Emissions by aerial routes from protected crop systems (greenhouses and crops grown under cover)

A position paper

Cecilia Stanghellini

Report 224

WAGENINGEN UR
For quality of life

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Wageningen UR Greenhouse Horticulture, Wageningen
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Summary for internet

This report describes the processes that may lead to emission of Plant Protection Products (PPP) from protected cultivation, through aerial routes. The introduction gives the background for this work and the limitations, outlining in particular why receptors other than air are not explicitly addressed here. Chapters 2 discusses the physical background of greenhouse air exchanges and the factors that affect it. Existing models for estimating ventilation of the different types of greenhouses are reviewed there. Chapter 3 gives a scientific argument about the processes and the factors that may affect aerial emissions of PPP from protected cultivations. The parameters that may have an high impact on the emission are identified there as well. A review of the knowledge needed and of the models that may be available for scoring each emission route is given in Chapter 4. In Chapter 5 a strategy is proposed to reduce/group the number of factors that are important (and to score their relevance) through some model calculations. An outline of the calculations that would be needed for ranking and eventually scoring the emissions and, possibly, highlight groupings of combinations that are similar with respect to emissions, is given.

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Summary

This report describes the processes that may lead to emission of Plant Protection Products (PPP) from protected cultivation, through aerial routes. The reasoning is built-up as follows:

- Aerial emissions of PPP from protected cultivations are related (through the pesticide properties) to the ventilation of the shelter (hereafter called greenhouse).
- The ventilation **requirement** of a greenhouses is dictated by the need to maintain an acceptable climate for the crop under given wheather conditions. The ventilation requirement determines (together with economics) greenhouse design in different climates.
- The **actual** ventilation at a given time (or ventilation rate) is heavily affected by wind speed, and thus highly variable in time, and may deviate from the ventilation requirement whenever the openings are not fully regulable.
- The emission along short-term emission routes (spray and gas during/after application, and during compulsory ventilation sometime after application) are determined by pesticide properties, droplet size and actual ventilation and must be scored through scenario calculations.
- In the long-term, actual ventilation will approach the ventilation requirement–though fully so only with computer-controlled openings. The long term emission of re-evaporated pesticides must be scored by linking re-evaporation to scenarios of the ventilation requirement (controlled openings), possibly accounting for limitations of the ventilation control (uncontrolled openings).

The introduction gives the background for this work and the limitations, outlining in particular why receptors other than air are not explicitly addressed here. Chapter 2 discusses the physical background of greenhouse air exchanges and the factors that affect it. Existing models for estimating ventilation of the different types of greenhouses are reviewed there. Chapter 3 gives a scientific argument about the processes and the factors that may affect aerial emissions of PPP from protected cultivations. The parameters that may have an high impact on the emission are identified there as well. A review of the knowledge needed and of the models that may be available for scoring each emission route is given in Chapter 4. In Chapter 5 a strategy is proposed to reduce/group the number of factors that are important (and to score their relevance) through some model calculations. An outline of the calculations that would be needed for ranking and eventually scoring the emissions and, possibly, highlight groupings of combinations that are similar with respect to emissions, is given.

Contents

1. Introduction

Working group emissions from protected crops

The Scientific Panel on Plant Protection Products and their Residues (PPR Panel) of EFSA has been asked to develop an inventory of protected crop systems (e.g. greenhouses and cultivations grown under cover) and emissions from these systems to relevant environmental compartments, and to provide guidance on the importance of emission routes including the circumstances under which they are relevant. To assist in the establishment of these products, the Working Group on Emissions from Protected Crops (WGEPC) has been established. The WGEPC consists of several members of the PPR Panel and five 'ad hoc experts' from Britain, Italy, the Netherlands and Poland.

Classifications

The first few meetings of the WGEPC were devoted to establish a framework apt to arrange conveniently possible emissions from application of plant protection products in the broad field of cultivation of crops under cover. The classifications on which agreement was reached on: 1) the construction of the cover, 2) the cultivation system and 3) the application type, are listed here below.

Cover construction types

The construction types differ in their influence on climatic and environmental conditions under which crops are grown. The construction type may therefore have influence on the emission of plant protection products from the system. The WGEPC considers six types of construction in or under which protected crops are grown :

GrH greenhouse
GlH glasshouse
LT low tunnel
PS plastic shelter
SH shade/net house
WT walk-in tunnel

Cultivation systems

Four cultivation systems are considered, depending on the crop being soil bound or not and whether the water, necessary for the growth of the crop, is recycled or not:

S+R soil with recycling
SL+R soilless with recycling
SnoR soil without recycling
SLnoR soilless without recycling

Application types

Furthermore, the way in which the plant protection product is applied may significantly influence the amount emitted and the relative importance of emission routes which may be considered. Application types are:

DI drip irrigation with / without recycling
FogFum fogging or fumigation
Inj injection into the soil
Spray spraying of the crop
SoilAp soil application

Potential emission routes

The following emissions routes were identified:

1. Spray drift emission at moment of application
2. Gas phase emission at moment of application
3. Gas-phase emission during compulsory ventilation prior to re-entry
4. Long-term gas phase emission through uncontrolled openings
5. Long-term gas phase emission through regulated openings
6. Discharge from circulation drainage tank
7. Discharge of rinsing water (cleansing of growing system)
8. Controlled discharge of condensation water (via drain / waste water basin)
9. Not controlled emission of condensation water
10. Leaching of applied substance

Run-off was recognized to be a possible emission route, but it was decided not to deal in detail with that.
This report covers emission routes to air (first five in the above list), whereas the emission routes are treated in a separate report (Van der Linden, 2009).

Scope of the present assignment

The working group felt the need for detailed background information about the factors that may affect the relevance of particular emission routes. In particular, Wageningen UR Greenhouse Horticulture (the group Greenhouse Technology), was contracted to write a "position paper" (the present document) on the emission by air from protected crops.

The Terms of Reference of the assignment are the following:

- Improving the scientific argument and review criteria for scoring and ranking of emissions through different (aerial) routes (drift, gas at moment of application, uncontrolled gas phase emission, controlled gas phase emission, forced ventilation) to different receptors (air, surface water and sediment, groundwater, soil, Sewage Treatment Plants). The relevance to non-target organisms (terrestrial environment) and birds and mammals as potential receptors is also to be considered.
- Critical review of available models and calculation methods and level of validation that could be used for ranking and eventually scoring of emissions.
- Provide an outline of scenario calculations that would be needed for ranking and eventually scoring the emission (supported by practical examples).
- Highlight need for data and/or models that may not be available yet. Identification of parameters with high impact on the emission ranking. In addition proposals how to reduce uncertainty when these calculations are applied at different cover systems, crop categories, growing systems and different geographical areas.

Present approach

1. In all aerial emission, the Plant Protection Product (PPP) will have to be carried away by the air stream leaving the enclosure, since the diffusion properties of most gases are usually several orders of magnitude lower than the diffusion caused by convective mass transfer.
2. The factors that affect air exchange of crop grown under cover (the driving forces)
3. Models for estimating air exchange under given conditions
4. From air exchange to emission of a PPP
5. What is missing

Reading guidance

In the early stages of its work, the workgroup decided to consider five receptors: air, groundwater, soil, sewage treatment plant and surface water. This report deals with air as the lone receptor of PPP's that leave the protected cultivation through aerial routes. Deposition on soil or surface water after a PPP has left the boundaries of the enclosure may be dealt with through the same models that are applied to field application of PPP, by using as boundary condition the configuration and concentration of the airflow exiting the enclosures.

2. Ventilation in protected cultivation

Air exchange of an enclosure

No protected cultivation system is perfectly sealed. There is always some air exchange that may be the carrier for PPP to reach receptors external to the protected cultivation. There most common ways to quantify the air exchange are:

- Total ventilation rate is the total amount of air exchanged in the unit of time and is indicated in m^3 per hour or m^3 per second. As this does not account for the size of the greenhouse, a better unit are the following.
- Specific ventilation rate is the amount of ventilation per unit soil surface area, $m^3\ m^{-2}\ s^{-1} = m\ s^{-1}$, and gives more information about the intensity of the process.
- Volume exchange rate per hour, h^{-1}, is a much used definition. It tells how many times per hour is the air within the enclosure refreshed. It is obviously related to the specific ventilation rate through the average height of the greenhouse H, m, since for each volume exchange, H m^3 are exchanged for each m^2 of floor area.

Even fully closed, a protected cultivation has some "porosity" (leakage ventilation). With closed ventilation openings, a leakage rate of 0.15 volumes per hour is estimated even for the well-sealed, modern Venlo glasshouses of Holland. A value of 0.5 h^{-1} is more reasonable for older glasshouses. At the other extreme, typical good-weather ventilation rates in the Mediterranean basin can be up to 50 air exchanges per hour.

Ventilation requirement

Greenhouses needs to exchange air with the ambient, in order to get rid of excess energy and of water vapour and to let carbon dioxide in, whenever this is not artificially supplied. In this latter case, however, carbon dioxide concentration is usually not measured, and thus not a factor determining the ventilation rate. As the example given in Fig. 1 shows, the amount of ventilation that is required depends primarily on two factors: a. the amount of sun energy entering the greenhouse and b. the amount of excess vapour (after condensation and ventilation for energy, "thermal" ventilation).

The amount of sun energy entering the greenhouse depends very obviously from the place, and from the properties of the cover. For instance, year sun radiation in Almeria is 6430 MJ m^{-2}, almost exactly double the figure for Holland. However, transmissivity of the cover is much less than the 75% typical of Dutch glasshouses and it is further reduced by whitewashing in the warmest months. A simple example as in Fig. 2 will clarify this.

THE VENTILATION REQUIREMENT IS DETERMINED BY THE CLIMATE AND THE PROPERTIES OF THE COVER

Even when the temperature in the greenhouse is as desired, ventilation may be necessary to prevent excessive humidity, which is linked to incidence of pathologies. The source of humidity is crop transpiration (largely determined by sun radiation). Condensation (which depends on outside conditions and cover proerties) may remove water vapour from the environment. Nighttime greenhouse ventilation is usually dictated by the need to get rid of humidity.

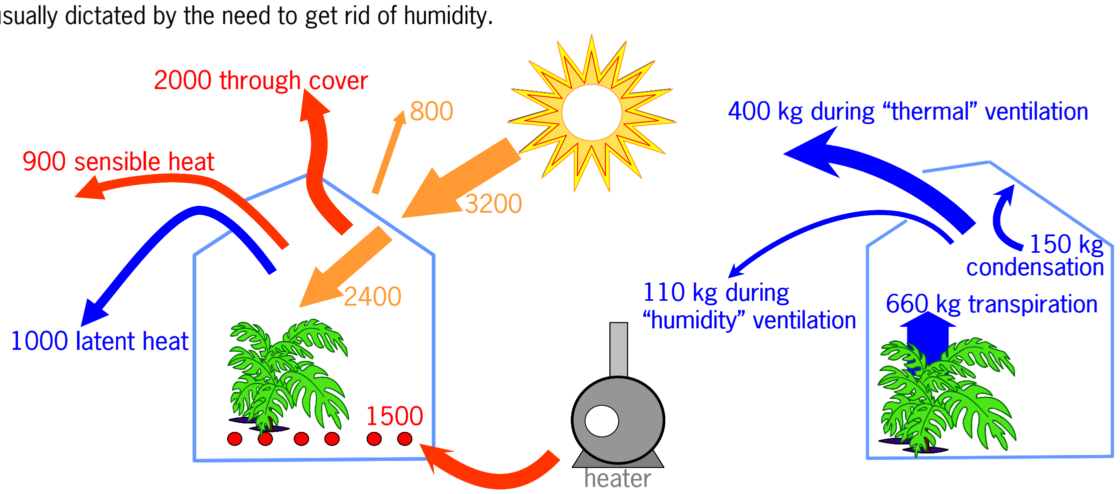

Figure 1. *Yearly energy (MJ m^{-2}, left) and vapour balance (kg m^{-2}, right) of a typical greenhouse in Holland. Ventilation is required to get rid of sun energy (when the greenhouse would become too warm) and also to get rid of water vapour when ventilation would otherwise not be required. Heating is needed to ensure at all times the required temperature whenever sun radiation does not suffice.*

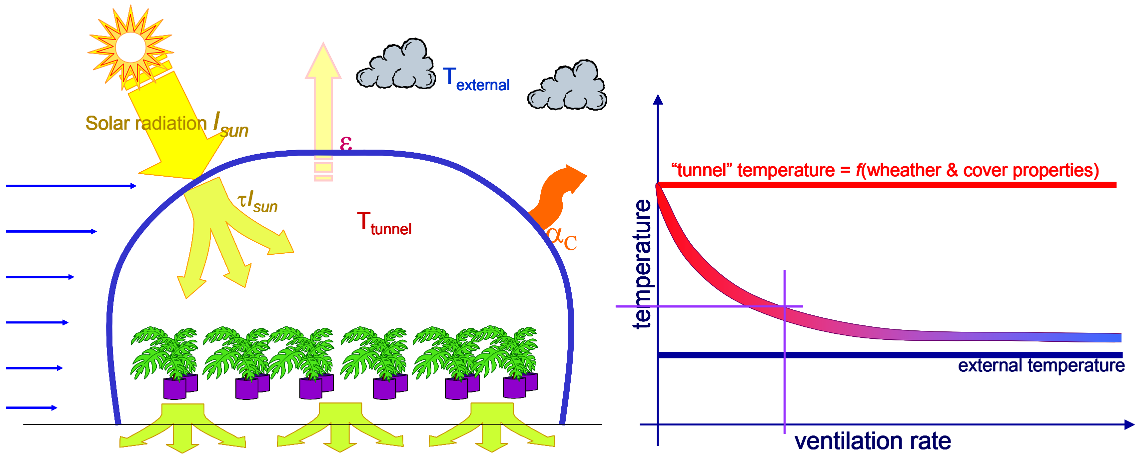

Figure 2. *Left: the temperature in an enclosure (tunnel) results from the balance of the energy fluxes in and out of the tunnel. These, in turn, depend on the properties (transmissivity, emissivity and heat transfer) of the cover. Right: the temperature in a ventilated enclosure approaches the external temperature with increasing ventilation rate. Regulated openings adapt the ventilation rate to changing weather, in order to maintain the internal temperature as desired.*

For example, let's assume we have a double-layer polyethylene covered tunnel ($\tau \approx 70\%$ and heat transfer coefficient $\alpha_c \approx 6$ W m^{-2} $^oC^{-1}$), with a sun radiation of 600 W m^{-2}. Assuming that the transpiration of the crop, ET_{crop}, absorbs 2/3 of the available energy, the temperature of the tunnel without ventilation can be estimated:

$$\tau I_{sun} - ET_{crop} = \alpha_c (T_{in} - T_{out}) \rightarrow T_{in} - T_{out} \approx \frac{0.33 \cdot 0.7 \cdot 600}{6} \approx 23\ ^oC \qquad \text{W m}^{-2} \ (1)$$

That is, if the outside temperature is, say, 25 ^oC, there would be inside around 50. No crop grows in such a temperature and, indeed, no greenhouse is closed in such conditions. Here we are even neglecting the fact that after a while the air will be fully saturated with vapour, and the inevitable condensation will release some of the energy that was removed by transpiration. The amount of ventilation that would be required to manage the inside temperature below an acceptable level depends on the amount of energy that each m^3 is able to remove from the greenhouse. This depends on the difference in enthalpy (sensible and latent heat) between the air inside and outside of the enclosure. As the latent heat (humidity) inside depends on the crop transpiration which, in turn, depends on the vapour removal, a very rough estimate is that the required ventilation rate will be between 5 and 20 volume exchanges per hour.

THE VENTILATION IS "KNOWN" IF CROP GROWTH INSIDE HAS TO BE POSSIBLE

In this paper we disregard a very recent Dutch development the so-called "closed greenhouse", which is fully conditioned in order to prevent ventilation, reducing thereby pest pressure and allowing for a high CO_2 concentration. Only a very minor area is equipped like that–mainly for demonstration–and after a couple of years of trials, the perception is developing that the economics do not support fully closed greenhouses, not even in the high-tech and relatively low-radiation environment typical of Holland. It is true that the present tendence is towards less ventilation, mainly through some measure of evaporative cooling. Nevertheless, ventilation is here to stay as a inherent feature of greenhouse production.

Actual ventilation

Ideally, therefore, the ventilation rate of a protected cultivation is regulated such as to ensure the best possible ambient temperature and humidity for the crop inside. Whenever the ventilation rate is non-regulable, one has to assume that cultivation will take place only so long as the conditions inside allow for crop growth, with some form of seasonal accommodation insofar as possible, either of the ventilation openings, as shown in Fig. 3, or of the transmissivity of the cover (whitewash). As productivity in this case is greatly limited by the lack of tools for climate management, whenever the value of the product makes it reasonable, the type of protection and its openings are better adapted to the external conditions. Indeed, the ventilation requirement, together with financial constraints, is the main factor accounting for the huge variability in greenhouse design in different places, such as shown in the

Figure 3. *Seasonal accommodation of ventilation in tunnels in Sicily, IT. From left to right, winter, early spring, late spring. The next step is complete removal of the cover.*

examples given in Fig. 4. All the openings shown there are regulable, either by hand or by a climate control computer. In first approximation, therefore, one would say that the ventilation rate can be known whenever the relationship between crop climate requirements and natural climate is known. Observe that this means that even in greenhouses with controllable openings, the feasibility of compulsory closure after PPP application is limited: conditions inside may soon damage productivity.

If one wants to know the amount of air exchange at any time, that is actual ventilation rate, wind speed plays obviously a major role, besides the amount, size and positions of openings. Models have been developed for determining the air exchange rate in function of wind speeds and characteristics of the opening, for the most common greenhouse types. De Jong (1990) developed and calibrated a model for multi-span Venlo greenhouses, Kittas and his associates modeled air exchange rates of single shelters, from a tunnel with continuous side openings (Kittas et al., 1995) to a greenhouse with side and ridge openings (Kittas et al., 1997). Muñoz (1998) modeled air exchange rate of multi-tunnels and Perez-Parra et al. (2004) of parral-type greenhouses. In an earlier paper Perez-

Figure 4. *Examples of regulated ventilation openings in greenhouses. Clockwise from top left: alternate ridge openings in a Venlo glasshouse, Holland; parral greenhouse with one-side ridge flap, Almeria, ES; multitunnel with "seagull wing" openings, protected by insect nets, Sicily, Italy and fully retractable roof, Barcelona, ES.*

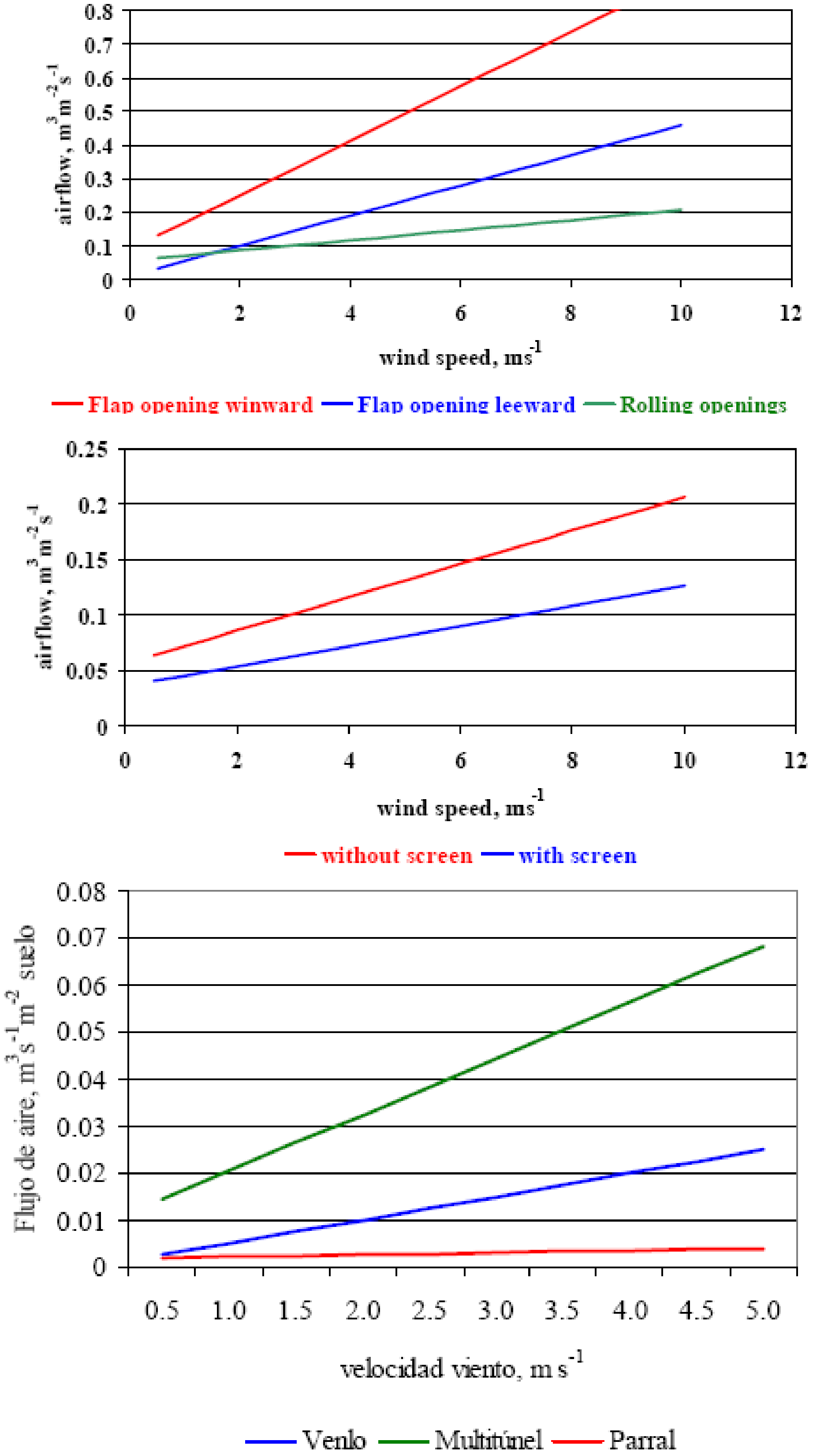

Figure 5. *Examples of the effect of various factors on the ventilation rate. Top: ventilation rate per unit area of the opening v wind speed, for zenital flaps in a parral greenhouse (see Fig. 4 top right) facing either windward or leeward and roll-type implementation of the same opening; Center: effect of insect nets on the airflow through a roll opening (the green line in the top panel); Bottom: specific ventilation rate (per m^2 soil surface area) of different types of greenhouses v wind speed. The parral greenhouse is with roll opening. After Munoz, 1998; Perez-Parra, 2002 and Perez-Parra et al., 2004, respectively.*

Parra et al. (2002) had quantified the reducing effect on ventilation of insect nets. Tanny et al. (2003) have measured the effect of a screenhouse on the ventilation rate, under wind speeds up to 3.5 m s^{-1}, and showed that it could still reduce air exchange by at least 50% with respect to open field. Altogether, it seems that the existing models allow for reasonable estimates of the ventilation rate, at a given wind speed, of all the major greenhouse types referred to in this work. Examples are given in Fig. 5, please note that, for instance, a flow rate of 0.04 m^3 m^{-2} s^{-1} (bottom panel) in a greenhouse 4 m high, equals 36 volume exchanges per hour.

THE EFFECT OF WIND IS IMPORTANT ONLY FOR “PASSIVE” OPENINGS

For the purpose of determining the chance of aerial emissions through ventilators, whenever the openings are computer-controlled, the opening fraction will automatically react to wind speed (large openings with little wind and the other way round) so that the resulting amount of air exchange is the right one to deliver the desired climate inside. Therefore, in this respect, greenhouses with natural ventilation through controlled openings may be treated similarly as greenhouses with forced ventilation, that is controlled air flow through fans installed in the greenhouse walls. Obviously, the effect of wind is very relevant–and should be accounted for–for all protected cultivations with non (or poorly) regulable openings.

3. From air exchange to emission

Transfer of mass in an air stream

Gas

When considering the diffusion from a source of gas in a moving air stream, it is necessary first to consider whether the diffusion is primarily driven by natural diffusion (that is, the molecular diffusivity of the gas) or by convection (that is, the gas is passively carried by the air stream). Obviously both processes are coexisting always, but it may be helpful to consider which one is predominant and how much.

There is a fundamental similarity between the molecular diffusion of heat, mass and momentum in the boundary layer around solids, and non-dimensional parameters are usually applied to account for the differences in the effective thickness of the boundary layer for heat and mass transfer. The most appropriate here is the Lewis number (Le), that is, the ratio of the thermal diffusivity of dry air ($\kappa = 1.51\ 10^{-5}\ m^2\ s^{-1}$) to the molecular diffusivity of the gas in air. When the Lewis number is not far from one, convection is the predominant process, and the gas flow (F, $kg\ m^{-2}\ s^{-1}$) out of an enclosure can be estimated from the specific ventilation rate (V, $m^3\ m^{-2}\ s^{-1}$) and the difference in concentration (C, $kg\ m^{-3}$) between in- and outside, as:

$$F = Le^{-0.667}\ V\ (C_{in} - C_{out}) \qquad kg\ m^{-2}\ s^{-1}\ (2)$$

For instance, for water vapour and CO_2 (two important gases in greenhouse management) the correction factor $Le^{-0.667}$ is respectively: 1.075 and 0.76, and is usually disregarded in calculations, since it is small compared with the inaccuracy connected to our knowledge of the ventilation rate. Obviously, contrary to vapour and CO_2, the natural, outside concentration of PPP is nihil, and eq(1) simplifies accordingly. However, when attempting to estimate the total emission over one period, one has to account for varying conditions of both the concentration inside and of the ventilation rate. The total emission (E, $kg\ m^{-2}$) of a gas from the moment of application to time t is then calculated by:

THE GASEOUS AERIAL EMISSION IS "KNOWN" IF THE VENTILATION RATE IS KNOWN

$$E = \mathrm{Le}^{-0.667} \int_0^t C_{in}(t) \cdot V(t) \cdot dt \qquad kg\ m^{-2}\ (3)$$

In Eqs(2) and (3) it is implicitly assumed that the concentration inside the greenhouse is homogeneous at any time. This is obviously not true, since usually there will be a vertical gradient, caused both by the weight of the material and by the fact that it will be applied in prevalence towards the crop. The position of the openings becomes relevant in the presence of a gradient. For instance, opening exclusively on the roof may cause Eq(3) to overestimate total

Figure 6. *Side-wall and zenital openings, all netted, in a parral greenhouse in Almeria, ES. The permeability of the nets may be further reduced by accumulation of dust.*

emission when a spatial mean of C_{in} is used (e.g. Duyzer at al., 2004), whereas a prevalence of side openings (Fig. 6) may have the opposite effect. Complexer calculations through Computational Fluid Dynamics (CFD) may be required for an accurate estimate in this case. Indeed, Bartzanas et al (2006) have demonstrated that a CFD-based caluculation of gaseous dispersion from a greenhouse, based on the similarity described here, could well reproduce measured dispersion rates of a tracer gas. In summary, the emission of a PPP from a greenhouse at any time, can be estimated from the ventilation rate, provided the molecular diffusivity of the gas is known.

Droplets

The above similarity in principle holds also for spray droplets. However, there are additional factors that have to be considered: one is the effect of gravity, and the other the reducing size of the droplets due to evaporation, e.g. Lebeau (2004). Nuyttens et al. (2004) have shown that one has to account for these factors when designing a spary boom for application in protected cultivations. A computer model for spray drift has been developed by Plant Research International, Wageningen, (IDEFICS, Holterman et al., 1995) which has been validated also recently with field application of spray on potatoes (van den Berg et al., 2006), and has been shown also to estimate correctly deposition of granulated substances (Holterman et al., 2006). Van Os et al. (1994) compared measurements of aerial emissions of a volatile (dichlorvos) and moderately volatile (parathion) substance and values estimated by IDEFICS, depending on a. the amount of pesticide brought into the greenhouse; b. a prefixed air exchange of the greenhouse; c. the evaporation of the pesticide after deposition and d. the total inactivation time of the pesticide. They showed that emission was highly dependent on the assumed ventilation rate and that, more unexpectedly, there was little effect of long term behaviour of ventilation on total emission. This is because they considered a leakage rate of 0.5 volume exchanges per hour even with closed windows. Very modern Dutch greenhouses have probably a smaller leakage rate, whereas such a value is probably underestimating true leakage of most commercial greenhouses. A proof-of-principle of estimating spray emission from protected crops by coupling IDEFICS to a ventilation model was given by Holterman at al., 1998.

SPRAY DRIFT IS A COMPLEX PROCESS BEST DESCRIBED BY MODELS

Application methods and their effect

Although an analysis of the effect of the type of application on the relevance of an emission route is outside of the

Figure 7. *Crop treatment through a spray-boom in a Dutch Venlo glasshouse. During application the ventilators are regulated the usual way by the climate control system.*

scope of this paper, a short overview of the two most common types of aerial application makes the task easier.

Aerial Treatment (FogFum)

The PPP suspended in water is injected in the exhaust stream of the combustion of a mixture of fuel and air, in a so-called fogger, usually hand-held, which is pointed obliquely high in the greenhouse. Aerial treatments are: Low Volume Misting (small droplets up to 25μm), fogging and fumigation. Droplets in the last two methods are so small that the effect of gravity on their displacement can be neglected. The application is rather fast, often less than one hour, and the ventilators are closed as much as possible. At the end of the application the greenhouse is full with a kind of fog that dissolves in a few hours, after which the ventilators are open again. Standard advice is to to keep the greenhouse sealed for at least four hours after application, which also increases effectivity. In order to prevent too high temperatures caused by the absence of ventilation, this application takes place usually in the evening. In most cases it is compulsory to ventilate well the greenhouse prior to re-entry of the personnel.

Crop Treatment (Spray)

This type of application can be performed through a hand-held spray device, or a spray-boom (Fig. 7), with nozzles aimed downwards, that moves slowly above the crop, or a spray-mast (vertical) with lateral nozzles, moving through a high crop (such as tomato). Droplet size vary between 10 and 400 μm, with a minimal fraction, however, smaller than 25 μm. Application in a large greenhouse takes usually some hours and the ventilators are not closed (see Fig.7) but regulated in the usual way. The treatment occurs usually at the end of the working day, and it may happen, partially at least, during daytime.

What is important?

In order to see which factors are relevant, we calculate total emission for FogFum, through eq(3) in a number of conditions. We will assume that the initial concentration of the chemical is uniform and that the position of the openings does not affect emission (which might be not far from the truth in this case), and that the Lewis number of the chemical (its diffusivity) is not far from one. An important characteristic of the chemical is its break-down rate, that we will describe here through its half life–the time it would take for its concentration to halve, even in the absence of any air exchange. Half life must be regarded here a compound indicator for all processes (besides ventilation) that would remove a chemical from the greenhouse environment. With this respect it is important to realize that in the reduced presence of UV radiation in glass/greenhouses, break-down time of of large molecules may be longer than in natural conditions. Other factors that are obviously important are a. how "porous" is the greenhouse even with closed ventilators and b. the strategy, that is, how long after application are the ventilators open/operated.

As Fig. 8 shows, only for chemicals with a relatively long half-life the porosity and ventilation strategy play a minor role–as nearly all chemical present in air after application will be emitted anyhow. Obviously the shorter the half life, the more relevant it becomes what happen in the greenhouse in the first few hours. For instance, in a porous greenhouse most emissions will have taken place before opening of the windows (Fig. 8, left), whereas in an airtight one, delaying the opening may reduce emissions significantly. It would be wrong, however, to extend the implications drawn from Fig. 8 to conditions where the underlying assumption of homogeneity would not apply, such as spray drift, or re-evaporation of deposited material (volatilization).

Fig. 8 shows the emission as a fraction of the substance present in the greenhouse air at time = 0. The application type (and the losses on emission routes 1 and 2) determine its relationship with applied dose. In most cases, however, new substance will be introduced into the greenhouse air by volatilization, for a [long] time after the end of the application (the starting time of Fig. 8). In this aspect it is worthwile to recollect the experimental and modeling stuydy by Leistra and Crum, 1990, who determined emission from the soil to the greenhouse air of a fumigant (metham-sodium) injected into the soil which was covered by a low-density polyethene film immediately thereafter. They found that up to 50% of the applied dose came back into the air within one week after application, permeating the film, and some 3% through leakages in the film. Volatilization in the second week (after removal of the film at day 7) added another 7% of dose to emission into the greenhouse air. Having no cover at all increased only slightly the emission.

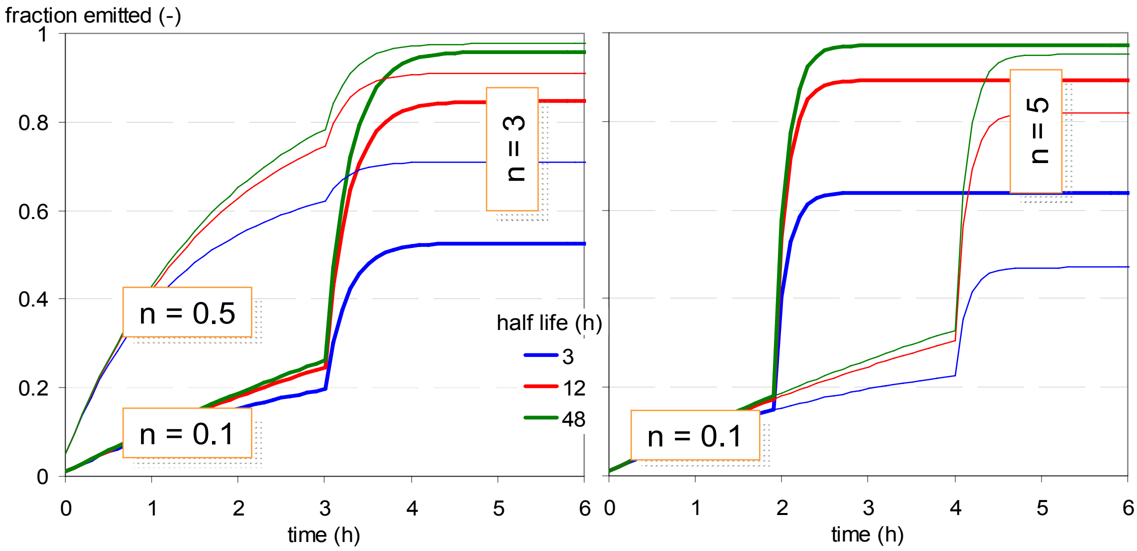

Figure 8. *Cumulative emission (as a fraction of the matter present in air after application, not of the applied dose) vs time, for chemicals having half life of 3 (blue), 12 (red) and 48 (green) hours, respectively, in a number of ventilation scenarios. Left: an air-tight or a leaky greenhouse (0.1 and 0.5 volume exchanges per hour, respectively), open after 3 hours with a ventilation rate of 3 per hour thereafter. Right: an air-tight greenhouse open either after 2 or 4 hours, with a ventilation rate of 5 per hour thereafter. Mean height of the greenhouse is assumed to be 5 m.*

In a more recent study by Schmidt et al. (2002), pesticides based on lindane, parathion, pirimicarb, procymidone, or tebufenpyrad were experimentally applied in two trial greenhouses and atmospheric concentration of these substances measured in the following 24 hours. The application rate was 750 g/ha active substance using a high volume spray technique. Assuming a mean greenhouse height of 3.75 m, the equivalent initial concentration was threfore about 20 $\mu g/m^3$. The indoor concentrations measured after 24 h in the middle of the greenhouse are given in Tab. 1, in decreasing order of volatility.

	Application		**24 h site 1**	**24 h site 2**	**mean**
	g/ha	*$\mu g/m^3$*	*$\mu g/m^3$*	*$\mu g/m^3$*	*% of initial*
Lindane			6.3	6.4	32
Parathion			1.2	1.2	6
Pirimicarb	750	20	0.64	0.75	3.5
Procymidone			0.28	0.46	2
Tebufenpyrad			0.026	0.19	0.5

Table 1. *Concentration of the indicated substances measured in the air of two non-ventilated greenhouses 24 h after high-volume application of 750 g/ha active substance (from Schmidt et al., 2002). The transformation in equivalent initial concentration has been made assuming a mean greenhouse height of 3.75 m.*

In conclusion, the more volatile the chemical, the more will be available in the greenhouse air to be carried away by ventilation. Volatility is known to be affected by temperature, besides the nature of the substance. Therefore, an estimate of the long term emission must be based firstly on an estimate of the long term source within the greenhouse, accounting for the factors that may affect it, and then on the ventilation rate. For volatile chemicals the worst case presumption seems to be that most of the applied dose will be carried eventually outside by ventilation.

4. Emission routes

Five possible aerial emission routes have been identified. Now we discuss how the ventilation rate and its variability–together with relevant properties of the material/application–may affect the chance of emission along all of them. As agreed in the WGEPC, we consider emission all flows leaving the external boundary of the greenhouse. In order to score impact on a particular receptor thereafter, the scoring of emission should be coupled to risk assessment models, as done f.i. with USES by Mensink (2004) for human exposure. Similarly, Duyzer et al., 2004 applied a modified "leeside vortex" model to determine air concentration of PPP's at selected distances from a greenhouse, based on the dosis applied, a form parameter of the greenhouse and the wind speed.

Spray drift

As heavy droplets will either evaporate (decading then into gas emission) or fall down, drift emission may take place only for a relatively short time after application (Brower et al., 1992). In the case of space application (FogFum) in a closed greenhouse, the emission depends obviously from the leakage rate, see Fig. 8. For instance, Duyzer et al., 2004 determined that in a well-sealed Venlo glasshose most emission would take place after re-opening (Fig. 9, right) whereas van Os et al., 1994, had determined the opposite in a more leaky glasshouse (Fig. 9, left). Observe that this is a contributing factor to the relevance of emission route 3, as Fig. 9 makes clear.
When space application takes place with significant leakages (or non controllable openings), the position of the openinings with respect to the applicator, and the presence of insect nets will also play an obvious role. This applies as well to crop (Spray) applications, since these seldom take place with closed vents. In view of the variability of air speed and of the internal air movement on the time and dimension scales that need to be considered here, the best way to score emissions would be to run a number of scenarios (coupling Computational Fluid Dynamics ventilation models and spray drift models) for some relevant type of greenhouses and opening configuration. The final score would obviously be dependent on greenhouse type and wind speed.

Gas at the moment of application

In term of time scale, this emission route is quite similar to the previous one. The way to address it would be the same: running scenarios. It would be simpler since only ventilation models would need to be run, accounting only for the diffusivity of the gas. A result of the scenarios could also be a broad classification of the ranges of diffusivities (property of the substance) that would result in significantly different scoring classes.

Gas-phase emission during compulsory ventilation prior to re-entry

This emission route refers to ventilation that takes place in order to remove the pesticide from the greenhouse air,

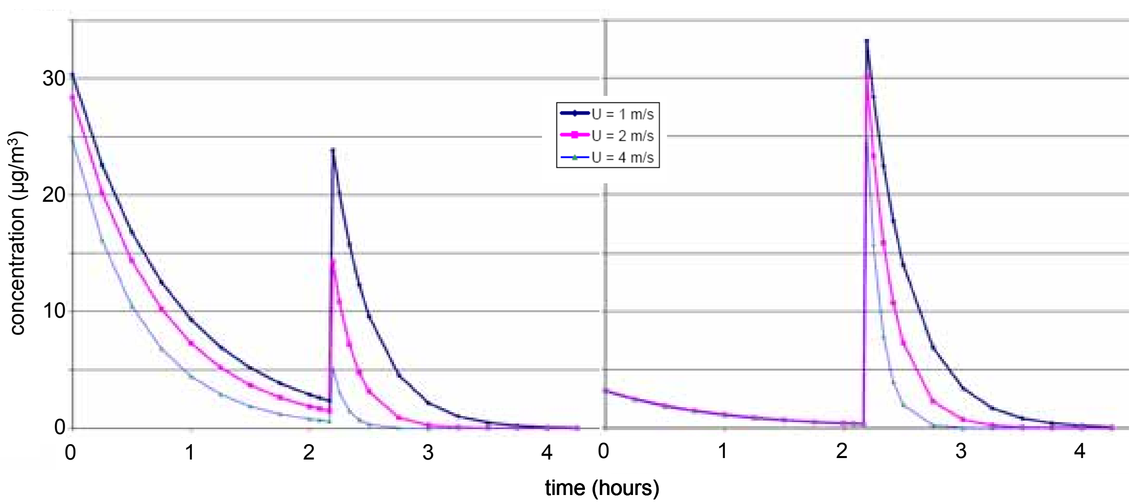

Figure 9. *Concentration 20 m leeway of a greenhouse after application of a dose of 1000 g per 3 ha at time = 0. Dutch Venlo glasshouse, left "leaky" and right well sealed, with three wind speeds. The emission caused by compulsory ventilation (E3) is represented by the peak after a bit more than two hours after application. Calculated by Duyzer et al., 2004, after calibration of the model with measurements.*

after application. Since for most PPP this is a compulsory practice prior to re-entry of the personnel into the greenhouse, it is necessary to score correctly the emission depending on the volatility of the pesticide and on the procedure, that is: time between application and opening of the ventilators. The leakage of the greenhouse in the period that it was close is a relevant factor, see Fig. 8.

Long-term gas-phase emission through uncontrolled openings

Residues arising from deposition following volatilisation are a new exposure term under Directive 91/414 and are distinct from spray drift. This is called long-term, since deposited material will evaporate in days (f.i. Martinez Vidal, 1997 and Egea et al., 1998) or even weeks (unpublished measurements, Wageningen UR Greenhouse Horticulture). The FOCUS (2008) group considered that the contribution from the deposition of volatilised residues to the receptors soli and surface water, was quantitatively less important than spray drift in the field. In a semi-closed environment, however, volatilization may lead to relatively high concentrations, which may result in significant emissions as soon as the ventilation rate increases. That means then that the long term ventilation rate is relevant for emission. As the openings are non or only partly regulated, the ventilation rate is determined primarily by the wind speed. Obviously important are the application type (how much PPP is deposited that may re-evaporate) and the volatility of the PPP (for how long it will go on evaporating). Scenarios are needed to score this emission route under various conditions (combinations of ventilation requirement, application type and volatility of the substance), since this is the one that is least likely to be “manageable” through regulation. Indeed, Garrat & Wilkins, 2004, implemented a 3-compartment (soil, plant and air) fugacity model to simulate pesticide behaviour in a greenhouse. The processes considered were soil-air exchange, leaf-air exchange, loss through greenhouse windows, metabolism in the plant, and loss from soil degradation or leaching. The model was applied to the results of Egea et al., 1998 on methamidophos residues in greenhouse air and crops and contrasted with the simulation of another sprayed pesticide, procymidone. The model reproduced the magnitude and diurnal fluctuations of airborne residues in the greenhouse.

Long-term gas-phase emission through regulated openings

This is actually the same as the previous, only made simpler by the fact that, being the opening automatic regulated, the long-term actual ventilation rate may be estimated through the ventilation requirement.

Summary

Table 2 attempts a scoring of the factors that are relevant for each emission route. They are grouped in: typical of the PPP (size of spray droplets, half-life and volatility); typical of the shelter (how porous is the cover anyhow, are the opening regulable, transmissivity for radiation) and of the weather (wind, temperature and radiation). Radiation is

Emission route	**substance**			**shelter**			**weather**		
	volatility	*drop size*	*half-life*	*porosity*	*regulability*	*transmis-sivity*	*radiation*	*wind*	*tempera-ture*
drift at application		↓↓		↑↑	↓	↓	↑	↑↑	↓
gas at application	↑	n.a.	n.a.	↑↑	↓		↑	↑↑	
compulsory ventilation	↑↑	n.a.	↑↑	↓	↓			↑↑	
long term uncontrolled	↑	n.a.	↑	↑	n.a.	↑	↓	↑	↑↑
long term controlled	↑	n.a.	↑	↑	↓	↑	↓	↑	↑↑

Table 2. *An overview of the factors (properties of the PPP and of the shelter, and weather factors) that may affect each possible emission route. An upward arrow means that emission increases by increasing the factor, whereas when increasing the factor (f.i. drop size) decreases the emission, a downward arrow is drawn. Two arrows indicate a strong relationship. Empy cells means that the factor has probably no influence on that emission route and n.a. means that the factor does not apply to that emission route. Radiation may reduce drif by fastening drop evaporation, and long-term emissions by increasing break-down rate of the PPP. Temperature increases both the ventilation requirement of the shelter and the volatility of the PPP.*

included since it may affect the half-life of the PPP and the evaporation rate of the droplets. Besides the obvious conclusions about the shelter (porosity increases emission and control of the openings decreases them), it may be worthwhile noticing that wind is very relevant for short term emissions, whereas climatic parameters such as temperature and radiation determine long term emission.
It may be useful at this point to attempt giving a structure to models/calculations that may be required for determining the factors that are likely to affect the emission along the routes listed above, as it is done in Table 3. With respect to the application type, for emission routes 1 and 2 it is relevant whether it is space application (uniform concentration of the PPP, supplied while protection is "closed"), or crop application, given with the purpose to create a gradient in concentration and usually while the protection is ventilated as necessary. This latter application may not necessarily lead to emission route 3. Another relevant characteristic of the application type is the size distribution of the droplets, which is relevant in the first place for the escape dynamics. Together with the weather (evaporation rate) the size of the droplets determine the duration of emission route 1.
The factors that have been mentioned as relevant for the construction are the "porosity" of the enclosure, which determines (together with the wind speed) the leakage ventilation; size, porosity and position of the openings; and whether they are automatically regulable or not. Non regulable openings may be included in the general "porosity" of the structure, except when they might be manually managed (before application of PPP or for seasonal adjustement.) The transmissivity of the cover material for sun radiation and heat determine (together with the weather) the ventilation requirement. Finally, the weather has been mentioned already: the most relevant factor is wind speed, since it determines the ventilation rate through all unregulated openings (including leakages). The sun radiation, temperature and humidity (in a minor role) determine both the evaporation of droplets and the ventilation requirement of the structure. Sun radiation (coupled to the transmissivity of the structure) may play a role in the break-down rate of deposited PPP.

	Application	**Construction**	**Weather**	**Models for scoring**
drift at application	• space treatment • drop size distribution	• leakage	• wind • radiation • temperature • humidity	drift models + computational fluid dynamics (CFD)
	• crop treatment • drop size distribution	• leakage • opening characteristics		
gas at application	• space treatment	• leakage	• wind	ventilation models + pesticide fate models
	• crop treatment	• leakage • opening characteristics		
compulsory ventilation	• space treatment	• leakage • opening characteristics	• wind	ventilation
long term uncontrolled	• all	• leakage • opening characteristics • cover material	• wind • radiation • temperature • humidity	ventilation + greenhouse climate+pesticide fate models
long term controlled	• all	• leakage • cover material	• wind • radiation • temperature • humidity	greenhouse climate +pesticide fate models

Table 3. *A summary of the factors–respectively of the application type, the construction and the weather/climate–that are relevant in determining the emission through the various routes. The last column gives suggestions about models that may be applied for scoring.*

5. Recommendations

The relevance of an emission route (once the application type is given) depends heavily on the combination of a number of factors of the structure and cover material and of the climate. Whereas there may be a sufficient knowledge on the effect of each of these single factors, a reliable scoring should be based on calculations of a number of combinations of these factors. It is therefore suggested that–within the life of this working group–an attempt be made to group/reduce the factors that will require a detailed risk assessment by a following working group. The calulations step that would be required to reduce the number of relevant parameters and to rank their relevance are:

1. Preselect three representative climates (wind, sun radiation and temperature), for instance Central, Southern and Continental EU.
2. Preselect as few as possible (4?) representative protected cultivation types, for instance: Venlo glasshouse, regulated multitunnel, unregulated parral, net house. Define their relevant characteristics (Tables 2 and 3).
3. Define relevant climate–cultivation type combinations. They will be less than 12, but more than 4, since some protection types can be found in more than one climate region.
4. For each combination:
    a. determine through a greenhouse climate model a typical [hourly] pattern of ventilation requirement along a crop cycle (a year or less).
    b. run scenarios for gaseous emission of PPP under (2?) preselected conditions of volatility and half-life of the PPP
    c. Run scenarios for drift emissions
5. Coupling the pattern of ventilation requirement (a) to the results of (b) and (c), makes it possible to score short term emission (1 to 3), depending on the weather at the moment of application, for each of the 20? combinations.
6. The long-term emissions (4 and 5) should be scored by coupling the pattern of ventilation requirement (E3) or of the actual ventilation, including the effect of wind, to a greenhouse climate model estimating the condensation and re-evaporation of the deposited substance (accounting for application type and volatility.)

6. Conclusion

In the § Introduction we have given the background for this work and the limitations, outlining in particular why receptors other than air are not explicitly addressed here. In § 2 we have discussed the physical background of greenhouse air exchanges and the factors that affect it. Existing models for estimating ventilation of the different types of greenhouses are reviewed there. § 3 gives a scientific argument about the processes and the factors that may affect aerial emissions of PPP from protected cultivations. The parameters that have an high impact on the emission ranking are identified there as well. A review of needed knowledge and of the models that may be available for scoring each emission route is given in § 4. Finally, § 5 gives an outline of the scenario calculations that would be needed for ranking and eventually scoring the emissions and, possibly, highlight groupings of combinations that are similar with respect to emissions.

The amount and variety of combinations, and of relevant parameters make impossible to attempt a ranking beforehand. For instance, the relative importance of the short- and long-term routes depends heavily on the volatility of the PPP. On the other hand, it is true (and it may be hoped) that a detailed sensitivity analysis of the scenario results obtained may allow for “clustering” and simplifications *a posteriori*. In addition, the estimated emissions may reveal a “natural” boundary between combinations (greenhouse type–climate) that are significantly different with respect to possible risks and/or regulations. In this way it would also be possible to highlight the defining factors with respect to which the claim may be substantiated (or not) that emissions from application of PPP under protected cultivation require different risk assessments than emissions from field application.

7. References

Bartzanas, T., N. Katsoulas, C. Kittas and A.A. Sapounas, 2006. Dispersion of Pesticides from a Naturally Ventilated Greenhouse: a CFD Approach, Acta Horticulturae, 718: 307-314

Berg, F. van den; Holterman, H.J.; Leistra, M., 2006. Emission of pesticides into the air during and after application to a potato crop In: Pesticide behaviour in soils, water and air; oral and poster abstracts. - London (UK) : SCI, 2006 - p. 1. Conference on Pesticide behaviour in soils, water and air; Warwick (UK), -03-27/ 2006-03-29

Brouwer, D.H., J.A.F. De Vreede, J.C. Ravensberg, R. Engel, J,J. Van Hemmen, 1992. Dissipation of aerosols from greenhouse air after application of pesticides using a low-volume technique. Implications for safe re-entry. Chemosphere, 24(9):1157-1169.

De Jong, T., 1990. Natural ventilation of large multi-span greenhouses. PhD dissertation, Wageningen University, 116 pp.

Duyzer, J., , van der Staay, M., Weststrate, H., Boertjes, B., Hollander, K. and Verhagen, H., 2004. De blootstelling van omwonenden van kassen aan gewasbeschermingsmiddelen via de lucht. TNO-rapport, R 2004/517: 72 pp.

Egea Gonzalez, F.J., Martinez Vidal, J.L., Castro Cano, M.L. and Galera Martinez, M. 1998. Levels of methamidothos in air and vegetables after greenhouse applications by gas chromatography. J. Chromatography A, 829:251-258.

FOCUS (2008). “Pesticides in Air: Considerations for Exposure Assessment”. Report of the FOCUS Working Group on Pesticides in Air, EC Document Reference SANCO/10553/2006 Rev 2 June 2008. 327 pp.

Garratt, JA; Wilkins, RM. A fugacity modelling approach to understand pesticide delivery and fate in greenhouses. Aspects of Applied Biology 2004, 71(2), 449-456.

Holterman, H.J.; Zande, J.C. van de, 1995. IDEFICS a computer model to quantify spray drift from field sprayers In: S.J.H. Crum en J.W. Deneer (eds) Workshop on the TOXSWA fate model. SC-DLO report 105: 59-63.

Holterman, H.J.; Michielsen, J.M.G.P.; Zande, J.C. van de, 1998. Spray drift in crop potection: validation and usage of a drift model Proceedings of the International Conference on Agricultural Engineering. Oslo, 24-28 August, Part 2 Paper no. 98-A-012: 577-578

Holterman, H.J., 2006. Off-target dust deposition for two granule application techniques: calculations based on the IDEFICS spray drift model. Aspects of Applied Biology, 77: 205 - 214.

Kittas, C., Boulard, T., Mermier, M. and Papadakis, G. 1995. Wind induced air exchange rates in a greenhouse tunnel with continuous side openings. J. Agric. Engineering Research 65:37-49.

Kittas, C., Boulard, T. and Papadakis, G. 1997. Natural ventilation of a greenhouse with ridge and side openings: sensitivity to temperature and wind effects. Trans. ASAE, 40: 415-425.

Lebeau, F., 2004. Modelling the Dynamic Distribution of Spray Deposits. Biosystems Engineering, 89(3): 255–265

Leistra, M and Crum, S.J.H., 1990. Emission of methyl isothiocyanate to the air after application of metham-sodium to greenhouse soil. Water, Air and Soil Pollution, 50: 109-121.

Mensink, B.J.W.G., 2004. Pesticide emissions from greenhouses. Proposal for the risk assessment system USES. RIVM, Bilthoven, report 601450014: 52 pp.

Muñoz, P. 1998. Ventilación natural de invernaderos multitúnel. Tesis doctoral, Universitat de Lleida.

Nuyttens, D.; S. Windey; B. Sonck, 2004. Optimisation of a Vertical Spray Boom for Greenhouse Spray Applications. Biosystems Engineering, 89 (4): 417–423

Pérez-Parra, J., Baeza, E.J., Montero, J.I., López, J.C., Pérez, C. and Antón, A. 2002. Effect of vent types and insect screens on ventilation of “parral” greenhouses. Acta Hort. 614(1):393-400

Pérez-Parra, J., Baeza, E., Montero, J.I. and Bailey, B.J. 2004. Natural ventilation of parral greenhouses. Biosystems Engineering 87:89-100.

Riemens, M.M.; Dueck, T.A.; Kempenaar, C.; Holterman, H.J. 2007 The EPOP model: estimation of pesticide spray depostion and effects on terrestrial non-target plants. Proceedings of the 17th SETAC symposium, Porto, Portugal, 20-24 May 2007, Society of Environmental Toxicology and Chemistry.

Schmidt, H., Siebers, J., Meier, U. and Holdt, G., 2002. Emissionen von Pflanzenschutzmitteln aus Gewächshäusern. Mitteilungen aus der Biologischen Bundesanstalt für Land- und Forstwirtschaft. 53. Pflanzenschutztagung. Heft 390, 365.

Tanny J., Cohen S., Teitel M., 2003. Screenhouse microclimate and ventilation: an experimental study. Biosystems engineering, 8(3): 331-341.

Van Os, E., H.J. Holterman and G. Klomp, 1994. Management of emission flows of pesticides from glasshouse. Acta Horticulturae, 372: 135-141.

Van der Linden, A., 2009. Emissions by “other routes than air” from protected cultivations. RIVM, Bilthoven, Report nr 607050004: 27 pp

Vidal Martinez, J.L., Egea Gonzalez, F.J., Glass, C.R., Galera Martinez, M. and Cano Castro, M.L. 1997. Analysis of lindane, □- and □-endosulfan and sulfate in greenhouse air by gas chromatography. J. Chromatography A, 765:99-108.

Zande, J.C. van de; Holterman, H.J.; Michielsen, J.G.P.; Stallinga, H., 2006. Temporal and spatial variability of spray drift around a sprayed field. Aspects of Applied Biology, 77 :295 - 302.