EMISSION OF PESTICIDES INTO THE AIR[1]

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Abstract. During and after the application of a pesticide in agriculture, a substantial fraction of the dosage may enter the atmosphere and be transported over varying distances downwind of the target. The rate and extent of the emission during application, predominantly as spray particle drift, depends primarily on the application method (equipment and technique), the formulation and environmental conditions, whereas the emission after application depends primarily on the properties of the pesticide, soils, crops and environmental conditions. The fraction of the dosage that misses the target area may be high in some cases and more experimental data on this loss term are needed for various application types and weather conditions. Such data are necessary to test spray drift models, and for further model development and verification as well. Following application, the emission of soil fumigants and soil incorporated pesticides into the air can be measured and computed with reasonable accuracy, but further model development is needed to improve the reliability of the model predictions. For soil surface applied pesticides reliable measurement methods are available, but there is not yet a reliable model. Further model development is required which must be verified by field experiments. Few data are available on pesticide volatilization from plants and more field experiments are also needed to study the fate processes on the plants. Once this information is available, a model needs to be developed to predict the volatilization of pesticides from plants, which, again, should be verified with field measurements. For regional emission estimates, a link between data on the temporal and spatial pesticide use and a geographical information system for crops and soils with their characteristics is needed.

Keywords: air quality, application technique, emission factor, emission model, glasshouse, measurement method, pesticides, regional emissions, spray drift, volatilization

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1. Introduction

In agriculture, pesticides are applied to the soil or to a crop. The application can be made using different techniques, which depends on the formulation type, the pest to be controlled and the timing of the application. The pesticide can be injected into the soil for use as a fumigant or sprayed onto the soil surface, possibly followed by it's incorporation into the soil top layer. In addition, seeds are sometimes treated with pesticides prior to planting. Crops can be sprayed, for example, with boom sprayers, tunnel sprayers or by aerial application or treated with systemic pesticides.

During the application, a fraction of the dosage is lost to the atmosphere. The application loss is defined as the total fraction of the dosage applied that does not reach the target area. The portion of this loss, in the form of droplets moving off-target (crossing the field border) through the air is referred to, here, as spray drift. Spraying pesticides through spray nozzles produces a spectrum of droplet diameters. Those droplets of smallest size within the spectrum are prone to become lost as spray drift. The larger droplets are carried away by the wind and may be deposited either just outside the target area or at some downwind distance. During aerial transport, the diameter of the droplets can decrease through evaporation of the carrier formulation, and/or the pesticide, and when the diameter is sufficiently small, the droplets or particles can remain airborne with the potential for long-range transport. The fine droplets (diameter less than 100 μm) may evaporate very rapidly. The evaporated component can also travel long distances. Loss during application through spray drift is not usually related to the physico-chemical properties of the pesticide itself, but is largely dependent on the application method, properties of the formulation and carrier (diluent) used and the environmental conditions.

In some cases it is not necessary to spray the whole cropped area (row application, spot spraying). Herbicides can be applied by spot spraying, which means that the sprayer (one or more nozzles) is only turned on when a weeded area is detected. This application method can result in a much lower total loss by both spray drift and volatilization than if the whole field were treated. The extent of reduction depends on the fraction of the field surface sprayed. The use of other formulation types, e.g. solid granules, will eliminate spray drift altogether.

After the pesticide is deposited on the target area, a fraction of this mass volatilizes in the course of time. Some of this volatilization loss may occur during the application period, because no application is instantaneous (e.g. about 15 min per hectare for soil surface spraying; fumigant application is much slower). Consequently, it is difficult to accurately assess the fraction of the dosage not reaching the target area through flux measurements, as the volatilization process can occur as soon as the surface has been sprayed and some time elapses before measuring equipment can be put in place. In particular for the more volatile compounds, the fraction of the dosage lost to the atmosphere by volatilization from the target surface can be substantial during the period of application. Majewski and Capel (1995) present an overview of the sources of emission, measurements and processes determining the emission of pesticides into the atmosphere.

The pesticide applied may be transformed into degradation products which are more toxic than the parent compound (e.g. the transformation of some organophosphorus insecticides to their oxygen analogs). The environmental effect of relevant transformation products should be included in any risk assessment. Moreover, the transformation products may have a much higher vapor pressure than the parent compound (e.g. the formation of the active compound methyl isothiocyanate from metam-sodium). For example, when assessing the health risks from the use of metam-sodium, the volatilization of methyl isothiocyanate and its subsequent dispersion in air should also be assessed. However, there is little information on the rate and the extent of the formation of transformation products for many of the pesticides applied to plants and soils. The transformation products may depend on soil and environmental conditions, which make it very difficult to predict under general conditions. In addition, much more specific chemical, soil and environmental information would be required. Under Directive 91/414/EEC of the European Union (EU, 1991), industry now has to consider relevant metabolites (transformation products) in the risk assessment process. Such information could be obtained using laboratory studies with radiolabelled compounds. Moreover, guidance is needed on the selection of relevant transformation products to be included in the risk assessment.

In this paper a review is presented of the research on the emission of pesticides into the air resulting from applications in agriculture. In addition, the information needed to improve the ability to evaluate the emission potential of pesticide is defined.

2. Emission during Application

To date, many measurements of pesticide emissions during application have been done on the spray drift of droplets or particles. In the United States, a consortium of agrochemical companies has established a Spray Drift Task Force, to collect drift data for current application techniques. These data have been used to develop an assessment method for aerial spray drift (Bird *et al.*, 1995a). In Germany, data on spray drift to adjacent watercourses for various techniques and crops have been collected by Ganzelmeier *et al.* (1995). In the United Kingdom (UK) a database on spray drift has been compiled by the Central Science Laboratory, which is used to underpin regulatory risk assessment, including the recent Local Environmental Risk Assessment for Pesticides (LERAP) arrangements (A Gilbert, personal communication 1999). The UK database has also supported development of the British Crop Protection Council (BCPC) Spray / Nozzle classification scheme (Doble *et al.*, 1985). In particular, the ability to measure a drift potential factor for different spray qualities under controlled conditions in the wind tunnel (Miller *et al.*, 1993) and to reconcile these results against field measured drift levels has enabled a revision of the BCPC scheme to include a drift potential factor (Southcombe *et al.*, 1997). As well as the scheme for classification of size spectra and drift potential of sprays, a more comprehensive basis for classification of application equipment and techniques by hazard has been made (Parkin *et al.*, 1994). Many measurements of spray drift to ditches just downwind of the treated field have been made in the Netherlands (Huijsmans *et al.*, 1997). A draft protocol for the measurement of drift is being discussed by the International Organization for Standardization (ISO, 1999), which would enable a (better) comparison of drift data from various sources that were obtained with different methods.

In most cases drift measurements have been limited to the determination of the mass of pesticide deposited on the surface adjacent to the treated field and on measurements of droplets in the air close to the ground leaving the target area using passive drift collectors. Little is known about the total fraction of the dosage which does not reach the target area. However, data from field experiments indicate that the emission during application can typically range from a few percent (e.g. Maybank *et al.*, 1974; Nordby and Skuterud, 1975; Grover *et al.*, 1988; Glotfelty *et al.*, 1990) to 20-30% (e.g. Nordby and Skuterud, 1977; De Heer *et al.*, 1985; Bird *et al.*, 1995b). In some cases it may be as high as 50% or even more (Warren, 1972; Armstrong, 1973; Wood and Stewart, 1976; Symons, 1977). The amount of pesticide not reaching the target area depends strongly on the application technique, the formulation and the environmental conditions (wind speed, temperature, humidity, atmospheric stability). Although the overall range for this loss is wide, it can be substantially narrowed for a given combination of application technique and environmental conditions.

A portion of the pesticide that does not reach the target area consists of gas-phase pesticide and small droplets or particles (aerosols) which are or have become so small that they cannot be captured effectively by drift collectors. For this fraction, measurement may be possible, but would require some form of iso-kinetic sampling (the flow rate of air into the orifice of the sampling unit is approximately the same as the flow rate of air around it).

3. Emission from Soil

After being deposited on the target area (soil surface and/or plant surface) or after injection or incorporation into the soil, volatilization is one of the processes which can affect the fate of the pesticide. The dominant factors that affect volatilization are the physico-chemical properties of a pesticide (e.g. vapor pressure, water solubility), its persistence in the soil, and environmental conditions (soil and air temperature, soil water content and soil organic matter). Some pesticides can occur both in a neutral and an ionic form in the soil solution. Depending on the pK_a of the pesticide, the volatilization from soil can also be substantially affected by the soil pH (Müller *et al.*, 1998). To illustrate the range in values for physicochemical properties of pesticides, data on basic properties for some common pesticides are listed in Table 1. Depending on the properties of the pesticide, soil and environmental conditions, cumulative volatilization losses range from a few percent to 50 % of the dosage (e.g. Glotfelty *et al.*, 1989; Majewski *et al.*, 1993; Bor *et al.*, 1995; Stork *et al.*, 1998a), or even more in some cases (Glotfelty *et al.*, 1984). The emission of fumigants from soil into the air is strongly affected by the application technique and volatilization losses can be as high as 90% of the dosage (e.g. Gan *et al.*, 1997; Gan *et al.*, 1998).

Soil fumigants form a class of pesticides that are unique since their vapor pressures are many orders of magnitude higher than those of other pesticides. Due to their high volatility, most of the dosage is lost to the atmosphere if no appropriate measures are taken, e.g. deeper injection, covering soil surface with plastic sheeting, soil compaction of the top layer or forming a water seal at the soil surface (Jury *et al*, 1997; Yates *et al.*, 1997b). For example, Yates *et al.* (1996) measured a loss of 64% of the dosage of methyl bromide after injection at a depth of 0.25 m and covering the soil surface with a polyethene film. Deeper injection of the fumigant into the soil results in a higher residence time in the soil. Consequently, a greater part of the dosage may be transformed into non-volatile compounds and volatilization loss is reduced (Yates *et al.*, 1996; Yates *et al.*, 1997a; Yates *et al.*, 1997b; Gan *et al.*, 1997; Gan *et al.*, 1998). Soil compaction (e.g. by rolling) and the supply of water to the soil reduce the air-filled porosity, thereby decreasing the diffusion of the pesticide through the gas phase of the soil system (Gan *et al.*, 1996, Gan *et al.*, 1998). A more drastic measure is wetting the surface soil (common practice in California), which results in a water cap on top of the soil profile.

TABLE 1

Physico-chemical properties of some common pesticides. Values for 25 °C if not stated otherwise. K_{om} = coefficient for the adsorption on organic matter.

Compound	Vapor pressure (Pa)	Water solubility (mg L^{-1})	K_{om} (L kg^{-1})
Atrazine	0.000039[†]	33[¶]	70[§]
Alachlor	0.0021[†]	242[†]	117[§]
Chlorpyrifos	0.0027[†]	1.4[†]	293[§]
Cyanazine	0.0000002[¶]	171[†]	55[§]
EPTC	4.53[‡]	375[†]	61[§]
Methyl bromide	190000[¶]	17500[¶]	2.4[§]
Methyl isothiocyanate	1700[‡‡]	8900[††]	3[§]
Metolachlor	0.0042[†]	488[†]	103[§]
Parathion	0.00089[¶]	11[¶]	1746[§]
Tri-allate	0.016[†]	4[†]	1164[§]
Trifluralin	0.0061[†]	0.18-0.22[†,¶¶]	3775[§]
2,4-D	0.000011[¶]	311[†,§§]	26-230[§,¶¶]

[†]Tomlin, 1997; [¶]at 20 °C, except for atrazine: 22°C, Tomlin, 1997; [§]Linders et al., 1994; [‡]Baker et al., 1996; [‡‡]at 20 °C, Siebering and Leistra, 1979; [††] at 20 °C, Smelt and Leistra (1974); [§§] at pH 1; [¶¶]value depends on pH.

Apart from the fumigants, there are other pesticides whose volatility requires their incorporation into the soil (e.g. tri-allate, vapor pressure of 16 mPa at 25 °C). However, most of the soil-applied pesticides are less volatile than tri-allate, and they can be sprayed onto the soil surface. Soil-incorporation may also be required to eliminate effectively harmful insects in the topsoil.

Another emission pathway for pesticides into the atmosphere occurs when pesticides are sorbed to soil particles and entrained into the atmosphere on wind blown particles (Glotfelty *et al.*, 1989). There are few data on the significance of this transport pathway and on the quantitative effects of soil and environmental factors that influence this process.

3.1 MEASUREMENT METHODS

Several methods have been used to determine the volatilization of fumigant after injection into the soil in the field, such as the aerodynamic-gradient method (Majewski, 1995; Yates *et al.*, 1996; Yates *et al.*, 1997b), a theoretical profile method (Yates *et al.*, 1997b), the integrated horizontal flux method (Yates *et al.*, 1997b) and the box method (e.g. Smelt and Smidt, 1994). The temporal uncertainty in the volatilization rate can be a factor of two. There is less uncertainty in the total loss of the fumigant into the air; this uncertainty has been estimated to be about 10% of the dosage (Yates *et al.*, 1996). The uncertainty in the total loss is a bulk uncertainty which is integrated over time. Therefore, errors are averaged out to some degree.

For pesticides which are sprayed on the soil surface or incorporated, the rate of volatilization can be measured in the field with different micro-meteorological methods, such as the aerodynamic-gradient (Parmele *et al.*, 1972, Glotfelty *et al.*, 1984; Majewski *et al.*, 1993), the Bowen-Ratio (except in moisture limiting situations; Majewski *et al.*, 1990), the theoretical profile (Majewski *et al.*, 1989; Whang *et al.*, 1993), the integrated horizontal flux (Glotfelty *et al.*, 1990, Majewski *et al.*, 1990) and the Eddy accumulation (Majewski *et al.*, 1993) methods. Majewski (1999) presents an overview of these methods and discusses their use and limitations. The difference in the measured total loss by volatilization between these methods is generally less than 20 - 25% (Majewski *et al.*, 1990). At a specific time, the measured volatilization rates resulting from using these methods may differ by a factor 2 - 5 from each other. Even for the same method such differences may occur. As weather conditions change with time, the volatilization loss can be expected to be different for each field experiment. Both temperature and soil moisture conditions at the soil surface have a great effect on the volatilization rate (e.g. Spencer and Cliath, 1974, Taylor and Spencer, 1990). For example, upon re-wetting of the soil surface, the volatilization rate can increase by a factor 2 - 5 (Spencer and Cliath, 1973; Spencer *et al.*, 1982; Spencer and Cliath, 1990). The variability in weather conditions makes it difficult to study the effect of only one factor on the volatilization rate. It should be noted that many of the micrometeorological methods that rely on measurements of horizontal wind speed produce highly uncertain flux estimates when the winds are calm. In fact, a zero wind gradient produces an unrealistic Richardson's number that affects the stability correction and the flux. Most of these methods were developed for turbulent conditions and, probably, should not be used when turbulence is practically absent (nighttime stable/inversion conditions).

In pesticide volatilization field experiments, measurements are mostly done under ideal conditions. The typical experimental field is situated in a flat area with no significant wind obstacles in the vicinity of the field. In practice, however, agricultural fields are also found in hilly areas. Hills and windbreaks may have an effect on pesticide volatilization but these factors have yet to be fully investigated in field experiments.

Field volatility experiments are costly, and volatilization chambers and wind tunnel systems containing a lysimeter (semi-field systems) have been developed to simulate field applications at a lower cost. In these systems, weather variables such as wind speed, air temperature and humidity and solar radiation can be controlled, so field conditions can be approximated by simulating the weather conditions as closely as possible (Stork *et al.*, 1998a; Stork *et al.*, 1998b, Kubiak *et al.*, 1993). The advantage of semi-field systems is that experiments can be reproduced and the factors affecting the volatilization process can be studied. Radio-labeled pesticides can be used to facilitate the analysis of the parent compound as well as any transformation products that are formed. In the volatilization chamber all terms of the mass balance of the pesticide in the soil and air compartments can be quantified, and the experimental time period can be easily lengthened (e.g. hours, days, weeks), therefore, the temporal variation in the volatilization rate as well as the total volatilization for a single application can be quantified.

The volatilization chamber method also has limitations. Substantial differences may occur between the atmospheric conditions in the field and the simulated weather conditions in the volatilization chamber. First, air turbulence in the chamber is likely to be different from that in the field (e.g. no larger scale turbulence). Further, there is always a flow of air in the chamber, so the situation in the field in which there is no wind cannot be simulated. Secondly, photo-degradation of pesticide at the soil surface can be investigated, but as air travel distances in the chamber are very short, photo-degradation in air cannot be effectively studied. Thirdly, the flow of air through the chamber might result in a light vacuum in the volatilization chamber, which might cause an advective transport component and result in measurement error unless similar gas advection occurs elsewhere in the field (i.e. not just in the chamber). A vacuum can be avoided by installing a pressing and a drawing transport blower (Maurer and Kubiak, 1994). The air pressure entering the chamber should be the same as that leaving the chamber. The influence of comparatively small pressure changes with time on the volatilization loss of pesticide is unknown.

Experimental results from volatilization chambers can be equivalent to results obtained from field studies, provided field soil and weather conditions are simulated accurately. Kubiak *et al.* (1995) simulated field applications of isoproturon and methyl parathion in the volatilization chamber and they measured the volatilization in the chamber system during the first 24 hours after application. Volatilization rates were not measured in the field, but there was a good correlation between the pesticide residues on the plants in both systems. However, when simulating field applications in the semi-field system, significant differences between the conditions in this system and those prevailing in the field can occur that may make evaluation of the experimental results more difficult. These differences can be due to differences in the initial penetration of the pesticide, differences in the soil surface temperature (caused by shielding of the soil surface from solar radiation in the semi-field system), and by soil puddling and compaction due to the higher intensity of the sprinkling events in the semi-field system than that of rainfall in the field. For example, due to a combination of such differences, Stork *et al.* (1998a) measured differences between the volatilization rates in the field and those in the wind tunnel system of up to about a factor 10.

The assessment of the volatilization potential of a pesticide using a volatilization chamber has been adopted by the Biologische Bundesanstalt (BBA, Germany) in their registration procedure (BBA, 1990). If the trigger value for the hydrolysis or photolysis half-life for the pesticide is exceeded, then the cumulative volatilization of the pesticide must be determined for the 24-hour period after application. Then further assessment, involving the subsequent stability of the pesticide in air, has to be made if a trigger value of 20% loss is exceeded. For the conditions mentioned in the guideline, the error in the cumulative volatilization measured was estimated to be ± 5%.

To evaluate the various methods to assess the volatilization potential of a pesticide after application, 18 laboratories (in Germany, Switzerland and England) measured the volatilization of three pesticides during the first 24 hours after application under the conditions required by the BBA (Walter *et al.*, 1996). The vapor pressures of these pesticides were $2 \cdot 10^{-6}$, $2 \cdot 10^{-4}$ and $3.5 \cdot 10^{-3}$ Pa (at 20 °C). The water solubilities (at 20 °C) of these compounds were 0.001-0.25 (only range reported), 55 and 4.3 mg L^{-1} respectively. For all the methods used in this inter-laboratory comparison (a ring test in which different laboratories measure the volatilization of a specific pesticide under conditions specified in a guideline), the initial volatilization loss for the compound with the lowest vapor pressure was well below the trigger value of 20%. For the other two compounds, some studies resulted in a value higher than the trigger value and some resulted in a lower value. For those compounds, the method to be used must be evaluated in more detail to assess whether it can give a representative and reliable value for the initial volatilization loss. Further guidance on the use of the method may also be needed.

4. Emission from Crops

The dominant factors that influence the volatilization of pesticides from crops are the physico-chemical properties (see Table 1), the persistence on the plant surface and the environmental conditions (atmospheric stability, wind, temperature and humidity). The persistence on the leaf surface depends on the various dissipation processes, such as photo-degradation (e.g. Liang and Lichtenstein, 1976, Devlin *et al.*, 1987), wash-off from the leaves by rainfall or irrigation (McDowell *et al.*, 1987; Willis *et al.*, 1992), and uptake of the pesticide by the plant leaves. Case studies on photodegradation have been briefly discussed by Leistra (1998). The fate processes on the leaf surface that affect the pesticide are not well understood. Some important factors include the nature of the plant, the age of the plant, the stage of development of the plant (e.g. seedling vs. fully mature), the characteristics of the leaf surface (e.g. waxy cuticle type), and the density and the height of the canopy (Taylor and Glotfelty, 1988). Furthermore, little is known on the effect of the formulation type on the waxy layer. The waxy layer may be partially destroyed when using a specific formulation.

A substantial fraction of the mass of pesticide may be sorbed onto the waxy layer of the plant leaves or, if no waxy layer is present, the sorption may be on other organic materials at the leaf surface layer. Volatilization can be affected by the initial distribution of the pesticide on the leaf surface. Depending on the formulation used for spraying, the pesticide may be uniformly distributed over the leaf surface or it may be concentrated in a number of areas that represent a fraction of the total leaf surface. Therefore, more research is needed on the factors that influence the volatilization of pesticides from plant surfaces. Volatilization experiments have been done for vegetative surfaces (e.g. Turner *et al.*, 1977; Taylor *et al.*, 1977; Grover *et al.*, 1985, Breeze *et al.*, 1992; Kubiak *et al.*, 1995; Van den Berg *et al.*, 1995; Smelt *et al.*, 1997, Stork *et al.*, 1998a) with cumulative volatilization losses ranging from less than a few percent to 60% or more in some cases. However, more experiments are needed to collect data on the volatilization of pesticides in the field for a range of environmental conditions and crop types.

The evaluation of pesticide emissions may be made more complex by the potential of several plant species to produce halogenated pesticides. For example, Gan *et al.* (1998) measured the production of methyl bromide by live *Brassica* plants in significant quantities (daily rates ranging from 20 to 40 ng methyl bromide per g dry plant material) as a result of uptake of Br^- by the plant roots from the soil.

4.1 MEASUREMENT METHODS

The rate of volatilization of pesticide from the plant leaves of most crop types can be measured in the field using the same micro-meteorological methods as those for the measurement of the volatilization rate from soil, e.g. the aerodynamic-gradient methods, the Bowen-Ratio method, and the theoretical profile shape methods. However, these methods will simultaneously measure any volatilization from the soil beneath the plants as well. The measurements on volatilization from a sprayed orchard are more complex because of the structure and height of the trees.

For assessment of the potential volatilization of pesticides from low crops the volatilization chamber and the wind tunnel-lysimeter methods as described in Section 3 are measurement methods that can be used at comparatively low costs. Volatilization can be studied under standard conditions or worst-case conditions. Since conditions can be controlled when chambers are used, experiments can be reproduced so the effect of the various factors governing the volatilization from plant surfaces can be studied. An advantage of the chamber method is that the sprayed surface can be limited to the plant surfaces only, since chambers sample small areas, but, small areal samples also introduce high uncertainty. Before spraying, surfaces other than the plant surfaces can be covered with paper that is removed after spraying (Kubiak *et al.*, 1993). The error in the cumulative volatilization measured with this system has been estimated to be less than 10%. The limitations of the chamber method have been mentioned in Section 3. To date, several volatilization experiments using a chamber or wind tunnel-lysimeter system have been reported (e.g. Stork *et al.*, 1998a; Müller *et al.*, 1998). However, more simultaneous field and chamber experiments are needed to verify the data obtained with the volatilization chamber.

5. Emission from Glasshouses

The use of pesticides in glasshouses can result in substantial emissions to the outdoor environment (Baas and Huygen, 1992). The rate of this emission is dependent on the ventilation rate of air in the glasshouse and the concentration of pesticide in the glasshouse air. The initial concentrations in the glasshouse air depend to a large extent on the application technique used. For example, much higher concentrations will occur when using a low-volume mister than when using a high volume technique (i.e. spray bar with 6 nozzles, pressure 1200 kPa). After application, a fraction of the mass deposited on the surfaces within the glasshouse volatilizes. Although the key processes that affect the volatilization are largely the same as those in the field, the environmental conditions in a glasshouse differ greatly, as they are much more controlled. Firstly, the short wavelength UV light (290-310 nm) responsible for many outdoor photolytic reactions may be filtered out by the glass, so photo-degradation occurring outdoors may not happen in the glasshouse. In some glasshouses, artificial light sources are used to promote the growth of the crop. Whether the light from these sources causes photo-degradation of the pesticide will depend on the UV spectrum that the lights emit. Secondly, there is little flow of air except when windows are opened. Furthermore, there is a tendency towards higher air temperature and humidity in the glasshouse compared with conditions in the field. In addition, the glasshouse may contain plastic materials, which may adsorb substantial amounts of pesticide in the glasshouse air following application (Bor *et al.*, 1994).

Because of the limited air movement in the glasshouse, limited ventilation with outside air, and relatively long residence time inside the glasshouse, the pesticide concentration in the glasshouse air can be expected to be high. The built-up of the concentration inside the glasshouse may result in a somewhat lower rate of volatilization from the plant leaves than under similar outdoor conditions which may be offset, somewhat, by higher temperature conditions inside the glasshouse.

The rate of emission of the pesticide from the glasshouse into the atmosphere can be determined by measurement of the fractional rate of ventilation of the glasshouse air (by leakage in the glasshouse structure, open windows and/or open doors) and the concentration of the pesticide in the glasshouse air. In the Netherlands, such measurements have been done to estimate the emission of pesticides into the air in a region with many glasshouses (e.g. Baas and Bakker, 1996). Models to predict the ventilation rate have been discussed by Fernandez and Bailey (1992).

The emission of pesticides from a glasshouse can be reduced in several ways. Firstly, the rate of ventilation in the glasshouse can be lowered by improvement of the structure. Secondly, comparatively high concentrations in air may be avoided by selecting an application technique with coarser spray droplets in combination with a less volatile pesticide (lower vapor pressure). The opening of windows and doors should be avoided when there are high concentrations of the pesticide in the glasshouse air, e.g. during application and the first few hours thereafter. Ultimately, emissions can even be avoided by technical means; this would necessitate the installation of a device to clean all exhaust air from the glasshouse but this would prove to be expensive. A feasibility study by Van Os *et al.* (1993) indicated that the reduction in emission out of the glasshouse by cleaning the air by carbon filters may be limited and not very practical. However, when assessing the health risk of glasshouse workers, the effect of measures reducing the emission of pesticide out of the glasshouse into the atmosphere should be taken into consideration, because a lower emission out of the glasshouse may result in greater concentrations in air prevailing in the glasshouse.

In most countries only about 1% of the agricultural area or less is under glass, so the contribution of the emission from glasshouses to air compared to the total emission of agricultural pesticides may be of limited importance. For the risk evaluation of the exposure of people living in the neighborhood of glasshouses as well as that of nearby ecosystems, the emission of pesticide from the glasshouse into the atmosphere can be an important issue.

6. Estimation and Modeling

6.1 EMISSION DURING APPLICATION

To date several spray drift models have been developed, such as AgDrift (Bird *et al.*, 1997), PEDRIMO (PEsticide DRift MOdel, Kaul *et al.*, 1996), IDEFICS (IMAG program for Drift Evaluation from FIeld sprayers by Computer Simulation, Holterman *et al.*, 1994, Holterman *et al.*, 1997) and others (Thompson and Ley, 1983; Walklate, 1992; Hashem and Parkin, 1991). The AgDrift 1.0 model has been tested and documented (Bird *et al.*, 1997). The PEDRIMO model has also been tested against measurements and a summary of these tests is given by Kaul *et al.* (1996). The IDEFICS model is being evaluated and documentation is not yet available. Although these models were not developed for the assessment of the fraction of the particles and droplets that remains air-borne and the vapor phase pesticide formed by evaporation of the droplets, they can be used to estimate this loss. To do this assessment some adjustment of the model may be required. Moreover, standardization of the definition and description of alternative pesticide application methods would promote a better use of available models and datasets (Gilbert, 1999, personal communication).

6.2 EMISSION AFTER APPLICATION

An estimate of the initial volatilization rate after spraying on the soil surface can be made using the physico-chemical properties of the pesticide: vapor pressure, water solubility and the coefficient of the sorption on organic carbon (or organic matter). A good indicator of volatilization is the effective vapor pressure of the pesticide, i.e. the vapor pressure that is in equilibrium with the concentration in the liquid phase and the mass sorbed on the soil surface. A good correlation (n =

12, $r^2 = 0.988$) between the logarithm of the ratio of the vapor pressure divided by the water solubility and the organic carbon sorption coefficient (K_{oc}) and the logarithm of the volatilization rate as measured during the first day after application, was obtained by Woodrow *et al.* (1997), although more data sets are needed to verify this method. Further, some of the data used in the correlation were obtained by residue analysis, which provides an indirect measurement of the total flux. For soil incorporated pesticides, the above mentioned ratio had to be multiplied by the application rate divided by the depth of incorporation ($r^2 = 0.93$). For surface applied pesticides, a similar approach has been made by Smit *et al.* (1997), who estimated the cumulative loss by volatilization during the first 21 days after application from the fraction of the pesticide in the gas phase of the soil system. For normal to moist field conditions r^2 was calculated to be 0.76 ($n = 22$) and, for dry soil conditions, it was 0.89 ($n = 7$). For this correlation only flux measurements were used. Both approaches take the effect of temperature on the physico-chemical properties of the pesticide into account. The uncertainty in the calculated pesticide flux density and that in the cumulative loss depends on the quality of the underlying data used in the respective correlation.

A screening-level estimate of the initial volatilization rate after spraying of the crop can be made using the vapor pressure of the pesticide. Woodrow *et al.* (1997) observed a good correlation between the logarithm of the volatilization rate and the logarithm of the vapor pressure. For the cumulative loss from plant surfaces an estimation method has been developed by Smit *et al.* (1998). Using literature data on volatilization rates from plant surfaces as measured in the field or in volatilization chambers, the best correlation was found between the logarithm of cumulative loss by volatilization (over a period of the first 7 days after application) and the vapor pressure of the pesticide ($n = 19$, $r^2 = 0.78$). The correlation between the cumulative loss and the ratio between the vapor pressure and the sorption coefficient on organic carbon was less clear ($r^2 = 0.55$).

For a more accurate calculation of the volatilization flux a model is needed which describes a pesticide's fate in the soil and the exchange with the lower part of the atmospheric boundary layer. For soil fumigants, a number of models have been used (e.g. Leistra, 1972; Jury *et al.*, 1983; Van den Berg, 1992; Baker *et al.*, 1996; Freijer *et al.*, 1996; Wang *et al.*, 1997; Wang *et al.*, 1998). In these models processes occurring in the soil are described at various levels of detail. Relevant processes are the transformation of fumigant in the soil and the diffusion and convection of fumigant in the gas phase. The transformation of fumigant is mostly described with first-order kinetics (Siebering and Leistra, 1979; Wagenet *et al.*, 1989; Leistra and Crum, 1990; Van den Berg, 1992). For the diffusion in the gas phase of the soil, several models have been used to calculate the diffusion coefficient (Troeh *et al.*, 1982; Millington and Quirk, 1960; Currie, 1965; Freijer, 1994). Convective transport of fumigant can be caused by changes in soil water content, temperature and air pressure at the soil surface. A model for air flow driven by air pressure changes at the soil surface has been described by Chen *et al.* (1995). The description of the process of exchange of soil fumigant across the soil-air interface is mostly simple. At the interface, a zero concentration of pesticide is incorrectly assumed or a thin stagnant air layer is assumed to exist through which the pesticide must diffuse (molecular diffusion), before it can enter the turbulent air. Van den Berg *et al.* (1999) compared volatilization rates of methyl isothiocyanate from soil after application of metam-sodium into the soil computed with a model based on this simple air boundary layer concept with measured data. The computed rates corresponded roughly to those measured in the field. However, the existence of a stagnant boundary layer is questionable because it precludes mass flow of air across the soil-air interface. Therefore, this concept should be considered a simplification of the processes that really occur at the boundary layer. For an adequate description, a direct coupling of the soil profile with the atmosphere is needed, which would require a sophisticated model. In a recent study on the volatilization from bare soil, Baker *et al.* (1996) developed a method to estimate the transport resistance of the boundary layer for general atmospheric conditions. This approach is rather complex (the resistance is expressed as a function of the Schmidt and Reynolds numbers) and it requires many meteorological input data. During the first day after application, the computed volatilization flux of EPTC (S-ethyl dipropylthiocarbamate) corresponded well to the measurements, but thereafter the model overestimated the volatilization flux. Further testing of the model is needed under different weather and soil conditions.

For soil surface applied pesticides, reliable models are not yet available, although a comprehensive model is being tested (Scholtz *et al.*, 1997). The initial distribution of the pesticide in the soil needs to be described adequately. Freijer *et al.* (1996) proposed the concept of a thin pesticide layer on the surface from which all pesticide is leached into the soil at the time of the first rainfall. The model should also take into account other loss processes at the soil surface, such as photodegradation and the occurrence of non-equilibrium conditions in the partitioning of the pesticide over the soil phases. The most important factors that determine the volatilization rate from soil are the soil moisture conditions and the soil surface temperature. These two variables change considerably, not only from day to day but during the course of a 24-hour period, as well. When the top few mm of the soil surface layer dries out, the moisture content can decrease below a few percent. At these low moisture conditions, pesticides are much more strongly adsorbed to soil and so the volatilization flux can be expected to be at a low level (Spencer and Cliath, 1973). Upon rewetting of the soil surface, the volatilization flux increases substantially (Spencer and Cliath, 1973; Spencer *et al.*, 1982;

Spencer and Cliath, 1990). Further research is needed to determine the importance of this process with respect to the vapor pressure of the pesticide. Temperature can have a large effect on the partitioning of the pesticide between the gas and liquid phases (e.g. the Henry coefficient). A higher temperature results in higher concentrations in the gas phase. Diurnal temperature differences can be substantial (more than 10 °C), in particular under clear skies. More research is needed on the description of the soil water content and temperature gradients and their temporal behavior in the top few mm of the soil profile to improve the volatilization model for soil surface applications of pesticide.

A model in which exchange between the air and soil compartment is described with the simple concept of a stagnant air boundary layer (e.g. as in the Behavior Assessment Model (BAM) (Jury *et al.*, 1983, Jury *et al.*, 1984), the PESTicide Leaching and Accumulation model (PESTLA) (Van den Berg and Boesten (1999), Van den Berg *et al.*, (1999)) is useful for screening purposes. However, it should be noted that rough estimates may not be good enough for a further step in the risk assessment procedure (higher tier), so computations would be needed with a model which describes all relevant processes adequately and which has been tested and validated against field measurements.

No models are currently available for estimating the volatilization flux of pesticides from plants. More research is needed on describing the processes and factors that affect the fate of the pesticide on the plant leaf, such as the development stage of the plant, the characteristics of the plant leaf surface, photo-degradation, uptake by the plant and wash-off of the pesticide by rainfall and irrigation. There is no model yet available that handles photo-degradation on plant surfaces. A plant growth model may be a good starting point for modeling processes occurring on the plant leaf surface.

After spraying a pesticide on the crop, a fraction of the dosage is deposited on the soil surface. Furthermore, a part of the mass deposited on the plant surface may be washed off and be deposited on the soil surface. When measuring the volatilization rate, losses of pesticide from both the soil and the plant surfaces contribute to this flux. In the field it is not possible to distinguish between the two volatilization sources, which would make model testing more difficult. However, the volatilization of pesticide from solely the plant surface can be measured in a volatilization chamber.

7. Regional Emissions

Information is needed on the location and timing of the applications of a pesticide in an agricultural area to assess the total risk of transport of the pesticide via the air to great distances from this area. This involves information on the type of application of the pesticide (crop, soil), the mode of application (equipment and technique), the dosage and the temporal and spatial distribution of the application of the pesticide. The degree of detail in the data requirements for this type of assessment depends on the scale for which the assessment is made. An emission estimate has been made for 9 pesticides used in North America (Scholtz and Voldner, 1992; Scholtz *et al.*, 1997). Sales and use information were collected or disaggregated to a county level by crop type (county diameter ranging from 30 to > 140 km) and this information was aggregated on the surface area of the relevant crop for the area studied, using 127 km grid cells on a polar stereographic projection. For each grid cell, annual and seasonal emission factors were calculated for each pesticide -mode of application combination using submodels for volatilization from soil and crops. In general, the distribution of pesticide emissions corresponded to the usage pattern. As expected, the largest emissions occurred in spring and summer. Because simplifications were made in the procedure for the calculation of the emission factors, the results should be treated with caution. Further model development and testing is needed, in particular on the volatilization from crops.

The availability of detailed information on pesticide use differs from country to country. In the State of California, US, pesticide use permit data are compiled in each township. Using this information, the mass of pesticide used can be calculated on a weekly or monthly basis. For the entire US, county level pesticide use data were compiled by the US Geological Survey based on the 1992 Agricultural Census. The data are available in both map and digitized geographical information system–compatible data bases. In the Netherlands, the use of pesticides is estimated from relevant factors, such as the crop type, the total area of the crop, the dosage, the time of application and the frequency of use (ISBEST: Information System for Pesticides, Lentjes and Denneboom, 1996). This information is linked with a geographical information system in which data are available on the total crop area for each county. For several groups of pesticides, the total estimated use has been compared with information on pesticide sales. For soil fumigants the correlation was good (105%), but for the fungicides (72%), insecticides (51%) and herbicides (62%), there was a substantial difference between estimated use and pesticide sales (Smidt *et al.*, 1999). At present, such data on regional pesticide use are not available in Germany.

For emission of pesticides from bare soils, the organic matter content of the topsoil should be taken into account. The geographical information system could provide data on the occurrence of a soil type in the area studied, together with the organic matter content in the topsoil. In the US, a national data base, the State Soil Geographic Database (STATSGO), containing organic matter estimates at a map scale of 1:250,000, with some assumptions of spatial homogeneity and intended for regional application has been developed (United States Department of Agriculture, 1994). A more detailed data set named the Soil Survey Geographic (SSURGO), which is as detailed as map scale 1:12000, is under development. For a screening assessment, the emission of pesticide from bare soil can be estimated with a model such as the Behavior Assessment Model (Jury *et al.*, 1983). Using this model, output on the emission of pesticide into the air can be provided on a daily basis.

Using information from a geographical database and linking these data to information on the volatilization flux of pesticide from each soil type under a given set of environmental conditions, estimates on the total emission into the atmosphere from applications in the region can be obtained for each pesticide-soil combination. A similar approach can be followed to obtain estimates of the total emission for each pesticide-crop combination.

8. Conclusions and Recommendations

Little information is available on the fraction of the dosage of the pesticide that misses the target surface and is lost, mostly via spray drift, during application. Because 30% or more (greater than 50% in some cases) of the dosage can be lost during application, depending upon application technique, formulation and environmental conditions, more data are needed for a range of application types and weather conditions. In addition, these data are also necessary to verify the computed results of drift models and in further model development and verification. Standardization of the definition and description of alternative application techniques would promote a better use of both models and data. Accurate data on the loss during application are necessary for an adequate risk evaluation and they are also needed to identify measures which could reduce pesticide losses during application.

The volatilization of soil fumigants and soil incorporated pesticides following application can be reasonably well estimated with models. With the help of a model for fumigant behavior in soil, the effect of possible measures to reduce the emission into the air can be quantified. However, as the differences observed between the computed and measured rates were sometimes substantial, further development and testing is needed to improve the reliability of both models and measurement methods.

The rate and extent of volatilization of many soil surface-applied pesticides is uncertain. Further model development is needed as well as accompanying laboratory and field studies investigating those factors that can affect the volatilization process, in particular the soil water content and the temperature gradients within the top few mm of the soil. This should result in better descriptions relating the pesticide volatilization rate with the physico-chemical properties (vapor pressure and water solubility) and the sorption to soil particles under variable temperature and soil water conditions.

The stagnant air-boundary layer concept is commonly used for screening pesticides on their volatilization potential. For a more advanced risk assessment, a volatilization model is needed which describes the processes at the soil surface adequately, in particular for soil surface-applied pesticides.

More data are needed on the volatilization of pesticides from plant surfaces. More data are also needed about the effect of the various processes on the fate of the pesticide on the plants, such as photo-degradation, uptake by plant leaves, sorption and wash-off. These data are required for the development of a model describing the fate of pesticides on the crop.

Depending on pesticide properties, application technique, soil, crop and environmental conditions, volatilization losses of pesticides after application range from less than a few percent to 50-60% of the dosage or even more in some cases. It should be noted that incorporation of the pesticide into the soil directly after spraying greatly reduces the volatilization of the pesticide compared to soil surface applications.

Chamber-lysimeter experiments are useful tools to determine the volatilization potential of pesticides from plants and soils. The data obtained in such experiments can also be used for further model development. The chambers are also useful tools to asses the fate of the transformation products. The duration of the experiments in such systems can be prolonged to at least several weeks, so a better insight into the processes that affect pesticide volatilization can be obtained with varying time periods and controlled environmental conditions. Many important weather conditions in the field (e.g. wind speed, air temperature, solar radiation) can be reasonably well simulated in volatilization chambers, but the simulation of some processes in the chamber, such as rainfall and turbulence needs to be improved. The degree and type of air turbulence in the field cannot be representatively simulated in the volatilization chamber.

Because in most countries only about 1% of the agricultural area or less is under glass, the emission from glasshouses may be of limited importance. However, for the risk evaluation of exposure of man and ecosystems to pesticides in air in the neighborhood of a glasshouse area, the emission of pesticide from the glasshouse into the atmosphere should be considered.

For regional emissions detailed and accurate input data are needed which involves information on the spatial and temporal pesticide use, soil and/or crop type, the dosage applied and the mode of application. This information should be linked with a geographical information system that provides information on the area of a specific soil or crop with its characteristics within a predefined area.

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