the mouse[8]. Phosmet is classified as a mild skin and eye irritant with transient effects.

DERMAL ABSORPTION

A rat dermal absorption study was performed and submitted by the registrant[23]. Methylene bridge-labeled ^{14}C-phosmet was applied to the clipped backs of male rats at three dosages: 1.68, 15.08 and 77.43 mg/29 cm^2 (0.058, 0.52 and 2.67 mg/cm^2). A protective appliance was used to prevent loss of material from the skin. Four animals were sacrificed at 0.5, 1, 2, 4, 10 and 24 hours post treatment. Urine, feces, blood, carcass and the dosed area of the skin was monitored. Phosmet that penetrated but remained in the skin was considered as absorbed. Radioactivity recoveries ranged from 95% to 112%. Absorption was most rapid in the first half-hour for all time periods: rates of 0.9% at the high dose to 4.7% at the low dose were measured. Dermal absorption rates at 24 hours, corrected for the % recovery and mean amount on the protective device, were 0.89%, 3.80% and 13.1%/24 hours at 2.67, 0.52 and 0.058 mg phosmet/cm^2. Although absorption rates decreased as dosage increased, total absorption was proportional to the applied dose. Absorption rates in µg/cm^2/hr for the low to high doses were 0.32, 0.82 and 0.99 µg/cm^2/hr. The amount of absorbed ^{14}C-phosmet in the skin at 24 hours ranged from 33% at the low dose to 74% at the high dose. There was no mention of the possible metabolic fate of this phosmet fraction. The amount of radioactivity excreted in the feces was a consistent 3-6% while the amount of absorbed phosmet excreted in the urine ranged from 13% at the high dose level to 45% at the low dose level. Realistic worker exposure estimates would be less than the lowest dose tested (0.058 mg/cm^2), therefore the dermal absorption rate of 0.54%/hr based on the 24 hour absorption data (13.1%/24 hr) was used in subsequent calculations.

WORKER EXPOSURE ESTIMATES

An exposure study was submitted by the registrant in which two volunteers in a commercially-sprayed pear orchard simulated various work tasks that homeowners would perform in a garden[24]. Dermal dosimeters (24.6 cm^2) were placed at 11 exterior locations and 2 sites under the outer clothing (coveralls). Additionally, respiratory monitoring was performed. Hands were monitored by the cotton glove and hand-rinse methods. Both methods were performed for comparison purposes. The data presented demonstrated that exposure measured by the hand-rinse method was 44% of the cotton glove method. A commercial pear orchard in

California was sprayed with 4.8 pounds phosmet A.I./acre with an airblast sprayer. The subjects worked 2, 30-minute periods each day on days 0, 1, 2, 3, 4, 5 and 7 post application.

One person was monitored on day 14. Each 30-minute period was evenly divided into picking fruit and "inspecting" the trees. A third time period was performed for 3 days to evaluate the hand-rinse method. Recovery of phosmet from spiked samples was variable, 60-124%. All respiratory samples and patches under the clothes were below the level of detection (0.5 µg for respiratory, 0.002 µg/cm^2 for gauze patches). Therefore only the head, neck and hands were used to estimate exposure.

The study appeared well conducted but several concerns were raised. No monitoring was performed to estimate homeowner exposure during an application. This would potentially be a more significant period for exposure than harvesting or "inspecting" trees. The study used a 30-minute exposure period which likely is an underestimation of the time homeowners would spend harvesting fruit from trees. The registrant based their exposure estimates only on the exposed skin (head, neck and hands) because no residue was found on the dermal patches under the outer clothing. The 30-minute exposure period gave scant time for phosmet to transfer through the clothing. A typical homeowner may wear contaminated clothing for up to 12 hours. During the spring and summer months when phosmet is likely to be applied, homeowners could be expected to wear short-sleeved shirts and possibly short pants. Thus, the forearms and or legs would be major areas of exposure yet were not included in this study. To the registrant's credit, they monitored intimate contact with the treated crop (harvesting) immediately after application, which few homeowners would be expected to do. The data provided were reworked to estimate homeowner exposure following an application of phosmet and the results presented in Table 2. The following notes apply:

1. Thirty-minute exposure period was used.
2. Respiratory exposure was negligible.
3. Hand exposure - 4 hours.
4. Dermal exposure - 12 hours.
5. No clothing penetration.
6. Exposure sites were head, neck, forearms and hands.
7. A 0.54%/hour dermal absorption rate was used.
8. A 44% cotton glove factor for hand exposure estimation was included.
9. Average homeowner weighs 54.8 kg.
10. Homeowner works 10 days/year in treated plants.
11. Homeowner would be exposed for 40 years of 70-year lifetime.

Table 2. Phosmet exposure estimates for homeowners working in treated pear trees, Estimates assume 30-minute exposure, 10 days exposure/year and 40 years.

Days After Application	Gross Exposure ±SD (mg/d)	Absorption/Day (µg/kg/d)	Daily Dose/Year (µg/kg/d/yr)	Daily Dose/ Life (µg/kg/d/70yr)
0	6.39 ± 0.90	4	< 1	< 1
1	8.16 ± 2.41	5	< 1	< 1
2	7.55 ± 0.82	4	< 1	< 1
3	5.90 ± 1.35	3	< 1	< 1
4	5.69 ± 0.69	3	< 1	< 1
5	4.49 ± 0.88	2	< 1	< 1
7	3.76 ± 0.46	2	< 1	< 1
14	2.10 ± 0.43	< 1	< 1	< 1

CDFA WH&S, Blewett. Phosmet. 1987.

The hand exposure contribution to total exposure ranged from 66-96% with a mean of 85%, thus the hands represent the most significant area of exposure. The data presented show that homeowners would be subject to minimal phosmet exposure.

The registrant has submitted a dislodgeable residue study for phosmet on grapes, pears and oranges as an estimation of worker exposure for these and similar crops[25]. Reentry intervals of 0, 14 and 100+ days for grapes, pears and oranges, respectively, were calculated based on an acceptable daily exposure level of 1 mg/kg by the registrant. Currently there is no reentry interval (spray is dry) for phosmet. The worker exposure estimates were made by the use of dislodgeable residue data, the appropriate transfer coefficient from Popendorf and Leffingwel1[26] and an assumption of an eight-hour workday. Commercially grown grapes, pears and oranges were periodically sampled for 27-28 days post application for phosmet residues. The grapes were treated at ~1 lb A.I./acre, pears at 4.8 lbs A.I./acre and oranges at 15 lbs A.I./acre. Both phosmet and its oxon were monitored. Total phosmet equivalents were calculated by multiplying the quantity of oxon by the ratio of their respective LD_{50}'s (5.07) to reflect the greater acute toxicity of the oxon. The transfer coefficients used were 6.6 µg/hr/ng/cm^2 for citrus and 3.8 µg/hr/ng/cm^2 for pears and grapes. Two trials were performed for each crop and the data combined where appropriate. The two sets of pear data were determined to be from separate populations, therefore only the trial with the highest residue level was used. The exposure and the daily dose for grapes, pears and oranges are presented in Tables 3, 4 and 5, respectively. The following notes apply:

1. Transfer coefficients used were 3.8 µg/hr/ng/cm^2 for grapes and pears and 6.6 µg/hr/ng/cm^2.
2. Transfer coefficients estimate dermal exposure including clothing protection.
3. Respiratory exposure was negligible.
4. An 8-hour workday was assumed.
5. Average worker weighs 54.8 kg.
6. Dermal absorption rate of 0.54%/hour was used.

Table 3. Dislodgeable residues and exposure estimates for grape workers. Estimates assume eight-hour workday and transfer coefficient of 3.8 µg/hr/ng/cm^2.

Days After Application	Sum of Residues (µg/cm^2)	Exposure/ Day (µg/kg/d)	Absorption/ 12 Hours (µg/kg/d)	Absorption/ 24 Hours (µg/kg/d)
0	1.72	954	62	124
1	1.22	677	44	88
3	0.96	532	34	69
4	0.84	465	30	60
6	0.96	532	34	69
9	0.72	399	26	52
13	0.62	344	22	45
20	0.23	128	8	16
27	0.27	150	10	19

ln residue = 0.286 - 0.068 X* r = -0.95

ln exposure = 6.6 - 0.068 X r = -0.95

*X= Days After Application CDFA WH&S, Blewett. Phosmet. 1987.

Table 4. Dislodgeable residues and exposure estimates for pear workers. Estimates assume eight-hour workday and transfer coefficient of 3.8 ug/hr/ng/cm2.

Days After Application	Sum of Residues (µg/cm²)	Exposure/ Day (µg/kg/d)	Absorption/ 12 Hours (µg/kg/d)	Absorption/ 24 Hours (µg/kg/d)
0	5.04	2796	181	363
1	5.95	3300	214	428
2	4.64	2574	167	336
3	4.65	2580	167	334
4	4.44	2463	160	319
5	4.16	2307	150	299
7	3.76	2086	135	270
10	3.56	1975	128	256
14	2.03	1126	73	146
21	1.13	627	41	81
28	1.02	566	37	73

ln residue = 1.74 - 0.066 X* r = 0.98
ln exposure = 8.05 - 0.066 X r = 0.98

*X= Days After Application CDFA WH&S, Blewett. Phosmet. 1987.

Table 5. Dislodgeable residues and exposure estimates for orange workers. Estimates assume eight-hour workday and transfer coefficient of 6.6 µg/hr/ng/cm^2.

Days After Application	Sum of Residues (µg/cm^2)	Exposure/ Day (µg/kg/d)	Absorption/ 12 Hours (µg/kg/d)	Absorption/ 24 Hours (µg/kg/d)
0	11.8	11369	737	1473
1	11.0	10599	687	1374
3	10.4	10020	649	1299
5	10.8	10406	674	1349
7	9.9	9539	618	1236
10	9.2	8864	574	1149
14	8.9	8575	556	1111
21	7.9	7612	493	986
28	7.1	6841	443	887

ln residue = 2.42 - 0.017 X* r = -0.98

ln exposure = 9.30 - 0.017 X r = -0.98

*X=Days After Application CDFA WH&S, Blewett. Phosmet. 1987.

To estimate the daily dose/year and daily dose/life the daily exposure value for each crop at 1 day post application was used. This value was chosen to represent the highest exposure possibly expected and could be assumed to be a "worst case" scenario. The estimates for each crop are presented in Table 6.

Table 6. Phosmet exposure and daily dose estimates for grapes, pears and oranges. Estimates based on 100 workdays/year for grapes, 30 days/year for pears and oranges.

Crop	Exposure/Day (µg/kg/d)	Absorption/ 24 hours (µg/kg/d)	Daily Dose/ Year (µg/kg/d/yr)	Daily Dose/ Life (µg/kg/d/70yr)
Grapes	677	88	24	14
Pears	3300	428	35	20
Oranges	10599	1374	113	65

CDFA WH&S, Blewett. Phosmet. 1987.

It is stressed that the figures in Table 6 may be a gross overestimation of exposure, particularly since individuals would not be expected to work continuously in fields that had been recently treated.

Phosmet is the active ingredient in several dog collars. A registrant has submitted data on the release rate of these collars[27]. The collars contain an average of 3.3 g of available phosmet as the active ingredient. The release rate over the useful life of the collar is ~9 mg/day. During the first week the collars release ~59 mg/day. Thus there is a potential for exposure to individuals handling the collar or petting a dog wearing the collar.

Phosmet was used in the Apple Maggot Eradication Program in the north coast counties of California in 1986. Apple maggot hosts (apple, crabapple and hawthorne) in commercial orchards or residential areas were subject to spraying with phosmet or other control measures when apple maggot adults were detected in the vicinity. Phosmet was selected for use in the program due to its efficacy, relative safety compared to other insecticides and rapid degradation in the environment.

A worst case exposure scenario for the Apple Maggot Eradication Program would entail a small child (22 kg, 2019 cm^2 exposed surface area) playing in a treated area immediately after the spray has dried. The spray concentration of phosmet during the Apple Maggot Eradication Program was 0.75 lbs A.I. per 100 gallons of water with plants sprayed to the point of dripping. If five gallons per application were used then each treated tree or shrub received approximately 0.60 oz of phosmet. It was assumed that skin exposure would equal the highest recorded residue concentration (2.84 ug/cm^2) as measured by the CDFA Environmental Monitoring Unit[13] to date and total dermal exposure would last 12 hours. A dermal absorption value of 0.54% per hour was used. Additionally, it was assumed the child would eat two 125 g apples with a phosmet level of 2.20 ppm. The dermal exposure was calculated to be 5734 µg with an additional 550 µg from the apples. The resulting total absorbed phosmet was 898 µg or 41 µg/kg. From these calculations a worst case exposure scenario from the Apple Maggot Eradication Program would approximate half the average daily release from a pet collar.

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