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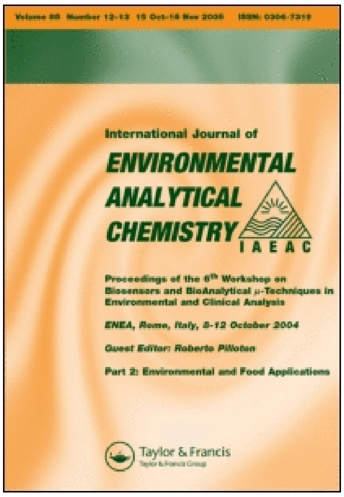

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B. Waymann[a]; H. Rudel[a]
[a] Fraunhofer-Institut für Umweltchemie und Okotoxikologie, Schmallenberg, FRG

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INFLUENCE OF AIR VELOCITY, APPLICATION DOSE, AND TEST AREA SIZE ON THE VOLATILISATION OF LINDANE

B. WAYMANN and H. RÜDEL

Fraunhofer-Institut für Umweltchemie und Ökotoxikologie, P.O. Box 1260, D-57377 Schmallenberg, FRG

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The volatilisation of lindane from soil and French beans was tested in a wind tunnel under defined conditions. Volatilised lindane was determined directly by passing a partial air stream through an adsorbent. Applications of a lindane formulation onto soil and plant surfaces were performed using a moving nozzle.

Soil volatilisation experiments were conducted using different air velocities (0.4, 1.1, 1.7 m/s). At higher air velocities the volatilisation rate increased from 12% to 31% within 24 h (initial dose 100%). For plant experiments with different velocities (0.4, 1.0, 2.0 m/s) the volatilisation rate increased from 52% to 62% at the highest velocity. Additionally, as higher air velocities were applied, air concentrations of lindane during the first hour decreased from 1.61 $\mu g/m^3$ at 0.4 m/s to 0.61 $\mu g/m^3$ at 2.0 m/s.

In soil experiments with different application doses of lindane (33 mg/m^2, 117 mg/m^2) the volatilisation rate was decreased (23%) at the higher application dose in comparison to the lower dose (39%). The volatilisation rate was also influenced by the size of the treated area in the wind tunnel. From a larger soil surface (0.84 m^2) a lower amount of lindane (23%) volatilised than from a smaller surface (31% at 0.28 m^2).

KEYWORDS: Volatilisation, lindane, wind tunnel, air velocity, soil, French beans

INTRODUCTION

Since the importance of pesticide volatilisation for the dissipation of pesticides in the environment has been recognized, several laboratory investigations were conducted in order to analyse the mechanisms involved (for reviews refer to Spencer and Cliath[1] or Taylor and Spencer[2]). However, the experimental conditions mostly were different from the situation in the field. E.g., most researchers did not use the technical formulations of the pesticides, which may influence the volatilisation behaviour. Additionally, for testing soil volatilisation, the pesticides were incorporated in the soil yielding an even distribution of the compounds in the total soil layer[3,4]. Consequently, the results probably are not relevant under field conditions.

Since 1992, the Federal Biological Agency (Biologische Bundesanstalt, BBA) has required data on the volatilisation potential of pesticides for risk assessment within the registration process in Germany[5]. Therefore, new interest has been focused on experiments investigating this feature under more realistic conditions. Several large-scale systems for the determination of volatilisation from soil and whole plants or plant stands have been presented recently[6,7,8,9,10,11]. The main emphasis was placed on model chamber systems[7,8,10,11] where climatic parameters are controlable. In the present study, our wind tunnel system[8] was used for the investigation of experimental parameters which influence the volatilisation rate. Some of these parameters have not yet been recognized.

In Germany, the experimental requirements for the performance of volatilisation studies with pesticides for registration purposes are laid down in a guideline[5]. For model chamber systems, the following standard parameters are recommended: air velocity > 1 m/s, temperature 20°C, relative air humidity 35–50%. Additionally, the experiments had to be conducted as close as possible to field conditions. Therefore, in this study

- all applications were performed using a moving nozzle which is also licensed for field applications,
- aqueous preparations of formulated lindane were sprayed onto soil and plant surfaces, and
- after soil applications the pesticide spray was not incorporated into the soil layer but instead, used directly.

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MATERIALS AND METHODS

A detailed description of the wind tunnel system and the performance of the experiments was published by Rüdel und Waymann[8]. The reproducibility of the data obtained with this system was confirmed by two volatilisation tests with lindane from plant surfaces under identical conditions. The difference between the volatilisation rates determined in both tests was below 1% absolute and below 2% relative[8].

Soil volatilisation tests were performed with sieved silty sand soil (75–79% sand, 1.1–1.5% organic carbon content; origin: Neustadt a.R., FRG). During the tests, a 3 cm-layer of the soil was kept at approximately 60% of the maximum water holding capacity by underlaying with a layer of wet burnt clay granules in stainless steel bowls with 0.14 m^2 surface area[4].

For the plant volatilisation tests, French beans (Phaseolus vulgaris) were grown in a climate chamber for four to six weeks (growth stage: blooming or first fruit bearing; height 30–40 cm). In each stainless steel bowl of 0.09 m^2 surface area and 10 cm height eight bean plants were planted.

In compliance with agricultural practice, the pesticide applications were performed using a moving nozzle (Teejet 8001EVS from Spraying Systems, Hamburg, FRG). The application equipment was built up following a description given by Kubiak *et al.*[11]. The distribution of the spray droplets on the target area was estimated visually. For this purpose, test applications of an aqueous solution of black ink were conducted on adsorbent paper. Due to border effects (uneven distribution of the spray drops at the edges), only a maximum area of 1 m^2 in the centre of the apparatus turned out to be suitable for the applications of the soil and plants. The equipment is flexible for treatments on target areas of different sizes (0.3–1 m^2).

Table 1 Volatilisation of lindane from soil surfaces within 24 h. Effects of different application doses, soil surface areas, and air velocities.

experiment	*S1*	*S2*	*S3*	*S4*	*S5*
soil surface area in the model chamber [m^2]	0.28	0.28	0.28	0.84	0.84
total volume sprayed [l/ha]	83	70	87	73	82
applied dose:					
lindane [mg/m^2]	128	112	139	117	33
Nexit fl. SC [l/ha]	1.59	1.40	1.74	1.46	0.41
air velocity [m/s]	0.4	1.1	1.7	1.1	1.1
volume stream [m^3/h]	1340	2700	4880	2880	2880
relative air humidity [%]	50	49	49	52	49
temperature [°C]	20	20	20	20	20
lindane in the model chamber [mg]	35.8	31.4	39.0	98.1	27.5
lindane concentration during 1. h [$\mu g/m^3$]	0.19	0.17	0.15	0.35	0.32
volatilised over 24 h [%]	12	31	31	23	39
total lindane mass flow [mg/m^2 in 24 h]	15	35	43	27	13

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The applications were performed by spraying the aqueous pesticide preparation on the target area where the bowls with soil or plants were placed. All soil applications were conducted using a pressure of 2.4 bar, a nozzle speed of 4 km/h, and a spraying volume corresponding to 70–87 l/ha (Table 1). The respective values for all plant applications were: nozzle speed 1 km/h, application pressure 2.0 bar, and spraying volume corresponding to 353–412 l/ha (Table 2). The manufacturer stated a mean volumetric droplet diameter of approximately 500 µm for the EVS8001 nozzle at a pressure of 1–3 bar.

Between two and six soil bowls (total area 0.28–0.84 m^2) or four plant bowls (total area 0.36 m^2) were lined up in the centre area of the application device. Immediately after treatment, the bowls were transferred into the volatilisation chamber of the wind tunnel. The arrangement of the sample bowls in the wind tunnel is shown in Figure 1.

The formulation used was Nexit flüssig SC (Shell Agrar, Ingelheim, FRG) with 800 g lindane per litre. The standard application doses for the soil tests were 110–140 mg lindane per m^2. The air velocities applied were 0.4 m/s, 1.1 m/s and 1.7 m/s over a treated test area of 0.28 m^2. Further experiments were conducted using a larger test area in the wind tunnel (0.84 m^2; air velocity 1.1 m/s). The influence of the application dose on the volatilisation rate was tested for lindane doses of 33 mg/m^2 and 117 mg/m^2 (test area 0.84 m^2; 1.1 m/s). The application doses for the volatilisation tests with beans varied between 141 and 165 mg lindane per m^2.

Table 2 Volatilisation of lindane from plant surfaces (french beans) within 24 h. Effect of different air velocities.

experiment	*P1*	*P2*	*P3*
surface of plant bowls in the model chamber [m^2]	0.36	0.36	0.36
total volume sprayed [l/ha]	412	390	353
applied dose:			
lindane [mg/m^2]	165	156	141
Nexit fl. SC [l/ha]	2.06	1.95	1.77
air velocity [m/s]	0.4	1.0	2.0
volume stream [m^3/h]	1370	2780	4670
relative air humidity [%]	49	47	45
temperature [°C]	20	20	20
lindane in the model chamber [mg]	59.4	56.1	50.9
lindane concentration during 1. h [$\mu g/m^3$]	1.61	0.84	0.61
volatilised over 24 h [%]	52	55	62
total lindane mass flow [mg/m^2 in 24 h]	86	86	87

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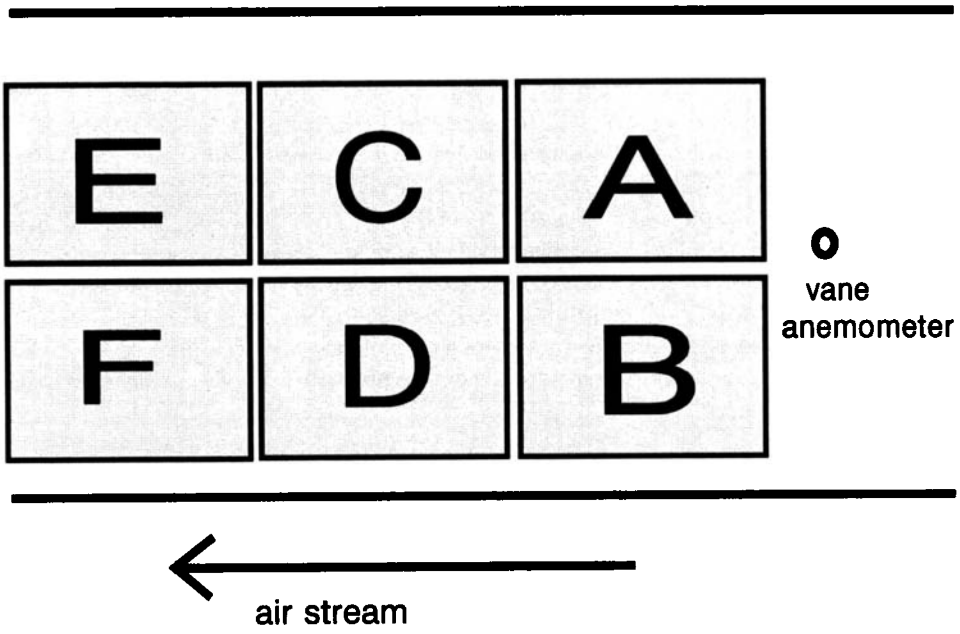

Figure 1 Arrangement of the soil bowls in the wind tunnel. For the different experiments between two (i.e. only bowls A and B) and six sample bowls (bowls A to F)) were used.

The actual lindane amount applied onto the bowls was calculated by subtracting the application losses (residues determined in the nozzle and outside the bowls) from the total lindane amount in the aqueous preparation used for application.

The volatility of lindane was determined by direct measurement of the amount of volatilised substance. This was achieved by trapping organic compounds on polyurethane foam (PUF) out of a partial stream (approximately 1–5% of the main air stream). The main air stream volume was determined following the protocol of an appropriate guideline[12] as described elsewhere[8], while the partial stream volume was quantified using a rotary piston gas meter. After sampling, the PUF plugs were extracted with toluene[13,8] and the extracts were analysed by gas chromatography[8]. Volatilisation was measured for five or six intervals between 0 and 29 h after treatment (Table 3); for a better comparison all data were interpolated for the 24 h volatilisation rates using a calculation method described by Timme *et al.*[14] (data given in Tables 1 and 2). The adsorption of volatilised compounds to the surface of the test system was investigated in control experiments. Due to the surface material (mainly stainless steel) and due to the advantageous ratio of a low surface area of the test system (approximately 10 m^2) to the high total volume stream (at least 1300 m^3/h), the adsorption observed was below 0.1% of the pesticide amount applied.

During each test the air velocity was monitored at the entrance of the model chamber using a vane anemometer (diameter 10 cm) which was installed at a height of approximately 5–10 cm above plant or soil surfaces, respectively. Additional measurements using a thermoanemometer probe were performed to characterize the air flow through the model chamber of the wind tunnel. At a volume stream of 2800 m^3/h, the air velocity measured at the entrance of the empty model chamber was 1.2 ± 0.2 m/s (mean value ± standard deviation from a grid of 9 measuring points).

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RESULTS AND DISCUSSION

In this study, the volatilisation of surface-applied lindane from soil and French beans (Phaseolus vulgaris) within 24 hours after application was determined. For a better comparison of different doses and to follow the BBA-guideline[5], all volatilisation data are expressed as percent lindane volatilised within 24 h (in relation to the initial dose of 100%). The complete volatilisation rate data from all samplings are listed in Table 3.

Table 3 Volatilisation rate (% of applied) versus time for soil and plant experiments.

	volatilisation rate for different time intervals [%] (summarized data)					
experiment	*0–1 h*	*0–3 h*	*0–5 h*	*0–7 h*	*0–23 h*	*0–29 h*
S1	0.7	1.9		4.3	11.0[d]	14.2
S2	1.5	4.3	8.4[a]	26.9[c]	31.2[e]	35.0
S3	1.9	5.4		11.4	28.5[d]	35.2
S4	1.0	3.1		6.1[a]	20.8[d]	26.7
S5	3.3	9.2	14.3	20.8[b]	38.1	42.5
P1	3.7	13.2		23.1[b]	49.5	57.8
P2	4.2	11.7		23.3	54.1	60.1
P3	5.6	15.6		28.1	61.4	67.7

Differing sampling intervals: a, 0–6 h; b, 0–8 h; c, 0–21 h; d, 0–22 h; e, 0–25 h.

The results of a set of soil experiments at different air velocities revealed that the air velocity had a strong influence on volatilisation from soil (Table 1). For soil experiments S1–S3, the volatilisation rate within 24 h increased from 12% (velocity 0.4 m/s) to 31% (1.1 m/s and 1.7 m/s).

When the air velocity was low (0.4 m/s), volatilisation seems to be limited by the capacity of the air to take up the volatilised lindane. Due to the lower exchange rate of the air in the wind tunnel at low air velocities, the air concentration of lindane was higher as compared to higher velocities. However, the mass flow expressed as mg lindane per hour was higher under high air velocity conditions (Table 1).

When sufficient pesticide was accessible for volatilisation, the transport of the lindane enriched air from the soil surface seems to be the speed controlling step of the whole process. The competitive process of the volatilisation is the adsorption of lindane by the soil.

The air volume relevant for the saturation with pesticide was not the total air stream passing through the wind tunnel. In the air conditioning part of the wind tunnel system before entering the model chamber, the air stream was straightened by a honeycomb grid[8]. Due to this device, vertical mixing of the air in the model chamber was very low. Therefore, only a thin layer directly above the soil could be enriched with lindane. After passing the treated area, the air was homogenized by a turbulence. Since the air sampling system was placed behind the turbulence, the lindane concentrations in the air determined were approximately two orders of magnitude lower than the theoretical saturation concentrations that could have been reached in the air directly above the treated area.

The volatilisation tests with treated soil surface areas of different sizes in the wind tunnel clearly show that this parameter influenced the volatilisation rate (Table 1). In experiments S2 and S4 an increase of the soil surface area by a factor of three only caused an increase of the lindane concentration in the air by a factor of not more than approximately 2 (application doses were identical with 1.40 l/ha and 1.46 l/ha). For the larger test area, a volatilisation rate of only 23% of the initial amount was measured compared to 31% for the smaller area (within 24 h). This seems to indicate that the air stream passing directly above the soil surface was saturated with lindane after a distance of a few cm of the soil surface. In this case, the absorption capacity of the air was already nearly reached, when passing over the more lee-side part of the soil surface. A possible interpretation of the observed effects is, that only a limited amount of air got in contact with the treated surface, and that the saturation of this volume of air was the limiting factor for the volatilisation of lindane. Therefore, with larger treated areas the air got in contact with a larger surface and the air concentration increased, but since the air was already enriched with pesticide vapour the concentration did not increase by the same factor as the soil surface (surface area 0.28 m^2: lindane concentration during the first hour 0.15–0.19 $\mu g/m^3$; surface area 0.84 m^2: lindane concentration 0.32–0.35 $\mu g/m^3$).

Investigations of the influence of the application dose on soil volatilisation (experiments S5 and S4) show that at a lower application dose, the amount volatilised (calculated as % of the initial amount) with 39% was higher than with the higher dose (23% volatilisation within 24 h). However, if volatilisation is expressed in terms of mass flow, a higher volatilisation was observed for the test using the higher lindane dose (13 mg/m^2 for the low dose, 27 mg/m^2 for the high dose).

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This effect could also be interpreted as result of the saturation with lindane of the air mass which was in contact with the soil. The uptake by the air was hindered, although more lindane was accessible for volatilisation. Therefore, it may not always be meaningful to test the soil volatilisation of substances with a high vapour pressure at the highest application dose. The absolute amount volatilised under these conditions is higher (larger mass flow), but the volatilisation rates expressed as a percent value (% volatilised in relation to the initial dose) may be lower than for lower doses. Since the threshold value of the BBA-guideline[5] is defined as a percent value (20% volatilisation of the amount applied within 24 h), this factor may be important for the evaluation of volatilisation experiments in special cases.

Spencer *et al.*[4] also investigated the volatilisation of lindane out of soil. They found that the volatilisation process during a period of 14–23 days was controlled not by the thickness of the air boundary layer above the soil but by the adsorption of lindane to the soil (control of the volatilisation within the soil). A comparison with our results is not possible, because Spencer *et al.*[4] tested the volatilisation of lindane after incorporation into the soil while for the volatilisation experiments described here surface-applied lindane in formulation was used. Further, the duration of the experiments was shorter (29 hours maximum).

The plant experiments were performed at higher application doses. In contrast to the soil experiments, the air stream in the wind tunnel was turbulent above and in the plant stand. This was caused by the unregular surface of the plant stand (plants of different height; leaves at different angles to the air stream). Therefore, the amount of air which came in contact with the surface was higher. This also became obvious from the higher lindane concentrations in the air during the plant tests than during the soil tests (up to 10 times higher; Table 2). Another difference between the soil and plant tests was the application of the spray drops onto a dry plant surface where the water evaporates relatively fast. Additionally, the sorption mechanism on plant surfaces is different from that on soil surfaces. Combined, the differences resulted in higher volatilisation rates for the plant tests than for the soil experiments.

The effect of different air velocities on lindane volatilisation from plants (experiments P1–P3; Table 2) was analogous to the results of the soil tests. Lower air velocities caused higher lindane concentrations in the air and lower volatilisation rates. The air concentrations of lindane determined during the first hour of each experiment were clearly dependent on the air velocity: the lindane concentration decreased from 1.61 $\mu g/m^3$ at 0.4 m/s to 0.61 $\mu g/m^3$ at 2.0 m/s (Table 2). The difference in the volatilisation rates between the extreme velocities was 10 percentage points (62% at 0.4 m/s in comparison to 52% volatilisation at 2.0 m/s; within 24 h).

CONCLUSIONS

The lindane volatilisation from soil and plant surfaces– expressed both as rate and in form of air concentrations– was strongly influenced by the air velocity above the treated area. At lower velocities, volatilisation seemed to be limited by the capacity of the air to take up volatilised lindane.

For the evaluation of volatilisation experiments more attention should be paid to the size of the treated area in the test system and to the application rates. Our results for lindane

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suggest that in some cases volatilisation may be overestimated, if too small an area is used. On the other hand, low volatilisation rates may be achieved by performing experiments using the maximum application dose on a large surface.

If these results are confirmed for other compounds, we suggest to amend the BBA-guideline[5] to include the size of the test area as one important parameter. To find realistic experimental conditions for the size of the treated area, field experiments are necessary. However, by testing the volatilisation rate in our wind tunnel system under constant conditions, ranking of different pesticides with respect to their volatilisation potential is possible.

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