GEM model for soilless cultures: review of process descriptions and suggestions for improvement

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Abstract UK The GEM model developed for soilless cultures consists of different submodels (A) for applications to crops grown on mats by drip irrigation, (B) for spray applications to crops grown on such mats, and (C) is for spray applications to crops grown in pots in an ebb/flood system (GEM-A, GEM-B, and GEM-C). The descriptions of the processes for pesticide behaviour in these submodels were reviewed, considering also their consistency with measurements available in the literature. For GEM-A it is recommended to include sorption to the mats, the foil surrounding the mats and the irrigation pipes and to include partitioning between the water in the mats and the plant roots. For GEM-B it is recommended to include direct contamination of the substrate mats and the troughs resulting from spray and Low Volume Mister (LVM) applications. For GEM-B and GEM-C it is recommended (i) to revise the procedures for calculating the initial concentrations in the air and the condensation water, (ii) to include deposition onto the roof by spray and LVM applications, (iii) to revise the procedure for calculating the volatilisation rates from the plant surfaces. For GEM-C it is recommended (i) to omit the sorption equilibration between the bottom 10 cm of the soil in the pots and the water on the ebb/flood tables, (ii) to revise the procedure for the flux in the gas phase between the greenhouse air and the top layer of the soil in the pots, and (iii) to use a crop-specific value for the fraction of the surface area covered by the pots.

Keywords: soilless cultures, pesticides, emissions to surface water

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Wageningen Environmental Research (WENR) values the quality of our end products greatly. A review of the reports on scientific quality by a reviewer is a standard part of our quality policy.

This report is a product of and has been approved by the working group on the development of scenarios for substrate cultivation in glasshouses in the Netherlands

Approved team leader responsible for the contents,

name: M. de Potter

date: June 2019

Preface

We thank Ton van der Linden (RIVM) for providing clarifications on the concepts in GEM in the summer of 2016 and for his input to the discussion on Annex 2 in autumn 2017. We thank also Marieke van der Staaij (WUR Greenhouse Horticulture) for providing help on the background of the assumptions with respect to the initial percentage of the dose in the air. Furthermore we thank Cecilia Stanghellini (WUR Greenhouse Horticulture) for discussions on the value to be used for the laminar boundary layer resistance.

Summary

The Greenhouse Emission Model (GEM) for soilless cultivation is used in the regulatory context from 2016 onwards. New insights based on experimental studies and users feedback led to the need for reviewing the underlying model concepts.

In this study model concepts were assessed that describe the fate of pesticides in soilless cultivation, considering the consistency with measurements available in the literature. Additionally, recommendations were given for model improvement. The effect of the proposed changes to the model will have to be assessed after the implementation of these changes in GEM.

The GEM model for soilless cultivations consists of three different model types:
GEM-A is for applications by drip irrigation with the nutrient solution to crops grown on slabs or bags (stonewool, coir, perlite etc.)
GEM-B is for spray applications to crops grown on such slabs
GEM-C is for spray applications to crops grown in pots in an ebb/flow system on tables or floors.

Below a summary is given of the main conclusions and recommendations per model type and aspect.

GEM-A GEM-B	**Sorption** For strongly adsorbing substances sorption to substrate, foil and irrigation pipes may significantly reduce the concentration in the water. It is recommended to include these sorption types in the model. GEM does not include partitioning of the pesticide in the plant roots. This process may have a considerable effect on the concentration in the water, though. It is recommended to include this process in the model and to collect supporting information on the time course of the mass of roots in substrate systems.
GEM-B GEM-C	**Fogging** Fogging is not common anymore in the Netherlands and has been replaced by so-called Low Volume Misting (LVM). Therefore we recommend to not consider this method anymore in future versions of the model.
GEM-B GEM-C	**Initial concentration in greenhouse air: spray applications** GEM-B and GEM-C assume that immediately after a spray application 8% of the dose is present in the air of the greenhouse. This may lead to exceedance of the saturated vapour pressure in the greenhouse air and of the water solubility in the condensation water which is not desirable and not plausible. The 8% was derived from measurements of cumulative emission of parathion-ethyl from a greenhouse over a period of about 13 h, while the model assumes that the 8% arrives instantaneously in the greenhouse air. Literature data point to lower fraction directly after application. We recommend to assume that the applied substance can only volatilize after being deposited to the plant, floor and roof first. Hence, the fraction which is applied directly to the air during spraying is (initially) zero.
GEM-B GEM-C	**Initial concentration in greenhouse air: LVM applications** For applications by the low volume mister (LVM) GEM-B and GEM-C assume that 35% of the dose is present in the air plus the condensation water immediately after application. This may lead to exceedance of the saturated vapour pressure in the greenhouse air and of the water solubility in the condensation water which is not desirable and not plausible. This number is based on about the same type of measurement as for the spray application in the previous paragraphs. We recommend to assume that the air contains initially a saturated vapour concentration and that the condensation water is initially free of pesticide.

GEM-B GEM-C

Direct deposition onto the mats and the troughs

GEM-B assumes that there is no direct deposition to the substrate mats and the troughs. However, an experiment carried out in a Dutch greenhouse for a spray application showed that this contamination route cannot be excluded. It is recommended to include this direct contamination in GEM-B, assuming that 1% of the dose ends up in the substrate and 0.3% in the troughs. These numbers are based on a limited experimental data.

GEM-B GEM-C

Direct deposition onto the roof

GEM-B and GEM-C ignore deposition onto the roof of the greenhouse during and immediately after spray applications or LVM applications. Measurements indicate that this deposition may be up to 1.5% of the applied dose. We recommend to include this process by assuming 0.1% deposition for spray applications and 1% deposition for LVM applications.

GEM-C

Direct deposition on the tables

GEM-C includes a direct contamination of the water on the tables close to 10% of the dose for spray and LVM applications. This is based on the fraction of the surface area of the tables that is not covered by the surface of the pots. It is likely that this direct contamination is a large source of contamination of the circulating water. We recommend therefore to check this approach by measurements for a few pot crops and for both spray and LVM applications. Furthermore we recommend to introduce a crop-specific value of the fraction of the surface area that is covered by the surface of the pots.

GEM-B GEM-C

Volatilization from the plant surfaces and the floor

GEM calculates the volatilisation rate from plant surfaces based on the assumption that the concentration in the air at the plant surface corresponds with the saturated vapour pressure. This has the consequence that in the first days after application the concentration in the air of the greenhouse is under most circumstances close to this saturated vapour concentration. However, measurements from different sources (Netherlands, Germany, Greece) indicate that the concentration in the air the first day after application is much lower than this saturated vapour concentration. We recommend therefore to revise this assumption.

In addition, we recommend to replace this reference mass per surface area by the mass per surface area deposited onto the plants and the floor during application.

GEM-B GEM-C

Deposition from the gas phase

GEM includes the description of deposition of pesticide onto the floor and plant surfaces resulting from diffusion from the gas phase. We recommend to omit this deposition process because this deposition is likely to have little influence on the concentration in the recirculation water.

GEM-C

Sorption equilibrium for pots on tables

GEM-C assumes that the water on the tables is in sorption equilibrium with the bottom 10 cm of the soil in the pots. However, there is almost no flow of water from the pots to the tables. Therefore we recommend to omit this sorption process.

GEM-C

Deposition and volatilization from pots on tables

GEM-C includes a flux between the air of the greenhouse and the soil in the pots based on the assumption that all pesticide molecules in the pots are concentrated in the top 2 mm and that the concentration in this top 2 mm is constant. We recommend to replace this approach by a sub-model that describes diffusion in the liquid and gas phase in the top 5 cm layer.

Samenvatting

Het Greenhouse Emission Model (GEM) voor substraatteelten is vanaf 2016 in gebruik als milieurisico-beoordelingsinstrument voor de registratie van gewasbeschermingsmiddelen (GBM). Naar aanleiding van experimentele toetsingen en feedback van gebruikers is besloten de concepten die ten grondslag liggen aan het gedrag van GBM in substraatteelten te evalueren en indien nodig verbeteringen voor te stellen.

In deze studie zijn de GEM modelconcepten geëvalueerd. Wanneer mogelijk zijn de concepten vergeleken met metingen uit de wetenschappelijke literatuur. Daarnaast zijn er aanbevelingen gedaan voor verbetering van de concepten per modeltype. Het effect van de voorgestelde wijzigingen zal moeten blijken uit een gevoeligheidsanalyse na aanpassing van GEM.

Het GEM model voor substraatteelten bestaat uit drie verschillende model typen:
GEM-A is voor toedieningen van middelen via druppelirrigatie aan gewassen die op matten groeien (steenwol, kokos, perliet etc)
GEM-B is voor toedieningen van middelen via spuiten aan gewassen die op dergelijke matten groeien
GEM-C is voor toedieningen van middelen via spuiten aan gewassen die groeien in potten in eb/vloed systemen.

Hierna volgt per model type en model aspect een samenvatting van de belangrijkste conclusies en aanbevelingen.

GEM-A GEM-B	**Sorptie** Er kan sorptie optreden aan het substraat in de matten, de folie die de matten omhult en de irrigatieleidingen. Deze sorptie typen zijn nu geen onderdeel van GEM. Omdat sorptie, met name voor sterk adsorberende stoffen, de concentratie in het water kan verlagen bevelen we aan om deze sorptie typen op te nemen in het model. GEM houdt geen rekening met een verdeling van het middel tussen de wortels en het water. Dit proces kan een aanzienlijk effect hebben op de concentratie in het water. Daarom bevelen we aan om dit proces op te nemen en om ondersteunende informatie te verzamelen over het verloop van de wortelmassa met de tijd in substraatsystemen.
GEM-B GEM-C	**Toepassingsmethode thermisch vernevelen** Ruimtebehandeling via thermisch vernevelen (foggen) is niet meer gebruikelijk in Nederland en is vervangen door de zogenaamde Low Volume Mister (LVM). We bevelen aan om deze methode niet meer mee te nemen in volgende versies van GEM.
GEM-B GEM-C	**Initiële concentratie in kaslucht: spuittoepassingen** GEM-B en GEM-C nemen aan dat meteen na een spuittoepassing altijd 8% van de dosering in de lucht van de kas aanwezig is. Dit kan in het model leiden tot overschrijden van de verzadigde dampdruk in de kaslucht en van de wateroplosbaarheid in het condensatiewater hetgeen in werkelijkheid niet zal gebeuren. Deze 8% is afgeleid uit eerdere metingen van de cumulatieve emissie van parathion-ethyl uit een kas gedurende 13 u, terwijl het model aanneemt dat deze 8% instantaan in de kaslucht aanwezig is. Metingen in de literatuur wijzen daarnaast op lagere fracties in de lucht na een spuittoepassing. We bevelen aan om aan te nemen dat alle gespoten middel neerslaat en dat er dus geen middel in de lucht of het condenswater aanwezig is direct na toepassing. Vervolgens zal het neergeslagen middel in de kaslucht terechtkomen door vervluchtiging.

GEM-B GEM-C

Initiële concentratie in kaslucht: LVM – toepassingen

GEM-B en GEM-C nemen voor toedieningen met LVM aan dat meteen na toepassing 35% van de dosering in de lucht van de kas plus het condensatiewater op het dak van de kas zit. Dit kan in het model leiden tot overschrijden van de verzadigde dampdruk in de kaslucht en van de wateroplosbaarheid in het condensatiewater. Dit zal in werkelijkheid niet voorkomen. Er zijn niet veel gegevens beschikbaar uit de literatuur over de concentratie in lucht en condenswater maar het is aannemelijk dat de lucht de verzadigde-damp concentratie van het middel bevat en dat het condenswater initieel geen middel bevat.

GEM-B GEM-C

Directe depositie op matten en goten

Directe depositie op de substraatmatten en de goten zit niet in GEM. Een experiment in een Nederlandse kas met een spuittoepassing heeft echter aangetoond dat deze route wel degelijk bestaat bij spuittoepassingen. We bevelen aan om deze directe verontreiniging in GEM-B op te nemen, aannemend dat 1% van de dosering in de substraatmatten terecht komt en 0.3% in de goten. De voorgestelde waarden zijn gebaseerd op een beperkt aantal metingen.

GEM-B GEM-C

Directe depositie op het dak van de kas

Uit metingen uit de literatuur blijkt dat directe depositie op het dak tot 1.5% van de dosering kan oplopen. Er wordt nu geen rekening gehouden met depositie op het dak. We bevelen daarom aan om dit proces op te nemen in GEM, aannemend dat er 0.1% depositie is voor spuittoedieningen en 1% voor LVM-toedieningen.

GEM-C

Directe depositie op de tafels

GEM-C houdt rekening met een directe verontreiniging van het water op de tafels van ongeveer 10% van de dosering voor zowel spuit- als LVM-toedieningen. Dit is gebaseerd op de fractie van het oppervlak van de tafels die niet bedekt is door de potten. Waarschijnlijk is deze directe verontreiniging verreweg de grootste bron van verontreiniging van het circulerende water. Daarom bevelen wij aan om deze directe verontreiniging te meten voor enkele gewassen die in potten groeien voor zowel spuit- als LVM-toepassingen. Verder bevelen wij aan om een gewas-specifieke waarde te gaan gebruiken voor de fractie van het oppervlak van de tafels die niet bedekt is door de potten.

GEM-B GEM-C

Vervluchtiging vanaf het blad van de plant

De berekening van de vervluchtigingssnelheid vanaf plantoppervlakken is gebaseerd op de aanname dat de concentratie in de lucht aan het plantoppervlak overeen komt met de verzadigde dampdruk. Uit metingen blijkt dat de concentratie in de kaslucht op de eerste dag na toediening veel lager is. We bevelen daarom aan om een reductiefactor te introduceren. De introductie van deze factor zal leiden tot verlaging van de vervluchtigingssnelheid.

Ook bevelen we aan om de referentiemassa per oppervlak te vervangen door de massa per oppervlak die direct na toediening op de planten terecht is komen (dit geldt ook voor de vloer).

GEM-B GEM-C

Depositie vanuit de gasfase

GEM houdt rekening met de depositie van middel op de vloer en plantoppervlakken ten gevolge van depositie vanuit de gas fase. Op basis van de nieuwe inzichten vindt er direct bij toediening depositie plaats op de verschillende oppervlakken (dak, blad etc.) en daarna vervluchtiging. We bevelen aan om deze depositie uit het model te halen omdat het weinig effect zal hebben op de concentratie in het recirculatie water.

GEM-C **Sorptie-evenwicht op tafels met potten**

GEM-C neemt aan dat het water op de tafels in sorptie-evenwicht is met de onderste 10 cm van de grond in de potten. Er vindt echter vrijwel geen waterstroming plaats van de potten naar de tafel. Daarom bevelen we aan om dit sorptieproces uit het model te verwijderen.

GEM-C **Depositie en vervluchtiging vanuit de potten**

GEM-C bevat een flux tussen de kaslucht en de grond in de potten gebaseerd op de aanname dat alle middel in de potten geconcentreerd is in de bovenste 2 mm en dat de concentratie in deze bovenste 2 mm constant is. Wij bevelen aan om deze benadering te vervangen door een sub-model dat diffusie in de vloeibare en de gasfase beschrijft in de bovenste 5 cm.

1 Introduction

The GEM model for soilless cultivations in greenhouses was developed in 2015 (Van der Linden *et al.*, 2015). In the past years, experience was gained with this model for regulatory use and the model was also tested (Van der Linden *et al.*, 2017; Wipfler *et al.*, 2019). Furthermore, sorption experiments were carried out with two pesticides and substrate materials (Boesten & Matser, 2017). The insights gained in this work led to the need to review the model and propose improvements to the model concepts in GEM. This work is reported in this report.

The GEM model for soilless cultivations consists of three different models (van der Linden *et al.*, 2015), further called GEM-A, GEM-B and GEM-C:

- GEM-A is for applications by drip irrigation to crops grown on slabs or bags (stonewool, coir, perlite etc.); see Figure 1
- GEM-B is for spray applications to crops grown on such substrates
- GEM-C is for spray applications to crops grown in pots in an ebb/flow system on tables or floors.

The proposed improvements consist of inclusion of new processes (e.g. sorption to stonewool) or of improved descriptions of processes that were already described in the GEM model. This report only describes the review and the model improvements. For an overview of the current GEM model concepts we refer to van der Linden *et al.* (2015) and Wipfler *et al.*(2015b).

The report is structured as follows. Chapters 2, 3 and 4 contain reviews of GEM-A, GEM-B and GEM-C, respectively, and chapters 5, 6, and 7 describe the proposed improvements for these three models.

Annex 3 provides an overview of the usefullness of OECD-309 studies for estimating the degradation rate in the recirculation water in GEM. This information may be useful in the longer term for developing additional guidance for estimation of this degradation rate.

Figure 1 *Tomato plants (left) and sweet pepper plants (right) growing on stonewool. On the right photograph the drip irrigation tubes are clearly visible (photographs by WUR Greenhouse Horticulture).*

2 Partitioning processes in the recirculating water in GEM-A and GEM-B

2.1 Sorption to substrate material and foil

GEM-A and GEM-B do not describe sorption to the substrate material and the foil surrounding the substrate mats. Boesten & Matser (2017) found significant sorption of the two isomers of dimethomorph to these materials. Therefore we propose here concepts for describing the sorption to these materials in GEM-A and GEM-B.

The GEM-A and GEM-B models consist of a system of perfectly mixed tanks/reservoirs (Van der Linden *et al.*, 2015). The cultivation part including substrate and pipes is one of these tanks. Let us assume that this tank contains a volume of water V_{sub} (m^3), a dry mass of substrate (e.g. stonewool) M_{sub} (kg), and a mass of foil M_{foil} (kg). We define the concentration in the water in the tank as c_w (kg/m^3), the mass of pesticide sorbed per dry mass of substrate as X_{sub} (kg/kg) and the mass of pesticide sorbed per mass of foil as X_{foil} (kg/kg). We assume linear sorption isotherms:

$$X_{sub} = K_{sub}\ c_w \qquad \text{Eqn 1}$$

$$X_{foil} = K_{foil}\ c_w \qquad \text{Eqn 2}$$

where K_{sub} (m^3 kg^{-1}) and K_{foil} (m^3 kg^{-1}) are the sorption coefficients of the substrate and the foil, respectively. The total mass of pesticide in the substrate tank, m_{sub} (kg), is described by:

$$m_{sub} = V_{sub}\ c_w + M_{sub}\ K_{sub}\ c_w + M_{foil}\ K_{foil}\ c_w \qquad \text{Eqn 3}$$

So c_w can be calculated as:

$$c_w = \frac{m_{sub}}{V_{sub} + M_{sub}\ K_{sub} + M_{foil}\ K_{foil}} = \frac{m_{sub}}{1 + \frac{M_{sub}}{V_{sub}} K_{sub} + \frac{M_{foil}}{V_{sub}} K_{foil}} \qquad \text{Eqn 4}$$

Assuming that sorption data for substrate and foil are available, there is still the problem of the estimation of M_{foil} and K_{foil}. In sorption studies both sides of the foil contribute to the sorption (Boesten & Matser, 2017). Therefore we recommend to halve the measured K_{foil} values from such studies. Let us consider the isomers of dimethomorph as an example. Boesten & Matser (2017) found K_{foil} values of 1.2-1.5 dm^3/kg for foil surrounding stonewool mats, so 50% of this is 0.6-0.75 L/kg. They reported a value of 0.03 kg/dm^3 for M_{foil} / V_{sub}. So M_{foil} K_{foil} / V_{sub} is then about 0.02. Eqn 4 indicates that sorption to the foil will decrease c_w only to a very limited extend. It is furthermore likely that only the bottom part of the foil is in contact with the circulating water, so M_{foil} has to be reduced by something like a factor 2. Thus for substances like dimethomorph including sorption to the foil will decrease c_w by about 1%. It can be expected that sorption to the foil is related to the octanol-water partition coefficient, K_{ow}. Dimethomorph has a log K_{ow} of 2.6-2.7 (Boesten & Matser, 2017), so the effect may become larger for more lipophilic substances. Wipfler *et al.* (2015a) made GEM calculations for 23 pesticides applied in soilless cultures. The log K_{ow} values of these ranged between -1.5 to 6.4, their median was 3.0 and their 80th percentile was about 4.5. So about half of the pesticides had a higher log K_{ow} value than dimethomorph. Therefore we recommend to include this process in GEM-A and GEM-B.

Let us also consider the sorption of the isomers of dimethomorph to stonewool as an example. Boesten & Matser (2017) found a K_{sub} of about 1 dm^3/kg for clean stonewool and estimated K_{sub} to be

about 3 dm^3/kg for stonewool at the end of the growing cycle of sweet pepper. They measured a M_{sub} / V_{sub} ratio of 0.14 kg/dm^3 for clean stonewool and of 0.1 kg/dm^3 for stonewool at the end of the growing cycle (the ratio is lower at the end because aged stonewool retains more water). So M_{sub} K_{sub} / V_{sub} is then about 0.14 for clean stonewool and 0.3 for stonewool at the end of the growing cycle. It follows from Eqn 4 that sorption to the stonewool may lead to a concentration decrease of about 10-25% for dimethomorph. Sorption to coir is likely to be considerably higher than sorption to stonewool. So it seems worthwhile to incorporate sorption to substrate material in GEM-A and GEM-B.

2.2 Sorption to plastic pipes

The growing systems simulated by GEM-A and GEM-B consist of different reservoirs and the porous mats and these are connected by different types of tubes. The main transport tubes are PVC tubes with an inner diameter of about 3 cm. The irrigation water flows into the porous mats through polyethene tubes (inner diameter of 16-18 mm) and one polyethene capillary tube (inner diameter of about 4 mm) for each plant; these capillary tubes are about 80 cm long. GEM-A and GEM-B do not consider sorption to any of the tubes and it is a point of debate whether this sorption should be included in GEM.

Boesten & Matser (2017) measured sorption of pymetrozine and dimethomorph to the PVC transport tubes and the polyethene capillary tubes in batch experiments. For pymetrozine the sorption was found to be too low to be measurable. The sorption coefficient of the two isomers of dimethomorph was found to be 0.07 and 0.12 L/kg for the PVC transport tubes and 0.14 and 0.16 L/kg for the polyethene capillary tubes. These values have to be halved when used for simulating the behaviour of pesticides in the culture systems because in the batch experiments both the sorption to the inner and outer part of the tubes is measured whereas the pesticide is only in contact with the inner part in the culture system. So it is clear that for part of the compounds the sorption to these tubes is measurable and thus the question is to what extent the concentration in the culture systems will decrease as the result of the sorption to these tubes ?

Let us consider a system that consists of a reservoir with a volume V_{res} (m^3) and of a plastic tube with an inner surface area S (m^2) and a length L (m). The reservoir and the plastic tube are filled with water containing a pesticide and this water is pumped continuously through the tube. We assume that the sorption to the plastic tube is at equilibrium as the result of the continuous circulation. The total mass of pesticide in the system is m_{sys} (kg) and the sorption coefficient K_{tube} (m^3/kg) is defined by

$$X_{tube} = K_{tube}\, c_w \qquad \text{Eqn 5}$$

where X_{tube} is the mass of pesticide sorbed per mass of plastic (kg/kg). So K_{tube} is a distribution coefficient expressed in m^3 of water per kg of plastic. The mass balance of this system then is given by

$$m_{sys} = \left(V_{res} + V_{tube} + M_{tube}\, K_{tube}\right) c_w \qquad \text{Eqn 6}$$

where V_{tube} is the volume of water in the tube (m^3) and M_{tube} is the mass of plastic of the tube (kg). If we define p as M_{tube}/L, the expression for c_w becomes:

$$c_w = \frac{m_{sys}}{V_{res} + L\left(S + p\, K_{tube}\right)} \qquad \text{Eqn 7}$$

Thus the effect of the sorption to the tubes on c_w can be assessed by considering the quotient Q defined as:

$$Q = \frac{V_{res} + L\, S}{V_{res} + L\left(S + p\, K_{tube}\right)} \qquad \text{Eqn 8}$$

So Q gives the ratio of the concentration in the water without considering sorption divided by the concentration including sorption to the plastic tubes. For a system with three different types of tubes the expression for Q becomes

$$Q = \frac{V_{res} + L_1S_1 + L_2S_2 + L_3S_3}{V_{res} + L_1S_1 + L_2S_2 + L_3S_3 + L_1p_1K_{tube,1} + L_2p_2K_{tube,2} + L_3p_3K_{tube,3}} \quad \text{Eqn 9}$$

where the indices 1, 2, 3 indicate the three different types of tube. It then follows that the relative contribution to the concentration decrease of the sorption to each type of tube is given by its $L\ P\ K_{tube}$ product.

Let us now consider a realistic culture system, i.e. the system used by Wipfler *et al.* (2019) to test the GEM model. The volume of water in the mats (V_{res}) was about 800 dm^3. The lengths and diameters of the three types of pipe were: (1) the main transport PVC pipes had an inner diameter of 28 mm and a total length of 15 m, (2) the polyethene irrigation tubes had an inner diameter of 16 mm and a total length of 12 × 13.2 = 158 m, (3) the polyethene capillary tubes had an inner diameter of 4 mm and a total length of 12 × 13.2/0.25 × 0.8 = 507 m. So the volumes of the three types of tube (i.e. L_1S_1, L_2S_2, L_3S_3) were 9, 32 and 6 L, respectively. The p value was 0.24 kg/m for the PVC pipes, 0.074 kg/m for the irrigation tubes and 0.015 kg/m for the capillaries (based on Boesten & Matser, 2017 for the PVC pipes and the capillaries; the value for the irrigation tubes was measured for this report). The sorption coefficient for the polyethene irrigation tubes ($K_{tube,2}$) can be expected to be equal to that of the polyethene capillaries ($K_{tube,3}$) because these are made of the same material. Based on the experiments with dimethomorph we assume that $K_{tube,1}$ = (0.095/0.15) $K_{tube,3}$ = 0.63 $K_{tube,3}$. We can then calculate Q as a function of $K_{tube,3}$ for this system.

Figure 2 shows that a significant decrease of the concentration will occur only if the K_{tube} of the polyethene tubes is higher than about 1 dm^3/kg. For dimethomorph this K_{tube} was estimated at half the measured values described above, so 0.075 dm^3/kg. This K_{tube} gives Q = 0.998, so a negligible decrease. It can be expected that K_{tube} increases with increasing octanol-water coefficient (dimethomorph has an octanol-water coefficient of about $10^{2.7}$). Some pesticides have considerably higher octanol-water coefficients. So we recommend to include this sorption to plastic pipes in GEM but to set K_{tube} default to zero and to require that non-zero values of K_{tube} are based on sorption measurements of the pesticide considered.

It is also interesting to consider the relative contribution of the three types of pipes to the concentration decrease. Eqn 9 shows that this is given by the ratios $L_1p_1K_{tube,1}$: $L_2p_2K_{tube,2}$: $L_3p_3K_{tube,3}$. Using the above parameter value these ratios are 19 : 100 : 65, so the polyethene irrigation tubes have the largest contribution and the PVC tubes the smallest. So estimates of K_{tube} can best be based on measurements of the sorption to the polyethene irrigation tubes and the polyethene capillary tubes.

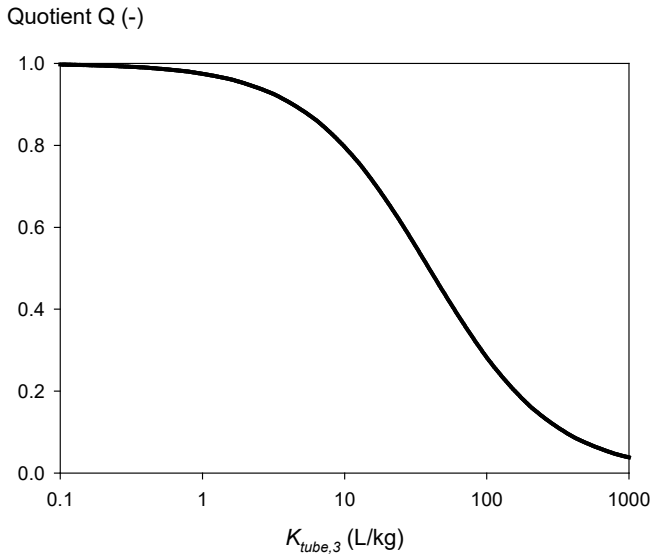

Figure 2 *Effect of the pesticide sorption coefficient of the capillary polyethene tubes ($K_{tube,3}$) on the quotient Q as calculated with Eqn 9 for the parameter values described in the text.*

2.3 Partitioning into plant roots

Boesten & Matser (2017) made exploratory calculations on the significance of partitioning of pesticides into the roots and found that this may lead to some 30% decrease in the concentration of the isomers of dimethomorph at the end of a sweet-pepper growing cycle. It seems therefore worthwhile to develop a procedure to incorporate this process in GEM.

These exploratory calculations were based on Briggs *et al.* (1982) who established the following relationship between partitioning of pesticides into roots and the octanol-water partition coefficient:

$$RCF = 8.2 \times 10^{-4} + 3.02 \times 10^{-5} \left(K_{ow}\right)^{0.77} \qquad \text{Eqn 10}$$

where *RCF* is the root concentration factor (m^3/kg) defined as the concentration in the roots divided by the concentration in the external solution (with concentration in roots defined as mass of pesticide in roots per mass of wet roots), and K_{ow} is the octanol-water distribution coefficient (-). For e.g. dimethomorph ($K_{ow} = 10^{2.65}$) this equation gives a RCF of about 4 dm^3/kg. So *RCF* is defined by:

$$RCF \equiv \frac{\mu}{c_w} \qquad \text{Eqn 11}$$

where μ is mass of pesticide in roots per mass of wet roots (kg/kg). Briggs *et al.* (1982) measured only uptake from nutrient solutions and did not test whether this partitioning into the plant roots is a reversible process (i.e. will pesticide molecules that are taken up by roots return back to the aqueous phase if the concentration in this phase decreases). There may be information available in the literature on this reversibility but it is beyond the scope of this report to check this. Furthermore there may be degradation in the plant roots. Assuming that it is a reversible process without degradation in the roots seems to be a conservative approach for soilless cultures because all pesticide molecules taken up by the roots may also be released again into the water in the substrate system. Therefore we propose to incorporate it as a reversible process. The total mass in the substrate tank (including sorption to the substrate) becomes then:

$$m_{sub} = V_{sub}\, c_w + M_{sub}\, K_{sub}\, c_w + M_{roots}\, RCF\, c_w \qquad \text{Eqn 12}$$

where M_{roots} is the mass of wet roots in the system (kg).

This gives the following expression for c_w

$$c_w = \frac{m_{sub}}{1 + \frac{M_{sub}}{V_{sub}} K_{sub} + \frac{M_{roots}}{V_{sub}} RCF} \qquad \text{Eqn 13}$$

Boesten & Matser (2017) estimated tentatively a M_{roots}/V_{sub} ratio of 0.1 kg/dm^3 at the end of the sweet-pepper growing cycle. So, taking again dimethomorph as an example, $M_{roots}\,RCF\,/V_{sub}$ becomes then 0.4. The above equation shows that in this case partitioning into the plant roots may decrease the concentration in the water distinctly. Therefore we recommend to incorporate this partitioning in GEM-A and GEM-B and to collect data on the course of time of the development of the mass of wet roots in soilless cultures.[1]

[1] In this context the following information may be relevant (personal communication Chris Blok, 2018): the fraction of dry material of roots ranges between 10 and 12%; the root mass increases the first six week up to a maximum value and remains thereafter fairly constant; in case of measurement of dry roots mass by ignition, it has to be kept in mind that fresh stonewool contains 1.5% organic material (binder).

3 Review of processes influencing the concentration in the condensation water in GEM-B

3.1 Overview of GEM-B

According to van der Linden *et al.* (2015), the pesticide is in GEM-B applied by spraying, fogging or fumigation to a crop grown on a substrate system (e.g. stonewool).

Currently, application by fogging hardly occurs anymore in greenhouses in the Netherlands and it is not 100% clear which types of application are included in fumigation. Almost all pesticide applications in the context of GEM-B are either spraying or room treatments with the low-volume mister (LVM); see **Figure 3** and **Figure 4**). Therefore we recommend to limit the application methods in GEM-B to spraying or LVM. The most important characteristics of these application methods are as follows. Pesticides are sprayed in typically 800-2000 L water per ha, leading to considerable dripping of spray liquid from the leaves. When pesticide is sprayed, the intention is to spray both the top and the underside of the leaves (so part of the nozzles is directed to the underside); however, the retention of spray liquid on the top of the leaves will be better than on the underside of the leaves because more liquid will drip from the underside, leading to e.g. 75% of the dose ending up on top of the leaves and 25% ending up on the underside of the leaves; see van Os *et al.* (2004, 2005) for comparisons between top and underside in chrysanthemum and tomatoes. Applications by the LVM take place by blowing spray liquid into a strong air stream, generating very fine spray droplets. The principle of the LVM application is that the cloud of droplets that is thus generated will be deposited on the top of the leaves within about 1 hour after application. The amount of spray liquid used with LVM is about 200 L/ha. The consequence is that LVM applications will not lead to deposition on the underside of the leaves.

Figure 3 *Equipment for spray applications (left) and a spraying event (right) in a greenhouse where tomatoes are grown on stonewool (photographs by WUR Greenhouse Horticulture).*

Figure 4 *Equipment for applications with the low volume mister (photograph by WUR Greenhouse Horticulture).*

The model distinguishes the following compartments in the greenhouse containing pesticide:
- the air (60 000 m^3 ha^{-1} in the scenario considered)[2].
- the condensation water present on the roof (0.532 m^3 ha^{-1} in the scenario considered)
- the plant surface
- the greenhouse floor
- the recirculation water in the substrate (the mats) on which the crop is grown
- the recirculation water in a number of tanks.

The interaction between these compartments via the air is schematically shown in **Figure 5**.

The condensation water on the walls of the greenhouse is not included because this flows into the soil (and thus does not become part of the recirculating water).

It is assumed the pesticide can enter the water in the substrate only via de condensation water flowing from the roof. So direct contamination of the circulating water e.g. by stem flow shortly after application is ignored.

[2] Van der Linden *et al.* (2015) gave 50 000 m^3/ha but this appeared not to be correct.

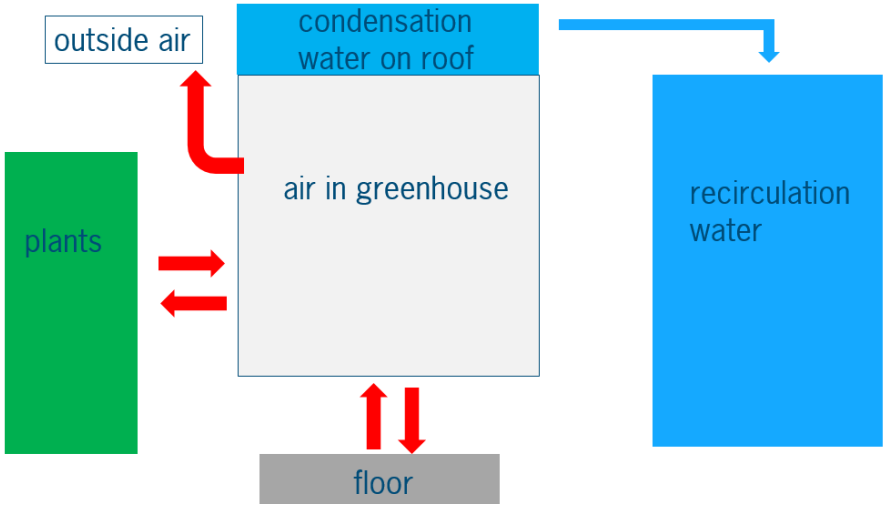

Figure 5 *Schematic representation of the exchange between the different compartments via the air in the greenhouse. The condensation water is assumed to be present on the roof. The red arrows are gas fluxes and the blue arrow is a water flux.*

The model assumes three types of surfaces in the greenhouse for exchange of pesticide with the greenhouse air (**Figure 5**): the plants, the greenhouse floor and the greenhouse roof where the condensation water is present.

The model includes simulation of the following quantities as a function of time:
- the mass of pesticide on the plants per surface area of greenhouse, A_p (kg/m^2)
- the mass of pesticide on the greenhouse floor per surface area of greenhouse, A_f (kg/m^2)
- the sum of the mass of pesticide in the greenhouse air and the mass of pesticide in the condensation water on the greenhouse roof, again per surface area of greenhouse, A_{a+w} (kg/m^2).

Based on the above quantities, the model simulates the following concentrations in the greenhouse:
- the concentration in the air at the plant surface, $c_{a,p}$ (kg m^{-3}),
- the concentration in the air at the floor surface, $c_{a,f}$ (kg m^{-3}),
- the concentration in the greenhouse air, $c_{a,g}$ (kg m^{-3}),
- the concentration in the condensation water on the roof, $c_{w,cds}$ (kg m^{-3})
- the concentration in a number of recirculation tanks.

It is assumed that $c_{a,p}$ and $c_{a,f}$ correspond with the saturated vapour pressure at the prevailing greenhouse temperature, as long as A_p and A_f exceed zero; otherwise $c_{a,p}$ and $c_{a,f}$ are zero as well.

The GEM-B model assumes instantaneous equilibrium between the air in the greenhouse and the condensation water present on the roof of the greenhouse. The model does not include separate simulation of the mass of pesticide in the greenhouse air, so if initially part of the dose stays in the greenhouse air, this means that the areic pesticide mass is added to A_{a+w}. The concentration $c_{a,g}$ is calculated from A_{a+w} assuming that the ratio of $c_{a,g}$ and $c_{w,cds}$ equals that of the concentrations corresponding with a saturated vapour pressure and water solubility using Henry's law.

Figure 6 shows that almost 100% is present in the condensation water for air-water partitioning coefficients below 10^{-7}; at a air-water partitioning coefficient of 10^{-5} there is about 50% in the air and 50% in the condensation water and at a air-water partitioning coefficients below 10^{-3} only a negligible fraction is present in the condensation water.

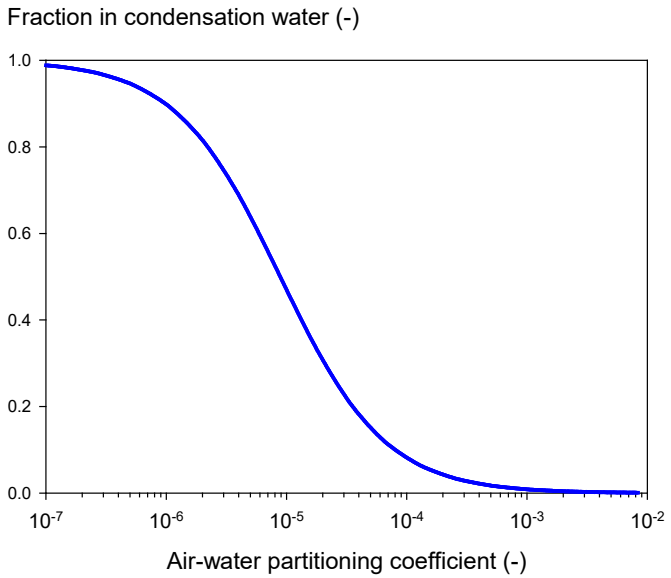

Figure 6 *Fraction of total mass of pesticide present in condensation water as a function of the air-water partitioning coefficient considering the total mass in a volume of air of 60,000 m^3 and a volume of condensation water of 0.532 m^3.*

3.2 Initial distribution of the dose

The application procedure assumes that part of the pesticide is deposited on the plants, that another part is deposited on the greenhouse floor and that the remaining part stays in the greenhouse air (Appendix D of van der Linden *et al.*, 2015). The distribution of the dose over these three compartments was not reported by van der Linden *et al.* (2015), so they are given Table 1 for the spray applications and in **Table 2** for the applications by fogging or the low volume mister. The fraction deposited on the floor was based on expert knowledge; this fraction does depend both on the density of the plants and the amount of leaves.

Van der Staay (personal communication, 2017) indicated that the 8% staying in the greenhouse air for the spray applications (Table 1) was based on measurements by Baas & Huygen (1992). Baas & Huygen (1992) measured concentrations in the air in a greenhouse after a high-volume spray application of parathion-ethyl (which has a saturated vapour pressure of 0.9 mPa at 25°C and a water solubility of 12 mg/L) to a tomato crop over at least 13 h after application (see their p. 54). The dose was 2.54 g for a surface of 256 m^2, so about 0.1 kg/ha. The value of 8% can be found in Table 19 of Baas & Huygen (1992); this is the cumulative emission (as a percentage of the dose) based on the measured concentrations over these at least 13 h and the flow rate of the air out of the greenhouse. The emission was 1% in the first hour, 2% in the next three hours, 2% in the following hour and 3% in the last period (at least 8 h; see their Table 19). In the GEM model it is assumed that these 8% are in the air immediately after application and that these are instantaneously in equilibrium with the 0.532 m^3 condensation water per ha. The measured concentration of parathion-ethyl in the gas phase immediately after application was at most 0.02 mg/m^3 (Table 4.14 of Baas & Huygen), i.e. considerably below the concentration corresponding with the saturated vapour pressure (0.11 mg m^{-3}). The volume of the greenhouse was 901 m^3 (p. 20 of Baas & Huygen); so the mass in the air immediately after application was 18 mg. The dose was 2.54 g (p. 33) so only 0.7% of the dose was in the gas phase immediately after application. This indicates that use of this 8% from Table 19 of Baas & Huygen to estimate the initial mass in the air is difficult to defend.

The concentration staying in the air (in the form of molecules in the gas phase) cannot be much higher than that corresponding with the saturated vapour pressure. The concentration in air of a saturated vapour, $c_{a,sat}$ (kg m^{-3}), can be calculated from

$$c_{a,sat} = \frac{m_{mol}\, P_{sat}}{R\, T}$$ Eqn 14

where m_{mol} is the molar mass (kg mol^{-1}), P_{sat} is the saturated vapour pressure (Pa), R is the gas constant (8.31 J mol^{-1} K^{-1}) and T is the absolute temperature (K). GEM-B assumes an air volume of 60 000 m^3/ha. **Figure 7** shows that a vapour pressure of more than 10^{-2} Pa (so 10 mPa) corresponds with a pesticide mass of 0.08 kg in this air volume (i.e. 8% of a dose of 1 kg/ha). The vast majority of pesticides has a lower saturated vapour pressure than 10 mPa (Boesten *et al.*, 2018). So this 8% does not seem to be a realistic value. We recommend therefore to modify this approach.

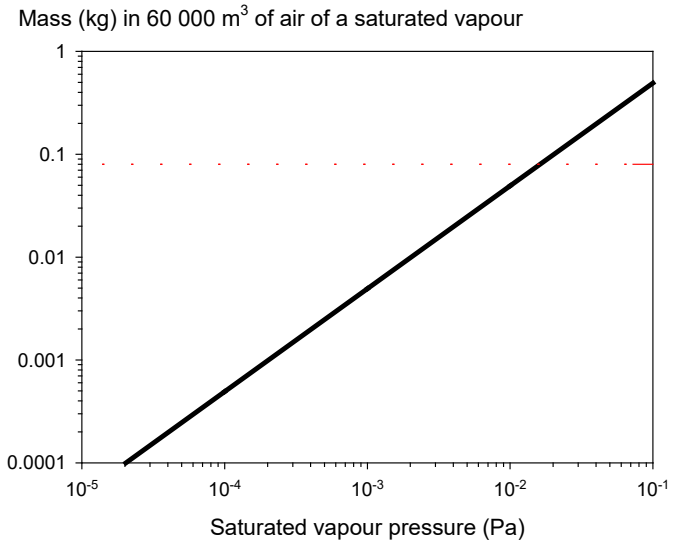

Figure 7 *Mass of substance present in 60 000 m^3 of air that contains this substance at its saturated vapour pressure shown as a function of this saturated vapour pressure. The horizontal dashed line corresponds with 0.08 kg.*

The source for the 35% staying in the air for the applications by fogging or the low volume mister (Table 2) is not clear. Application by fogging does not happen anymore in the Netherlands (personal communication Erik van Os, 2018) so we focus here on the low volume mister and recommend to exclude application by fogging from GEM-B. Baas & Huygen (1992) applied parathion-ethyl by a 'ruimtebehandelingstechniek' (Low Volume Mister, abbreviated LVM) at a rate of about 0.1 kg/ha. For the Low Volume Mister they report a cumulative emission of 23% for parathion-ethyl in their Table 19. They found a maximum concentration of gas plus droplets in the air of 1.74 mg/m^3 for this application technique (their Table 4.8) which corresponds with 56% of the dose of 2.8 g. These percentages seem inconsistent at first glance but emission is the product of the flow of air out of the greenhouse and the concentration in the greenhouse. For the Low Volume Mister they found a maximum concentration in the gas phase of 0.95 mg m^{-3}. The concentration corresponding with the saturated vapour pressure was about 0.11 mg m^{-3}. So the exceedance of this saturated concentration is considerable. However, Baas & Huygen (1992) did not discuss the possible causes of this exceedance. Probably such a large exceedance of the saturated vapour pressure is an artifact from the measurement method due to very small droplets in the air. Thus also for the low volume mister we propose to replace the assumption of 35% initially in the air by a more realistic approach.

As follows from the above, deposition of spray droplets on the walls of the greenhouse is ignored in GEM-B. However, this deposition is not relevant because the condensation water flowing from the walls does not flow into the recirculation system. Deposition of spray droplets on the roof of the greenhouse is ignored as well. This seems difficult to defend, especially for application by the low volume mister. Crum *et al.* (1988) found a deposition of methomyl on the roof of 0.07% after a spray application in cucumber. Crum *et al.* (1991) found a deposition of methomyl on the roof of 1.5% after applying with a low-volume mister and 0.05% after a spray application (greenhouse grown with tomatoes). Bor *et al.* (1994) found a deposition of parathion-ethyl on the roof of 1.2% of the dose

after applying with a low-volume mister and 0.4% after a spray application in a greenhouse grown with tomatoes. So we recommend to include deposition on the roof in GEM-B. These measurements are from 25-30 years ago when greenhouses were about 3 m high whereas current greenhouses whereas current greenhouses are 6-7 m high. It can be expected that deposition on the roof after spray applications is lower for higher greenhouses.

Van Os *et al.* (2012) recovered about 0.003% and 0.1% the dose in the condensation water (two experiments on two different days) after spraying highly soluble and non-volatile tracers to a tomato crop in a greenhouse with a gutter height of 6 m. It can be expected that the amount recovered in the condensation water is close to the deposition onto the roof under these circumstances.
The numbers are based on estimated (non-measured) volume fluxes of condensation water. Based on all this information we recommend to assume a deposition onto the roof of 0.1% of the dose after spray applications and of 1% of the dose after LVM applications. We recommend to underpin or revise these numbers by carrying out experiments in modern greenhouses.

It is not checked in GEM-B whether $c_{a,g}$ and $c_{w,cds}$ exceed the saturated vapour pressure and the water solubility, respectively. This may lead to problems immediately after application. Let us consider a spray application of 1 kg/ha. The 8% staying in the air give 0.08 kg/ha. In the scenario there is 0.532 m^3 condensation water and 60 000 m^3 air per ha. If the pesticide has a very low vapour pressure, the concentration in the condensation water becomes 0.08 kg / 0.53 m^3, i.e. 151 mg/L. Most pesticides have a lower water solubility, so for these pesticides $c_{a,g}$ may exceed the saturated vapour pressure which is not realistic. We recommend therefore to improve this aspect of GEM-B.

Table 1 *Distribution of a pesticide application by spraying over the different substance compartments in the greenhouse in GEM-B for the reference deposition crops (see Table B-2 of van der Linden et al., 2015).*

Crop type (reference deposition crops)	Fraction of applied dose		
	deposited on crop surface	deposited on floor	staying in greenhouse air
Cut flowers	0.80	0.12	0.08
Lettuce	0.80	0.12	0.08
Tomato and cucumber	0.72	0.20	0.08
Rose and gerbera	0.80	0.12	0.08
Very small young plants	0.00	0.92	0.08

Table 2 *Distribution of a pesticide application by the low volume mister over the different substance compartments in the greenhouse in GEM-B.*

Crop type (reference deposition crops)	Fraction of applied dose		
	deposited on crop surface	deposited on floor	staying in greenhouse air
Cut flowers	0.55	0.10	0.35
Lettuce	0.55	0.10	0.35
Tomato and cucumber	0.55	0.10	0.35
Rose and gerbera	0.55	0.10	0.35
Very small young plants	0	0.65	0.35

3.3 The fluxes in the gas phase

3.3.1 The flux between the air and the plant surfaces

Van der Linden *et al.* (2015) use rate equations for the exchange between the air and the plants. They use different approaches depending on the direction of the flux. This direction is based on $c_{a,g}$ and $c_{a,p}$: if $c_{a,g} > c_{a,p}$ then the flux is from the air to the plant surfaces and if $c_{a,p} > c_{a,g}$ then the flux is from the plant surfaces to the air.

It is assumed that $c_{a,p}$ corresponds with the saturated vapour pressure at the prevailing greenhouse temperature, as long as A_p exceeds zero; otherwise $c_{a,p}$ is zero as well. So let us consider a pesticide application. Immediately after application, $c_{a,p}$ will correspond with the saturated vapour pressure and this will stay so as long as A_p exceeds zero; so the switching between the flux from the plant to the air takes place as soon as A_p is rounded to zero by the computer. Such a dependence of the direction of the flux on the round-off procedure of the computer is undesirable.

Let us assume that in a future version of GEM-B, c_a cannot exceed the saturated vapour pressure at the prevailing temperature. Let us consider the first time step after application of the pesticide: A_p is zero and so the flux is from the air to the plant surfaces. However, the next time step $c_{a,p}$ equals the saturated vapour pressure and the flux will be in the opposite direction. This will continue until A_p has decreased to zero after which the flux will reverse. It seems unlikely that such frequent reversals in direction will happen in reality.

If the flux is from the air to the plants, the flux is described by

$$J_{d,p} = LAI \frac{c_{a,g} - c_{a,p}}{r_a} \qquad \text{Eqn 15}$$

where $J_{d,p}$ is the mass flux of pesticide (i.e. mass rate per surface area of greenhouse, so kg m^{-2} d^{-1}) from the air to the plants ('d' from deposition), LAI (-) is the leaf area index (which is fixed to 5)[3] and r_a is the laminar boundary layer resistance (d m^{-1}).

Van der Linden *et al.* (2015) wrote that they used for the laminar boundary layer resistance, r_a, a value of 1.16 $\times 10^{-3}$ d/m based on Jacobs *et al.* (2007) which use the unit s/m, so the value becomes 100 s/m. Jacobs *et al.* (2007) report this resistance of 100 s/m once but for the sum of (1) the aerodynamic resistance for the turbulent layer and (2) the laminar boundary layer resistance. Moreover, Jacobs *et al.* (2007) deal with air-water exchange of small water bodies in the field. It can be expected that exchange in a greenhouse proceeds at a slower rate because the wind speed is much slower. Stangellini (1987) studied heat exchange between plants in a greenhouse and found that this was dominated by forced convection, which has the consequence that the laminar boundary layer is almost non-existent (personal communication C. Stanghellini, 2018). She multiplied the LAI by a factor of 2 because the leaves have two sides. Based on the analogy between heat and substance transport, from her data an equivalent resistance of 200 s/m can be derived.

Inspection of the code showed that r_a is not fixed in the model but instead is calculated as:

$$r_a = \frac{d_{lam}}{D_{a,ref} \left(\frac{T}{T_{ref}} \right)^{1.75}} \qquad \text{Eqn 16}$$

where d_{lam} is the equivalent thickness of the laminar air boundary layer (m), set at 0.5 mm, $D_{a,ref}$ is the diffusion coefficient of the pesticide in the air (m^2 d^{-1}) at the reference temperature T_{ref} (K) and T is

[3] The definition of the LAI in greenhouses is a point of debate; we define it as the sum of the one-sided surface areas of the leaves divided by the surface area of the greenhouse. The total surface area available for deposition is the sum of the two-sided surface areas of the leaves.

the air temperature (K). This relationship between the diffusion coefficient and air temperature is identical to the one used in PEARL (Van den Berg *et al.*, 2016). However, in view of the fact that the laminar boundary layer is almost non-existent, it seems inappropriate to use an r_a that depends on the diffusion coefficient of the pesticide in the air. So we recommend to use r_a = 200 s/m = 2.32 × 10^{-3} d/m for all pesticide molecules. This has the consequence that r_a does not depend anymore from the temperature.

If the flux is from the plants to the air, it is described by:

$$J_{v,p} = \frac{A_p}{A_{ref}} \frac{c_{a,p} - c_{a,g}}{r_a} \qquad \text{Eqn 17}$$

where $J_{v,p}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the plants to the air ('*v*' from volatilisation), A_{ref} is a reference areic mass of 1 kg/ha so 10^{-4} kg m^{-2}.

The conceptual basis of the equation for $J_{d,p}$ is understandable because the surface area of the plants is proportional to the LAI. However, it is strange that the LAI is not included in the equation for $J_{v,p}$ and we recommend to include the LAI in this equation in future versions of GEM.

As described before, Stanghellini (1987) multiplied the LAI with a factor of 2 in her description of the heat flux to account for the fact that the leaves have two sides. This seems not appropriate in the equation for the volatilisation flux in case of LVM applications because then only the top of the leaves will be covered with pesticide by the spray application. So we recommend not to multiply with this factor in the equation for the volatilisation flux for LVM applications. For spray applications, we recommend to multiply with this factor of 2 because then both the top and the underside of the leaves are treated.

The A_p/A_{ref} term in the equation for $J_{v,p}$ goes back to Leistra & Wolters (2004). They justified this as follows: "The pesticide is assumed to be deposited on the leaves in spots of variable thickness. The thinner the deposit at a certain place, the sooner that place will be depleted by volatilisation. The concept is that the volatilising surface decreases in proportion to the decrease in mass of pesticide in the deposit." Wipfler *et al.* (2015a) showed that dosages of spray applications in substrate cultures range from about 0.01 to about 10 kg/ha so the A_p/A_{ref} term has a huge impact on calculated volatilisation fluxes.

So we checked the basis of this A_p/A_{ref} term. Let us consider (as a thought experiment) a simplified system: (1) a pesticide is sprayed onto a crop in a greenhouse which results in an A_p of 1 kg/ha, (2) the LAI, c_p, c_a and r_a are constant in time and c_p exceeds c_a (so flux from plants to the air). Eqn 17 then simplifies to:

$$J_{v,p} = \lambda A_p \qquad \text{Eqn 18}$$

with $\lambda = (c_{a,p} - c_{a,g})/(A_{ref}\, r_a)$. If we ignore dissipation due to penetration and transformation, the rate equation for A_p then simplifies to:

$$\frac{dA_p}{dt} = -\lambda A_p \qquad \text{Eqn 19}$$

Thus, the consequence of this concept is that A_p decreases exponentially with time under these conditions. The concept is based on a variable thickness of the deposition spots but it is not clear on which type of distribution of thicknesses of the spots this exponential decline is based. Further analysis using realistic distributions of these thicknesses (Annex 2) showed that the approach of Eqn 17 is only an approximation but that there is no suitable alternative approach, so we recommend to keep the concept in GEM-B that $J_{v,p}$ is directly proportional to A_p. This further analysis in Annex 2 showed also that the approximately exponential decrease applies to the remaining fraction of the dose; this can be achieved by replacing A_{ref} in Eqn 17 by the dose, so A_p immediately after application ($A_{p,i}$).

We did not analyse further the situation of repeated applications in which still some residue is left from the previous application. However, this is unlikely to occur because then there would be no need for the repeated application.

So we recommend to use the following equation:

$$J_{v,p} = LAI \frac{A_p}{A_{p,i}} \frac{c_{a,p} - c_{a,g}}{r_a} \qquad \text{Eqn 20}$$

This equation will work also for the situation where there is no pesticide on the plant because it will generate then a zero flux.

Holterman *et al.* (2012) also modelled the flux between the air and the plant surface in a greenhouse. They assumed that volatilisation from the plant surface and deposition onto the plant surface take place simultaneously. The sum of these fluxes is described as

$$J_p = LAI \frac{D_a}{d_{lam}} \left(\varphi_p \, c_{a,sat} - c_{a,g} \right) \qquad \text{Eqn 21}$$

where D_a is the diffusion coefficient in the air ($m^2\ d^{-1}$) and φ_p is the fraction of the plant surface covered with substance. In this equation the term $\varphi_p\ c_{a,sat}$ represents the deposition flux and the term $c_{a,g}$ represents the volatilisation flux. They assumed a d_{lam} of 0.001 m for a multi-span greenhouse and a walk-in tunnel (Table 13 of Beulke *et al.*, 2011). The fraction φ_p is calculated as

$$\varphi_p = \frac{A_p}{A_{p,crit}} \qquad \text{Eqn 22}$$

where $A_{p,crit}$ is the mass of substance per surface area of plants ($kg\ m^{-2}$) needed to cover the whole surface. This is estimated as:

$$A_{p,crit} = d_{\min} \rho \qquad \text{Eqn 23}$$

where d_{min} is the minimal thickness of the substance layer at the plant surface and ρ is the density of the solid or liquid phase of the pure substance ($kg\ m^{-3}$). If A_p exceeds $A_{p,crit}$ then φ_p is set to 1.
This minimal thickness of the substance layer was set to the thickness of 10 layers of molecules. The thickness of a single layer of molecules was calculated assuming that each molecule is a sphere. The volume of one molecule was estimated from the density of the pure substance and the molar mass. This results in d_{min} values of typically 10 nm. In combination with a typical ρ value of 1 kg/L and a LAI of 5, this gives a typical $A_{p,crit}$ value of 50 mg m^{-2}, so 0.5 kg ha^{-1}. This seems a reasonable value if the applied dose is about 1 kg/ha (50% surface coverage). However, if 50 g/ha is sprayed, this gives immediately after application already a fraction φ_p of 0.1 whereas the coverage of the plants with spray liquid will be about the same for doses of 50 g/ha and 1 kg/ha. Perhaps it is an idea to base the initial value of φ_p on characteristics of the application procedure (e.g. volume of water sprayed, type of nozzle). Another discussion point in this approach seems the need to include the LAI in the calculation procedure for φ_p. We conclude that the approach by Holterman *et al.* (2012) does not provide a straightforward alternative to describe the deposition flux.

3.3.2 The flux between the air and the floor surface

Van der Linden *et al.* (2015) used also rate equations for the exchange between the air and the floor. They used different approaches depending on the direction of the flux (as they did for the plants). Their equation for the mass flux of pesticide from the air to the floor (Eqn (9) in Appendix D) is not correct because of copy and paste of their Eqn (4). Inspection of the source code showed that the following equation is used:

$$J_{d,f} = \frac{A_f}{A_{ref}} \frac{c_{a,f} - c_{a,g}}{r_a} \qquad \text{Eqn 24}$$

where $J_{d,f}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the air to the floor ('*d*' from deposition), A_{ref} is set to the arbitrary value of 1 kg/ha and where $c_{a,f}$ equals the saturated vapour pressure as described in Section 3.1. The proportionality to A_f seems not defensible: why would the deposition rate increase with increasing mass of pesticide on the floor ?

The mass flux from the floor to the air is calculated with:

$$J_{v,f} = \frac{A_f}{A_{ref}} \frac{c_{a,f} - c_{a,g}}{r_a} \qquad \text{Eqn 25}$$

where $J_{v,f}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the floor to the air ('*v*' from volatilisation). Here there is some rationale for the proportionality to A_f: the more there is on the floor, the higher the flux. However, again the use of an arbitrary A_{ref} of 1 kg/ha seems difficult to justify and we recommend to replace A_{ref} by the A_f value immediately after application.

The above flux equations assume that the pesticide deposits are uniformly distributed over the floor surface. This is a point of discussion in view of the fact that only some 10-20% of the dose is assumed to be deposited onto the floor; probably the initial deposition in the paths between the plants is much higher than below the plant rows. A possible solution would be to assume that only the paths contain pesticide deposits and that the remainder of the floor is free of pesticide.

3.4 The flux from the condensation water to the circulating water

The mass flux of pesticide (i.e. mass rate per surface area of greenhouse) from the pool of condensation water to the circulating water (J_{circ}, kg m^{-2} d^{-1}) was not described by van der Linden *et al.* (2015). Therefore it is given here:

$$J_{circ} = f_{red} \; q_{cds} \; c_{w,cds} \qquad \text{Eqn 26}$$

where f_{red} is a reduction factor (-), q_{cds} is the volume flux of condensation water (volume rate per surface area of greenhouse, m/d) flowing to the circulating water, and $c_{w,cds}$ is the mass concentration in the condensation water (kg m^{-3}). The factor f_{red} is set to 0.1 in GEM-B; this was done because without this factor the simulated concentrations in the condensation water seemed too high based on expert judgement. However, as discussed before procedure for calculating the initial mass of pesticide in the condensation water will be revised. Therefore we recommend to omit this factor from the model and to test the revised model against measurements in the condensation water.

Crum *et al.* (1991) measured deposition on the roof and concentrations in condensation water after a spray application of methomyl. They found 0.05% deposition on the roof and the concentrations in the condensation water corresponded with 0.021% of the dose, so considerably less in the condensation water than deposited on the roof (see Section 6.4 for the cause of this difference). The maximum concentration in the condensation water was about 5 mg/L, i.e. 0.01% of its water

solubility of 55,000 mg/L). For an LVM application they found 1.5% deposition on the roof and the concentrations in the condensation water corresponded with 0.10% of the dose, so much less in the condensation water than deposited on the roof. The maximum concentration in the condensation water was again about 5 mg/L. Bor *et al.* (1994) measured deposition on the roof and concentrations in condensation water after a spray application of parathion-ethyl and found 0.4% deposition on the roof and 2.1% in the condensation water, so considerably more in the condensation water than deposited on the roof. The maximum daily concentration was 0.4 mg/L, i.e. 3% of the water solubility of parathion-ethyl. After an LVM application they found 1.2% deposition on the roof and 2.0% in the condensation water, so again more in the condensation water than deposited on the roof. For this LVM application the maximum daily concentration was 1.2 mg/L, i.e. 10% of the water solubility of parathion-ethyl. Parathion-ethyl and methomyl have comparable saturated vapour pressures (0.9 and 0.7 mPa) but their water solubilities differ strongly (12 versus 55,000 mg/L) and also their air-water partitioning coefficients differ strongly (about 10^{-5} versus about 10^{-9}), so these are interesting measurements for testing the model.

Van Os *et al.* (2012) sprayed a highly water soluble and non-volatile tracer in a tomato crop growing in a 6-m high greenhouse and found in a first experiment 0.05% of the dose in the condensation water after 24 h and 0.06% after 48 h (so in total about 0.1% of the dose); in a second experiment they found much lower concentrations (in total 0.003% of the dose).

At first glance one might think that the above approach for calculating the flux of pesticide (Eqn 26) would lead to an increasing concentration in the condensation water because only 10% of the pesticide molecules present in the volume flux of condensation water are transferred to the circulating water (so 90% of the molecules present in this volume flux stay in the condensation water on the roof, thus leading to an increase of the concentration). However, this is not the case because the total volume of condensation water is kept constant in the model (at 0.532 m^3/ha).

Van der Linden *et al.* (2015) do not explain why it is justifiable to assume equilibrium between the condensation water and the greenhouse air while they use rate approaches for exchange (1) between the greenhouse air and the plants and (2) between the greenhouse air and the floor. The surface area of the plants is usually larger than that of the greenhouse roof so one would expect that equilibration between plants and air proceeds faster than equilibration between roof and air.

Figure 8 indicates that the volume flux of condensation water flowing to the recirculation water is typically about 7 m^3 ha^{-1} d^{-1} from October to April and about 0.8 m^3 ha^{-1} d^{-1} from May to September (based on the line for 1992). As the volume of condensation water is 0.532 m^3 per ha, this means that even in summer the condensation water is usually completely 'refreshed' within less than a day. So most of the pesticide present in the condensation water will flow to the recirculating water in less than a day.

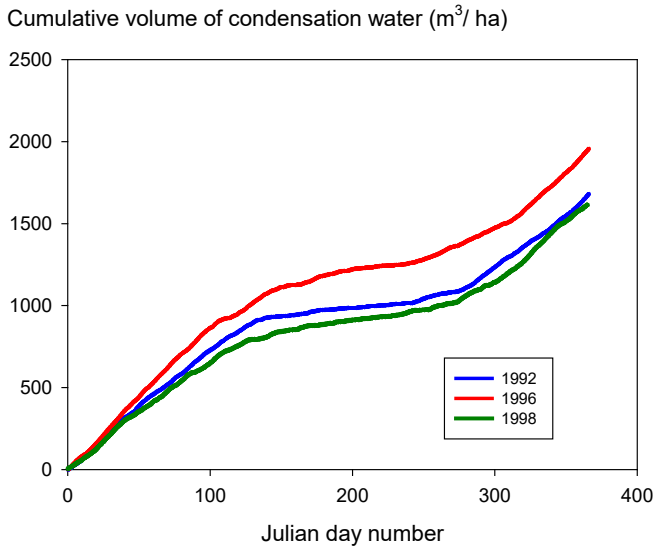

Figure 8 *Cumulative volume of condensation water per surface area of greenhouse flown into the recirculating water as a function of Julian day number as simulated by GEM for cut flowers (i.e. reference emission crop roses) for three different years.*

3.5 Dissipation kinetics from the air-water compartment due to ventilation and flow of condensation water

For the interpretation of model output it is useful to estimate the dissipation kinetics from the air-water compartment. Let us try to approximate these kinetics by considering only the loss processes from this compartiment, realising that this will give the fastest possible dissipation because volatilisation from plant and floor surfaces will increase the mass in this compartment. We consider only the time period after a pesticide application, so assuming that deposition fluxes can be ignored. We ignore furthermore here the degradation in air which is dealt with in the next section. There are then two loss processes left: pesticide leaving the system by the condensation water and pesticide leaving by ventilation. So the rate of decline of A_{a+w} can be written as:

$$\frac{dA_{a+w}}{dt} = -J_{vent} - J_{circ} = -H_a N_{vent} c_{a,g} - f_{red} q_{cds} c_{w,cds} \qquad \text{Eqn 27}$$

in which J_{vent} is the ventilation flux (kg m^{-2} d^{-1}), H_a is the volume of air per surface area of greenhouse (so 6 m) and N_{vent} is the ventilation rate coefficient (d^{-1}) which is 50 d^{-1} in GEM. The air-water partitioning coefficient (also called Henry coefficient) is defined as:

$$K_H \equiv \frac{c_a}{c_w} \qquad \text{Eqn 28}$$

The solution of this system is:

$$c_{a,g} = c_{a,g,i} \exp\left(-\frac{t}{\tau_{vc}}\right) \qquad \text{Eqn 29}$$

in which $c_{a,g,i}$ is the $c_{a,g}$ at the start and τ_{vc} is the time constant which is given by:

$$\tau_{vc} = \frac{H_a K_H + H_w}{H_a N_{vent} K_H + f_{red} q_{cds}} \qquad \text{Eqn 30}$$

in which H_w is the volume of condensation water per surface area of greenhouse (so 0.0000532 m).

The dependency of τ_{vc} from K_H was calculated for a q_{cds} for spring/summer conditions because then most pesticide applications take place, so q_{cds} = 0.8 × 10^{-4} m/d. Figure 9 shows that for K_H lower than about 10^{-9}, τ_{vc} approaches $H_{w,cds}$ / (f_{red} q_{cds}) which equals 6.65. Under these conditions the dissipation kinetics are controlled by the flow of the condensation water. For K_H larger than 10^{-4}, τ_{vc} becomes very close to 1/N_{vent} so 0.02 d. For K_H values between 10^{-8} and 10^{-4} the time constant has an intermediate value.

It is also relevant for interpretation of model results to know the fraction lost by ventilation defined by:

$$\Phi = \frac{J_{vent}}{J_{vent} + J_{circ}} = \frac{H_a N_{vent} K_H}{H_a N_{vent} K_H + f_{red} q_{cds}} \qquad \text{Eqn 31}$$

The relationship between Φ and K_H was calculated using the same parameters as described above. **Figure 10** shows that the ventilation is the dominant process if K_H exceeds 10^{-7}. The K_H of the vast majority of the pesticides exceeds this value so usually much more pesticide is lost by ventilation than by flow of condensation water in spring and summer when most pesticides are applied.

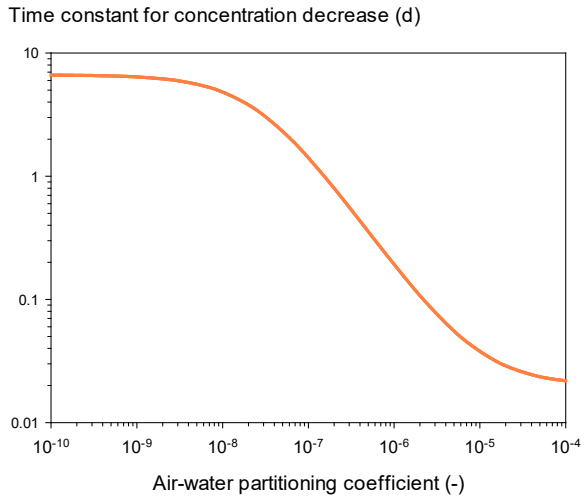

Figure 9 *The time constant τ_{vc} as a function of the air-water partitioning coefficient as calculated with Eqn 30.*

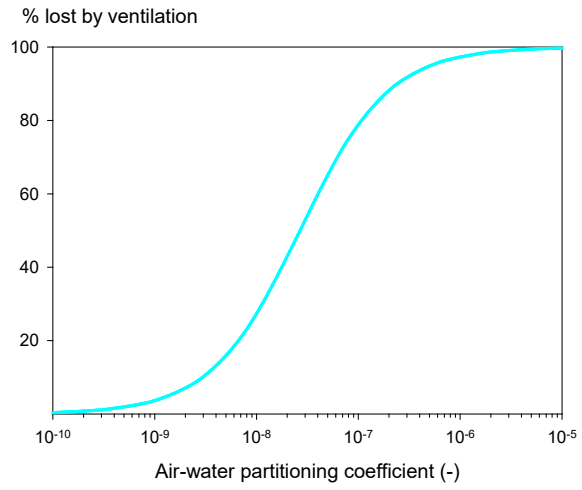

Figure 10 *The percentage lost by ventilation as defined by Eqn 31 as a function of the air-water partitioning coefficient.*

3.6 Need for including degradation in the air

GEM-B includes a first-order rate for degradation in the greenhouse air. Wipfler *et al.* (2015a) made calculations for some 24 pesticides that are commonly used in greenhouses. The half-lives for degradation in air ranged from 0.1 to 100 d and were usually in the range from 0.1 to 1 d. The source of these half-lives were pesticide dossiers at EU level. These half-lives were probably derived for outdoor air. The roofs of the greenhouse will absorb the UV radiation, i.e. the radiation with the highest energy, so the true degradation rates in the greenhouse are probably considerably slower than the rates estimated by GEM-B when using this dossier information.

Let us consider the possible effect of degradation in air by including this degradation in the simplified model described in the previous section (so Eqn 27). We then have to add a degradation term of minus $H_a\, k_{air}\, c_{a,g}$ to the right hand side of Eqn 27 where k_{air} is the first-order degradation rate coefficient in air (d^{-1}). As the ventilation flux equals $H_a\, N_{vent}\, c_{a,g}$, we can directly compare k_{air} and N_{vent} to assess the significance of degradation in air. GEM-B uses N_{vent} = 50 d^{-1} and the shortest half-life in air (i.e. 0.1 d) gives k_{air} = ln(2)/0.1 = 7 d^{-1}. So ventilation will usually be a much more important loss process. Because moreover the estimated values of k_{air} are likely to overestimate the true degradation rate in the greenhouse air, we recommend to omit degradation in air from GEM-B.

3.7 Initial speed of increase of the concentration in the air and its initial level

Let us now try to get a feeling how fast the initial increase of the concentration in the air will be as a result of the volatilisation processes. For simplicity we consider the following system: a greenhouse that is initially free of pesticide in which a pesticide is applied that ends up completely at the plant surface with ventilation as the only source of dissipation (so no condensation water). This leads to the following rate equation for the mass per surface area in the air, A_a (kg m^{-2}):

$$\frac{dA_a}{dt} = +J_{v,p} - J_{vent} = LAI\,\frac{c_{a,p} - c_{a,g}}{r_a} - H_a N_{vent}\, c_{a,g} \qquad \text{Eqn 32}$$

Note that the term $A_p/A_{p,i}$ was omitted from the volatilisation flux; this is defensible because we consider only the initial phase so when $A_p/A_{p,i}$ is close to 1.0. Some rearrangement, using $A_a = V_a\, c_{a,g}$ and realising that $c_{a,p} = c_{a,sat}$ gives:

$$\frac{dc_a}{dt} = \frac{LAI\, c_{a,sat}}{r_a\, H_a} - \left(\frac{LAI}{r_a\, H_a} + N_{vent}\right) c_a \qquad \text{Eqn 33}$$

The solution of this equation can be described as

$$c_{a,g} = c_{a,g,\infty}\left[1 - \exp\left(-\frac{t}{\tau_{vv}}\right)\right] \qquad \text{Eqn 34}$$

$$c_{a,g,\infty} \equiv \frac{LAI}{LAI + r_a\, H_a\, N_{vent}}\, c_{a,sat} \qquad \text{Eqn 35}$$

$$\tau_{vv} = \frac{r_a\, H_a}{LAI + r_a\, H_a\, N_{vent}} \qquad \text{Eqn 36}$$

where $c_{a,g,\infty}$ is the $c_{a,g}$ after infinite time and τ_{vv} is the time constant of this system (d). Using LAI = 5, r_a = 1.16 ×10^{-3} d/m, H_a = 6 m and N_{vent} = 50 d^{-1} gives $c_{a,g,\infty}$ = 0.88 $c_{a,sat}$ and τ_{vv} = 211 s. So within a few minutes a concentration in the air is reached that is very close to the concentration corresponding

with a saturated vapour pressure. If we set N_{vent} to zero (so no ventilation) we obtain $c_{a,g,\infty}$ = 1.00 $c_{a,sat}$ and τ_{vv} = 241 s; these numbers indicate that the volatilisation process dominates the ventilation process strongly, so the ventilation is hardly able to reduce the concentration in the air.

3.8 Concept of a saturated vapour concentration at the plant surface

The flux for the volatilisation from plant surfaces in GEM-B is so far based on the concept that above a deposit of pesticide at the plant surface the concentration in the air is equal to the saturated vapour concentration shortly after application. However, it is questionable whether this is the case because the pesticide molecules in such a deposit may behave differently from pesticide molecules in the solid or liquid state of the pesticide.

Baas & Huygen (1992) sprayed parathion-ethyl onto a tomato crop and found a $c_{a,g}$ = 0.2 $c_{a,sat}$ in the first half hour after application (their Table 4.14). They estimated N_{vent} at 4.6 d^{-1} under these conditions (their Table 12) and their H_a was about 3.5 m (their p. 20). The tomato crop was about 1.3 m high at the time of application (Bor *et al.*, 1994, p. 13), so we estimate LAI to be 2. Using an r_a of 2.32 $\times 10^{-3}$ d/m gives $c_{a,g,\infty}$ = 0.98 $c_{a,sat}$ for these conditions (and τ_{vv} = 344 s). The estimated LAI of 2 has hardly any effect on this 0.98: if we set LAI to 1.0 then we obtain $c_{a,g,\infty}$ = 0.96 $c_{a,sat}$.
So the measured concentration is only about 20% of the saturated concentration whereas it should be about equal to the saturated concentration. So either the r_a is too low or the concentration at the plant surface is only about 30% of the saturated concentration. As the r_a was based on independent measurements (via the analogy with heat exchange), it seems likely that the assumption that the concentration at the plant surface corresponds with saturated conditions, is wrong.

As follows from this example and from that at the end of Section 3.7, the time constant τ_{vv} is in the order of minutes and the concentration reached within these minutes is close to the saturated vapour concentration. We searched for some further literature data on concentrations in greenhouses shortly after spray applications to plants. We calculated the saturated vapour concentration with Eqn 14 based on pesticide properties from the PPDB pesticide properties database. **Table 3** shows that maximum concentrations for parathion-ethyl, pirimicarb, chlorothalonil and malathion are 5-26% of the saturated vapour concentrations. The 20% found by Baas & Huygen (1992) for parathion-ethyl (see previous paragraph) is nicely in the range of values found for this pesticide by Siebers & Mattusch (1996). The maximum concentration found for dinocap by Siebers & Mattusch is about equal to the saturated vapour concentration and that found for dichlofluanid is 3-5 times higher than the saturated vapour pressure. Siebers & Mattusch sampled air by pumping air through columns packed with sorbents so their concentrations should be considered as upper limits for the concentrations in the greenhouse air. The concentrations found for fenhexamid by Tsiropoulos *et al.* (2006) are about 100 times higher than the saturated vapour pressure. They sampled also the air by pumping air through columns packed with sorbents so probably the high concentrations are caused by spray droplets that were retained by the columns. So the data in **Table 3** confirm the observation by Baas & Huygen that the initial concentration in the air is far below the saturated vapour concentration.

Please note that there are is a vast amount of measurements available of concentrations in greenhouse air after spray applications (see e.g. references cited by Siebers & Mattusch 1996, Kazos *et al.* 2008 and Houbraken *et al.* 2017) so **Table 3** contains only a limited sample of available data. We recommend to collect and analyse all these data, hoping that this will give indications of the causes of the differences between the substances.

Leistra & Wolters (2004) fitted measurements on volatilisation of fenpropimorph in an outdoor wind tunnel to a model that was based on the following volatilisation flux:

$$J_{v,p} = \frac{A_p}{A_{ref}} D_a \frac{c_{a,sat}}{d_{lam}} \qquad \text{Eqn 37}$$

with A_{ref} = 1 kg/ha. D_a was a function of temperature as described in Section 3.3.1. They did so for four experiments in which fenpropimorph was sprayed onto beans, radish and sugar beet and used d_{lam} as a fitting parameter. They found d_{lam} values ranging from 0.5 to 1 mm under these outdoor conditions.

Let us assume that the concentration at the plant surface is not equal to the saturated vapour concentration but instead equal to a proportionality factor F times this concentration. In combination with our earlier proposals this leads to the following volatilisation flux:

$$J_{v,p} = \frac{A_p}{A_{p,i}} \frac{F\ c_{a,sat}}{r_a} \qquad \text{Eqn 38}$$

Combination of these two flux equations gives

$$F = \frac{A_{p,i}}{A_{ref}} \frac{D_a\, r_a}{d_{lam}} \qquad \text{Eqn 39}$$

Applying this equation to the data of Leistra & Wolters (2004) and assuming $r_a = 2.32 \times 10^{-3}$ d/m gave F values of 0.5, 0.6, 0.9 and 1.0. One may expect that the actual r_a in the windtunnel was lower than this value estimated for a greenhouse because the wind speed in an outdoor windtunnel is probably higher than in a greenhouse. Inserting a lower r_a in Eqn 39 gives a lower F, so this range of F from 0.5 to 1.0 is an overerestimation of the true range of F. So these results give also qualitative support for the hypothesis that the concentration at the plant surface is lower than the saturated vapour concentration.

Table 3 *Some measured concentrations in greenhouse air after spray applications collected from literature.*

pesticide	type of plant	measured concentration (µg m^{-3})		saturated vapour concentration (µg m^{-3})	reference
		maximum	average of first day		
parathion-ethyl	begonias	5	2.4	106	Siebers & Mattusch (1996)
	begonias	10	3.6	106	
	chrysanthemums	22	4.3	106	
	chrysanthemums	26	5.1	106	
	cucumbers	20	5.0	106	
	cucumbers	28	5.8	106	
dinocap	begonias	2.3	0.4	1.1	
	begonias	1.0	0.2	1.1	
	chrysanthemums	0.4	<0.2	1.1	
	chrysanthemums	0.8	<0.2	1.1	
	cucumbers	1.1	0.2	1.1	
	cucumbers	0.9	0.2	1.1	
pirimicarb	begonias	9	3.7	42	
	begonias	6	2.0	42	
	chrysanthemums	6	2.9	42	
	chrysanthemums	5	2.3	42	
	cucumbers	10	4.9	42	
	cucumbers	8	2.9	42	
dichlofluanid	tomatoes	14	5.0	5	
	tomatoes	25	4.5	5	
chlorothalonil	ornamental pl.	0.8		8	Kazos *et al.* (2008)
malathion	tomatoes	20		420	Tsiropoulos *et al.* (2006)
fenhexamid	tomatoes	6	1	0.05	

3.9 Dissipation at the plant surfaces

As described by van der Linden *et al.* (2015), both GEM-B and GEM-C include a first-order dissipation process for the pesticide molecules at the plant surfaces with a default half-life of 10 d.
This dissipation includes both penetration into the plants and degradation at the plant surface. Photodegradation is driven by UVB radiation (wavelength 280-320 nm) which does not reach the plant surfaces in greenhouse because of the glass. So the main dissipation process is expected to be penetration into the leaf.

Leistra (2005) reviewed the literature on pesticide penetration into the leaves. The half-life for penetration of many pesticide-plant combinations is in the range of a few hours to a few days, but also much longer half-lives are possible. Formulation adjuvants may decrease this half-life considerably (de Ruiter *et al.*, 2004). An example of fast penetration is the study by Leistra & Wolters (2004) who found rate coefficients for penetration of fenpropimorph into leaves of beans, radish and sugar beets ranging between 2 and 5 d^{-1} (corresponding with half-lives between 0.1 and 0.4 d).

We recommend to include the rate coefficient for dissipation in a sensitivity analysis of the GEM-B model. If this analysis shows that this coefficient has a large impact, notifiers may consider to determine this rate coefficient for their crop-pesticide combination as a higher tier.

3.10 Need to include direct contamination of circulation water

As described in Section 3.1, GEM-B does not include direct contamination of the circulation water by spray or LVM applications. Van Os *et al.* (2012) measured the distribution of highly water soluble tracers in a greenhouse after spray applications to a full grown tomato crop on stonewool. They used three different application procedures: (1) all nozzles of the vertical spray bar open, (2) lowest nozzle of vertical spray bar closed, (3) all nozzles open but stonewool slabs covered to prevent direct spraying and dripping onto these slabs. The applied water volumes were between 1300 and 1800 L/ha. They performed the experiment twice in the same greenhouse (on 27 October and 16 November 2009) using different tracers. In the first experiment, van Os *et al.* (2012) recovered 1% of the dose in the water in the mats for procedure (1) and 0.4-0.6% for procedures (2) and (3). In the second experiment they found values that were about ten times higher: 9% for procedure (1) and 4-6% for procedures (2) and (3). Recovery of 9% of the dose in the mats seems unrealistically high. In the experiment described by van der Maas *et al.* (2015) the surface area of the mats was 14% of the total surface area and the mats are covered with foil (except for the 10 × 10 cm surface of the pots in which the plants were introduced into the system). So it is difficult to envisage how 9% of the dose could end up in the mats. So we propose to assume as a default value a direct contamination of the mats of 1% of the dose after spray applications.

Van Os *et al.* (2012) measured also the deposition into the troughs that transport the drainage water to the tanks in the system. In the first experiment they found 0.2-0.3% of the dose in these troughs and in the second experiment 0.1-4.1%. We propose to ignore again the second experiment and to assume that 0.3% of the dose ends up in the troughs after spray applications.

Van Os *et al.* (2012) found for procedure (1) that 41-45% of the dose was recovered from horizontal filter collectors hanging above the mats. So the surface area of the foil that covers the mats will contain considerable amounts of pesticide which may drip partly into the troughs (e.g. also during a next spraying event).

The mats are not completely covered with foil: each plant grows in a pot with a 10×10 cm surface area which is uncovered. We calculated the total surface area of the pots for the experiment described by van der Maas *et al.* (2015); there were 276 pots so 2.76 m^2 in a greenhouse of 144 m^2, so the pots were about 2% of the total surface area. In combination with the 41-45% of the previous paragraph, one

would expect that about 1% of the total sprayed mass would end up in the pots. So this is more or less consistent with the 1% of the dose recovered in the mats by van Os *et al.* (2012) for procedure (1).

Duivestijn & Marang (2014, p. 23) point at the possibility of contamination of drainage water resulting from the cleaning of the troughs (using water under high pressure water) after the plants and the mats have been removed. This aspect was not considered in the measurements by Van Os *et al.* (2012) as they measured in the drainage water which flows only over part of the trough surface. There will usually be at least one month time between the last spray application and the removal of the plants. During this period pesticide present at the dry trough surface may volatilise depending on the saturated vapour pressure. We recommend to ignore this aspect for the time being in GEM-B but we recommend to measure for a number of pesticide-crop combinations the concentration in this cleaning water.

Given the importance of direct contamination and the wide range found in the duplicate experiments by van Os *et al.* (2012) we recommend to collect more experimental data on this contamination route. This should include both the mats and the troughs and also the cleaning of the troughs at the end of the growing cycle.

As there seem to be no measurements of contamination of the mats and the troughs after LVM applications, we propose to use the same default values as for spray applications (1% contamination of the mats and 0.3% of the troughs).

4 Review of processes influencing the concentration in the condensation water and in the water on the tables in GEM-C

4.1 Overview of GEM-C

The GEM-C model (described in Appendix E of van der Linden *et al.*, 2015) deals with emissions to surface water following applications to crops grown in pots in an ebb/flow system on tables or floors (Figure 12). The application technique may either be spraying, fogging, fumigation or application via the irrigation water.

Currently, application by fogging hardly occurs anymore in greenhouses in the Netherlands and it is not 100% clear which types of application are included in fumigation. Almost all pesticide applications in the context of GEM-C are either by spraying (see Figure 11) or by room treatments with the low-volume mister (LVM). Therefore we recommend to limit the application methods in GEM-B to spraying or LVM. In case of spraying of crops grown in pots the spraying is directed to the top of the plants only (so not to the underside of the leaves as in the case of GEM-B). The spray volume is typically 1000 L/ha. The procedure for LVM applications is identical to that in GEM-B (see Section 3.1).

Figure 11 *Spray application to Calathea growing in pots on tables (photograph by WUR Greenhouse Horticulture).*

The GEM-C model distinguishes the following compartments in the greenhouse containing pesticide:

- the air
- the condensation water on the roof
- the plant surfaces
- the top 2 mm of the substrate in the pots (which has 10% organic matter)
- the bottom 10 cm of the substrate in the pots (which has 10% organic matter)
- the recirculation water on the tables
- the recirculation water in a number of tanks.

The schematic representation of the fluxes and the partitioning in Figure 13 indicates that the concentration in the water on the tables is assumed to be perfectly mixed with the water in the bottom 10 cm of the pots (including sorption equilibrium in these bottom 10 cm of the pots).

The pesticide on the greenhouse floor is not included in GEM-C because the tables with the pots cover the surface area of the greenhouse to such a large extent that the deposition on the floor can be ignored.

Figure 12 *Photographs of crops growing on ebb/flood systems (photographs by WUR Greenhouse Horticulture).*

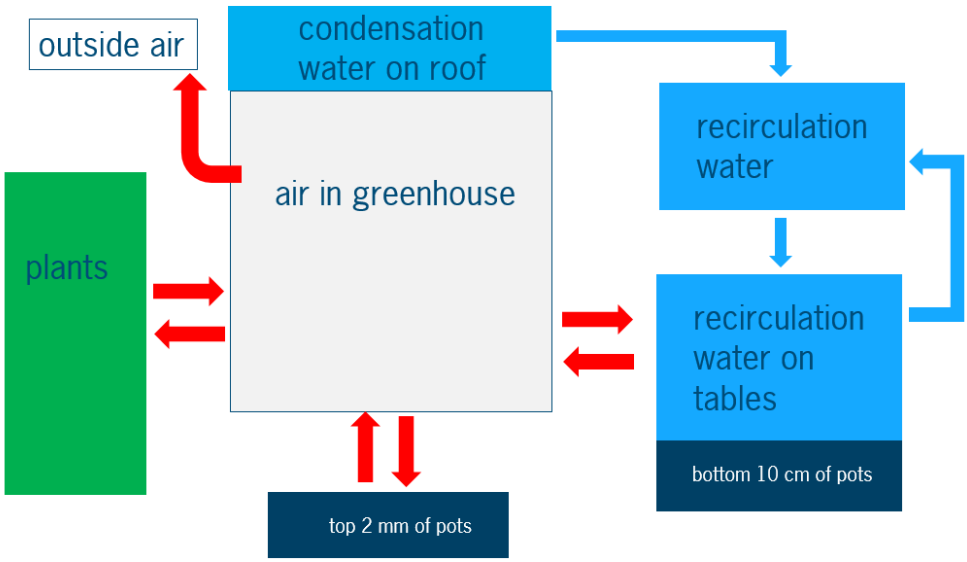

Figure 13 *Schematic representation of the exchange between the different compartments in the greenhouse in GEM-C. The red arrows are gas fluxes and the blue arrows are water fluxes. Only those compartments are shown that contribute to the concentration in the condensation water.*

4.2 Initial distribution of dose

As described by van der Linden *et al.* (2015), a pesticide application is distributed over the different compartments as follows:

$$A_{p,i} = f_p \; A_i \qquad \text{Eqn 40}$$

$$A_{a,i} = f_a \; A_i \qquad \text{Eqn 41}$$

$$A_{pot,i} = \left(1 - f_p - f_a\right) f_{pot} \; A_i \qquad \text{Eqn 42}$$

$$A_{tab,i} = \left(1 - f_p - f_a\right)\left(1 - f_{pot}\right) A_i \qquad \text{Eqn 43}$$

where A_i is the areic mass (i.e. mass per surface area of greenhouse) applied (kg m^{-2}), $A_{p,i}$ is the areic mass applied on the plants, $A_{a,i}$ is the areic mass staying in the greenhouse air (kg m^{-2}), $A_{pot,i}$ (kg m^{-2}) is the areic mass applied to the substrate in the pots, $A_{tab,i}$ (kg m^{-2}) is the areic mass that is deposited onto the recirculation water on the tables on which the pots are placed, f_p is the fraction applied to the plants, f_a is the fraction that stays in the air (-), and f_{pot} is the fraction of the surface area covered with pots (-).

Both f_p and f_a are fixed in the scenario. Van der Linden *et al.* (2015) did not provide the values used; inspection of the code showed that for spray applications f_a was 0.08 (so again 8%) and f_p was 0.80 (so same values as for cut flowers shown in Table 1). So then $A_{pot,i} + A_{tab,i} = 0.12\ A_i$ and this amount is subdivided between the pots and the tables using the above equations.

The value of f_{pot} was for all crops fixed to 0.3. So $A_{tab,i}$ equals then $0.7 \times 0.12\ A_i$ which corresponds with 8.4% of the dose. It seems quite likely that the contribution of this exposure route to the contamination of the recirculation water will exceed the other routes in this current version of GEM-C. So the parameter f_{pot} is likely to have a very large effect. The value of 0.3 seems realistic in view of the data in Annex 4. Frequency distributions of f_{pot} for crops grown on tables and floors (Figure 14) show that all f_{pot} values for crops grown on floors are below 0.5 whereas the values for crops grown on tables are sometimes higher (as is also illustrated by the middle photograph of Figure 12). We understood from Ctgb (personal communication Anne Steenbergh, 2018) that pesticide registrations are either provided for individual crops or for groups of crops growing on tables or floors. In view of the probably large effect of f_{pot} (to be confirmed by sensitivity analysis) we suggest to consider the possibility to use a crop-specific f_{pot}.

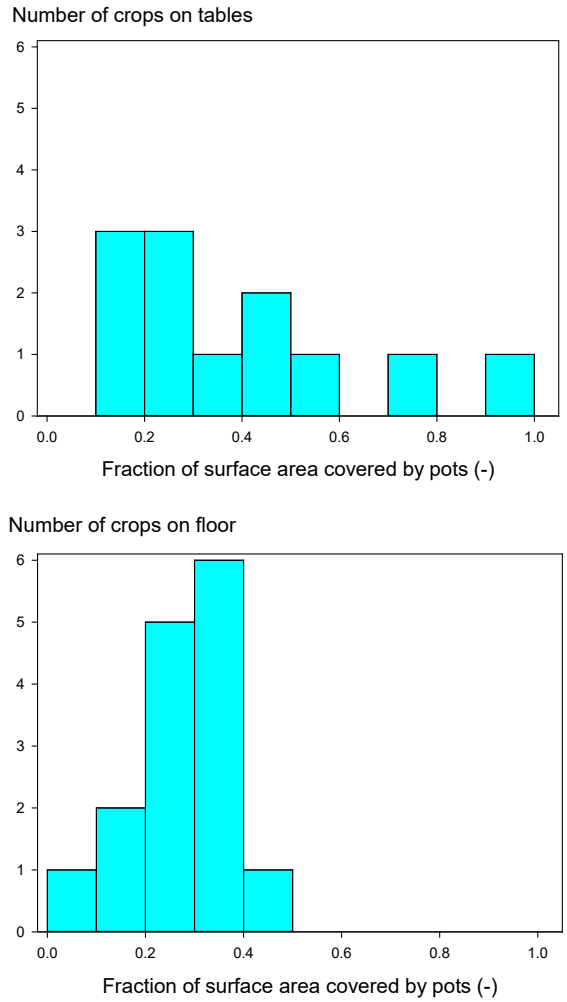

Figure 14 *Frequency distributions of the fraction of the surface area covered by pots (f_{pot}) for crops grown on tables (top) and crops grown on floor (bottom); data taken from Annex 4.*

For applications by the low volume mister f_a was 0.35 and f_p was 0.55 (so same values as in GEM-B; see Table 2), so then $A_{tab,i}$ equals then 0.7 × 0.10 A_i which corresponds with 7% of the dose. Thus in the current GEM-C version 35% goes into the air immediately after application so this route may in this current version be more important than the deposition onto the recirculation tables.

The basis of the initial fractions in the air (8 and 35%) is the same as for GEM-B, so we propose to modify this approach also here.

4.3 The fluxes in the gas phase

4.4 The flux between the air and the plant surfaces

Van der Linden *et al.* (2015) use in GEM-C rate equations for the exchange between the air and the plants similar to those of GEM-B. If the flux is from the air to the plants, it is described by

$$J_{d,p} = LAI\ D_a \frac{C_{a,g} - C_{a,p}}{d_{lam}} \qquad \text{Eqn 44}$$

where D_a is the diffusion coefficient of the pesticide in the air ($m^2\ d^{-1}$) and d_{lam} is the equivalent thickness of the laminar air boundary layer (m), set at 0.5 mm. This equation is equal to the corresponding equation in GEM-B (Eqn 15) considering that $r_a = d_{lam} / D_a$. Leistra (2005) reports

D_a values of three pesticides to range from 0.36 to 0.50 m^2 d^{-1}. This gives a range for r_a of 1.0×10^{-3} to 1.4×10^{-3} d m^{-1}, which is close to the r_a value of 1.16×10^{-3} d m^{-1} that was used in GEM-B.

If the flux is from the plants to the air, it is described in GEM-C by:

$$J_{v,p} = LAI \frac{A_p}{A_{ref}} D_a \frac{c_{a,p} - c_{a,g}}{d_{lam}} \quad \text{Eqn 45}$$

where A_{ref} is again set at 10^{-4} kg m^{-2}, so 1 kg/ha. Inclusion of the LAI in this flux equation seems logical because a larger surface area will lead to a larger flux. However, the LAI was not included in the $J_{v,p}$ flux in GEM-B. We recommend to include the LAI in the equation for $J_{v,p}$ in both GEM-B and GEM-C.[4]

4.4.1 The flux between the air and the surface of the pots

The top 2 mm of the substrate is assumed to be perfectly mixed and the gas fluxes for the exchange between the top 2 mm of the pots and the air are described by:

$$J_{d,pot} = f_{pot}\ D_a \frac{c_{a,g} - c_{a,pot}}{d_{lam}} \quad \text{Eqn 46}$$

$$J_{v,pot} = f_{pot}\ D_a \frac{c_{a,pot} - c_{a,g}}{d_{lam}} \quad \text{Eqn 47}$$

where $J_{d,pot}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the air to the pots, $J_{v,pot}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the pots to the air, and $c_{a,pot}$ is the concentration in the gas phase in the top 2 mm of the pots (kg m^{-3}). This concentration is derived from the conservation equation for the mass in this top 2 mm of substrate:

$$\frac{dA_{pot}}{dt} = +J_{d,pot} - J_{v,pot} - J_{dif,pot} - k_{pot}\ A_{pot} \quad \text{Eqn 48}$$

where A_{pot} is the mass per surface area in this top 2 mm (kg m^{-2}), $J_{dif,pot}$ is the downward diffusion flux (kg m^{-2} d^{-1}) in the gas phase from the top of this top 2 mm into the soil, and k_{pot} is the rate coefficient (d^{-1}) for degradation in this top 2 mm. This diffusion flux $J_{dif,pot}$ is calculated as:

$$J_{dif,pot} = f_{pot}\ D_{a,pot} \frac{c_{g,pot}}{d_{pot}} \quad \text{Eqn 49}$$

with

$$D_{a,pot} = \varepsilon^{1/3}\ D_a \quad \text{Eqn 50}$$

where $D_{a,pot}$ is the diffusion coefficient in the gas phase in this top 2 mm (m^2 d^{-1}), d_{pot} is the thickness of the layer (2 mm) and ε is the volume fraction of the gas phase (-)[5]. The concept behind this equation for $J_{dif,pot}$ is the assumption that the concentration below this top 2 mm is zero.

[4] Inspection of the code showed that the same equation for $J_{v,p}$ was used in GEM-B and in GEM-C and that the LAI was not included in this equation.

[5] Eqn 13 in Appendix E of van der Linden *et al.* (2015) states that the diffusion coefficient of the pesticide in the gas phase of the pot substrate is proportional to the volume fraction of liquid whereas it should be proportional to the volume fraction of gas. Inspection of the source code showed that this was a typo: the code assumes indeed that it is proportional to the volume fraction of gas.

The calculation of $c_{a,pot}$ is based on the assumptions that the substance is perfectly mixed and that there is equilibrium partitioning between the sorbed, liquid and gas phases in these top 2 mm assuming a linear sorption isotherm:

$$c_{g,pot} = \frac{A_{pot} / d_{pot}}{\varepsilon + \frac{\theta}{K_H} + \frac{\rho_{pot}\, m_{om}\, K_{om}}{K_H}} \qquad \text{Eqn 51}$$

where θ is the volume fraction of liquid in this top 2 mm, ρ_{pot} is the dry bulk density of this top 2 mm (kg m^{-3}), m_{om} is the mass fraction of organic matter in this top 2 mm (kg/kg) and K_{om} is the organic-matter/water distribution coefficient of the substance (m^3 kg^{-1}). The parameter values were as follows:

ε = 0.54, θ = 0.06, ρ_{pot} = 1 kg dm^{-3}, m_{om} = 0.10 kg/kg. The value for θ is remarkably low for a pot of soil with 10% organic matter whose bottom 5-cm layer is regularly saturated with water.

The above combination of assumptions with respect to the fluxes seem inconsistent: (1) the equations for $J_{d,pot}$ and $J_{v,pot}$ are based on the assumption that the concentration at the surface of these 2 mm equals $c_{a,pot}$, (2) the equation for $J_{dif,pot}$ assumes a linear decrease of the concentration from $c_{a,pot}$ at the soil surface to zero at 2 mm depth, (3) the equation for $c_{a,pot}$ assumes that $c_{a,pot}$ is constant over these top 2 mm. So (1) and (2) are consistent with each other but (3) is inconsistent with (1) and (2).

The equation for $J_{dif,pot}$ is based on only diffusion in the gas phase and thus ignores diffusion in the liquid phase. Diffusion coefficients of pesticides in air and water are typically 0.4 and 0.4 × 10^{-4} m^2 d^{-1}, respectively; so diffusion in the air proceeds 10^4 faster than in water. However, for many pesticides the concentration in air is more than a factor 10^4 lower than in water (i.e. $K_H < 10^{-4}$), in which case the diffusion flux in water will dominate the diffusion flux in air. Therefore we recommend to include diffusion in the liquid phase in the GEM-C model.

The thickness of 2 mm seems an arbitrary choice that requires further underpinning if the processes in this layer have a significant influence on the emission concentrations. We recommend to assess this significance by a sensitivity analysis. The thickness of 2 mm may be checked by simulations with PEARL for a few realistic time courses of the concentration in the air in the greenhouse.

The above equations for $J_{d,pot,}$ $J_{v,pot}$ and $J_{dif,pot}$ contain the factor f_{pot}; this has the consequence that these fluxes are mass rates per surface area of greenhouse (so not per surface area of pots). However, the equation for $c_{a,pot}$ does not contain f_{pot} so in this equation A_{pot} is assumed to be the mass per surface area of pots, which is incorrect because via the equations described in Section 4.2 A_{pot} was defined as mass per surface area of greenhouse. Let us illustrate this with the following example. Based on the parameterisation described in Section 4.2, A_{pot} equals 0.3 × 0.12 A_a = 0.036 A_a. Let us assume that a dose of 1 kg/ha was sprayed; this gives A_{pot} = 3.6 mg m^{-2}. Using the approach in GEM-C, this leads to a concentration in total soil of 3.6 mg m^{-2} / 0.002 m = 1.8 mg dm^{-3}. However, the true concentration in total soil in the pots is in this case 1.8/0.3 = 6 mg dm^{-3} because A_{pot} is deposited on only 30% of the surface area. We recommend to improve this aspect.

4.4.2 The flux between the air and the water on the tables

The gas fluxes for the exchange between the water on the tables and the air are described by:

$$J_{d,tab} = \left(1 - f_{pot}\right) D_a \frac{c_{a,g} - c_{a,tab}}{d_{lam}} \quad \text{Eqn 52}$$

$$J_{v,tab} = \left(1 - f_{pot}\right) D_a \frac{c_{a,tab} - c_{a,g}}{d_{lam}} \quad \text{Eqn 53}$$

where $J_{d,tab}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the air to the water on the tables, $J_{v,tab}$ is the mass flux of pesticide (kg m^{-2} d^{-1}) from the water on the tables to the air, and $c_{a,tab}$ is the concentration in the gas phase at the interface between the water and the air (kg m^{-3}). This concentration is calculated from the concentration in the water on the tables using Henry's law. This concept seems defensible and thus does not need to be revised.

In the above equations different descriptions are used depending on the direction of the flux (deposition or volatilisation). It is somewhat a matter of taste but we consider no need to use different descriptions because the direction of the flux is defined by its sign.

As discussed in Section 3.3.1 we recommend to replace the quotient d_{lam}/D_a by r_a.

4.5 The interaction between the recirculation water on the tables and the pots

The concentration in the water on the tables is assumed to be perfectly mixed with the water in the bottom 10 cm of the pots, and it is assumed that there is sorption equilibrium in these bottom 10 cm of the pots. Let us consider what happens on the ebb/flow tables: at the start of an irrigation event, a water layer of about 5 cm is formed on the tables and this water gradually drains away over a period of 15-20 min. Initially part of the water will flow into the pots because of the water layer of 5 cm and additionally by capillary suction. When the water layer has drained away, some of the water in the bottom centimetres of the pots will flow out (with lower concentrations because of the sorption), but this is only a small part of the recirculating water that is on the tables. The water that remained on the tables during the irrigation event does not mix with the water in the pots and the concentration in this water will not be lowered by the sorption in the pots. So the assumption of perfect mixing between the bottom 10 cm of the pots and the recirculation water on the tables seems to be too optimistic and this may lead to a considerable underestimation of emission concentrations for pesticides with high K_{om} values. We recommend therefore to revise this concept. A straightforward simple alternative is to assume that a certain fraction of the volume of water pumped on the tables is taken up by the pots together with the pesticide present in this water. This means that the adsorption in the pots will not influence the concentration in the recirculation water on the tables.

4.6 The flux from the condensation water to the recirculation water

Eqn (26) in Appendix E of van der Linden *et al.* (2015) suggests that the concentration in the condensation water is multiplied by 0.1. However, inspection of the code showed that the code uses instead Eqn 26 of our report.

5 Proposed changes in GEM-A

Whereas the previous chapters dealt with a critical review of different aspects of GEM, this chapter and the next chapters describe the proposed changes in GEM.

Following the recommendations in Chapter 2, the following processes have to be added to GEM-A:
- partitioning into the roots
- sorption to the substrate material (stonewool, coir etc.)
- sorption to foil
- sorption to the different types of plastic tubes.

This leads to the following of the total mass of pesticide m_{sys} (mg) present in the part of the system that starts where the water leaves the mixing tank and that ends where the water leaves the substrate system via drainage:

$$m_{sys} = V_{sub}\, c_w + M_{sub} K_{sub}\, c_w + M_{foil} K_{foil}\, c_w + M_{roots}\, RCF\, c_w + \left(\sum_{i=1}^{3} L_i\, S_i + \sum_{i=1}^{3} L_i\, p_i\, K_{tube,i} \right) c_w \qquad \text{Eqn 54}$$

with

$$\sum_{i=1}^{3} L_i\, S_i = L_{PVC}\, S_{PVC} + L_{pei}\, S_{pei} + L_{pec}\, S_{pec} \qquad \text{Eqn 55}$$

$$\sum_{i=1}^{3} L_i\, p_i\, K_{tube,i} = L_{PVC}\, p_{PVC}\, K_{tube,PVC} + L_{pei}\, p_{pei}\, K_{tube,pei} + L_{pec}\, p_{pec}\, K_{tube,pec} \qquad \text{Eqn 56}$$

where L is the length of a class of pipes/tubes (dm), S is the inner surface area (dm^2) of a class of pipes/tubes, p is the mass per length (kg/dm) of a class of pipes/tubes, K_{tube} is the sorption coefficient (dm^3/kg) of a class of pipes/tubes and where the subscripts *PVC*, *pei* and *pec* indicate the PVC transport pipes, polyethene irrigation tubes and polyethene capillary tubes, respectively. To be able to make these new elements of the model operational, default input values are needed for the new parameters. These are provided below.

Boesten & Matser (2017) measured for Cultilene stonewool foil a surface area per mass of 281 cm^2 g^{-1}. The dimensions of such a stonewool slab are 7.5×12×100 cm. This gives a surface area of 7.5×12×2 + 100×12×2 + 100×7.5×2 cm^2, so about 4000 cm^2. This gives a mass of foil per slab of 14 g which can be used to estimate M_{foil} for any greenhouse system.

The rule of thumb is that a full-grown tomato or cucumber plant has a mass of 5 kg of which about 18% consists of roots (fresh weight). There are two such plants per stonewool slab which gives for full-grown plants an M_{roots} per slab of 2×0.18×5=1.8 kg. The volume of such a slab is about 80 L, so the volume fraction of roots is quite small. We recommend a linear increase of M_{roots} from 0 to this maximum value in the first six weeks after application and to keep M_{roots} constant for the remainder of the growing period (based on footnote 1 on p. 20).

We recommend to base the default *RCF* on the Briggs-et-al. relationship with the octanol-water partitioning coefficient described in Section 2.3 and to check as part of a sensitivity analysis how critical this estimate is.

We recommend to use as default values S_{PVC} = 0.062 dm^2, S_{pei} = 0.020 dm^2 and S_{pec} = 0.0013 dm^2 (see Section 2.2). We recommend to use as default values p_{PVC} = 0.24 kg m^{-1}, p_{pei} = 0.074 kg m^{-1} and p_{pec} = 0.015 kg m^{-1} (see Section 2.2).

6 Proposed changes in GEM-B

6.1 Application methods

As indicated before, we propose to limit the application methods in GEM-B to (1) spraying and (2) room treatments with the LVM.

6.2 Partitioning processes in the recirculating water and in the substrate

The same changes are proposed as for GEM-A (see previous chapter).

6.3 Initial distribution of the substance

There are two simple options for the concentrations in the air and the condensation water immediately after application: (1) these concentrations correspond with saturated vapour pressure and water solubility, (2) these concentrations are zero. Let us have a look at available measurements. Baas & Huygen (1992) sprayed parathion-ethyl onto a tomato crop and found a maximum concentration in greenhouse air of about 0.02 mg/m^3 (their Table 4.14) which is about 20% of the concentration under saturated conditions. As part of the same experiment Bor *et al.* (1994) measured a daily maximum concentration in the condensation water of 0.4 mg/L which is about 3% of the water solubility. The measurements of Baas & Huygen indicate a rapid decline of the concentration in the air: the maximum of 0.02 mg/m^3 was measured during the first half hour and was at about 50% of this level up to 4 h. So both the concentration in the air and in the condensation water were much lower than corresponding with saturated vapour pressure and water solubility. Crum *et al.* (1991) sprayed methomyl onto a tomato crop in a greenhouse and found a maximum concentration in the condensation water of 5 mg/L which is only 0.01% of the water solubility of methomyl. Thus, these measurements indicate that it is not realistic to assume that the greenhouse air and the condensation water contain concentrations corresponding with saturated conditions immediately after a spray application. This is not surprising because the substances are sprayed as a wettable powder or an emulsifiable concentrate. So the substance is in a droplet of water that evaporates in the air or at the leaf surface. After evaporation, the substance is present in a residue that consists for a large part of the formulation additives.

In view of the foregoing we propose to assume that immediately after a spray application, the air and condensation water are free of substance. In combination with the 0.1% deposition onto the roof (see Section 3.2), 1% direct contamination of the water in the mats and 0.3% direct contamination of the water in the troughs (see Section 3.10), and the distribution over crop and floor as given in Table 1, this leads to the initial distribution between crop, floor, roof, troughs and mats as shown in Table 4.

Table 4 *Recommended initial distribution of pesticide after spray applications for the reference deposition crops.*

Reference deposition crop	Fraction of applied dose				
	deposited on crop surface	deposited on floor	deposited on roof	dripped into mats	deposited on troughs
Cut flowers	0.857	0.129	0.001	0.01	0.003
Lettuce	0.857	0.129	0.001	0.01	0.003
Tomato and cucumber	0.772	0.214	0.001	0.01	0.003
Rose and gerbera	0.857	0.129	0.001	0.01	0.003
Very small young plants	0.00	0.989	0.001	0.01	0.003

In Section 3.2 we recommended to check whether c_a and $c_{w,cds}$ exceed the saturated vapour pressure and the water solubility, respectively. If the initial values of c_a and $c_{w,cds}$ are set to zero, there is no need for this check because there is no mechanism in the model that could lead to exceedance of these values.

Let us now consider LVM applications. Baas & Huygen applied parathion-ethyl also to tomatoes with a LVM. They found a maximum concentration in the greenhouse air of 0.95 mg m^{-3} which is much higher than the saturated vapour concentration of 0.11 mg m^{-3}. This 0.95 mg m^{-3} was measured in the first 15 min after application; between 125 and 190 min after application the concentration had decreased to 0.03 mg m^{-3} and between 190 and 300 min after application to 0.01 mg m^{-3}. If it is assumed that the concentration after 300 min is zero, the daily average concentration in the greenhouse air becomes 0.03 mg m^{-3}, i.e. about 30% of the saturated vapour concentration. As part of the same experiment Bor *et al.* (1994) measured a daily maximum concentration in the condensation water of 1.2 mg/L which is about 10% of the water solubility. As part of the same experiment Bor *et al.* (1994) measured also a deposition on the greenhouse roof of 1.2% of the dose. So the 1.2 mg/L in the condensation water is the result of both dissolution of the direct deposition and of the partitioning between the air in the greenhouse and the condensation water. Crum *et al.* (1991) applied methomyl to tomatoes with the LVM and measured only the sum of the concentration in the gas phase and the concentration in the droplets. Immediately after application they found a total concentration of about 15 mg m^{-3} (i.e. 30 times the saturated vapour concentration of 0.05 mg m^{-3}) and this total concentration decreased to about 0.04 mg m^{-3} within 1 hour. In the condensation water they measured a maximum concentration of about 5 mg/L, i.e. 0.01% of the water solubility of 55.000 mg/L. These results are not so easy to interpret but it seems defensible to assume that immediately after an LVM application the air in the greenhouse contains the saturated vapour concentration and that the condensation water is initially free of pesticide (the latter is the same assumption as for the spray applications). So we propose to follow this approach and to test whether simulated results are in line with the measured concentrations in the condensation water by Crum *et al.* (1991) and Bor *et al.* (1994).

In combination with the 1% deposition onto the roof (see Section 3.2), 1% direct contamination of the water in the mats and 0.3% direct contamination of the water in the troughs (see Section 3.10), and the distribution over crop and floor as given in Table 2, this leads to the initial distribution between crop, floor, roof, troughs and mats as shown in Table 5.

Table 5 *Recommended initial distribution of pesticide after LVM applications for the reference deposition crops. These fractions apply to the applied mass per surface area of greenhouse (A_i) minus the mass per surface area of greenhouse corresponding with the saturated vapour concentration ($A_{a,sat}$) because it is assumed that the concentration in the air initially is equal to this concentration.*

Reference deposition crop	Fraction of (Ai - Aa,sat)				
	deposited on crop surface	deposited on floor	deposited on roof	dripped into mats	deposited on troughs
Cut flowers	0.827	0.150	0.010	0.010	0.003
Lettuce	0.827	0.150	0.010	0.010	0.003
Tomato and cucumber	0.827	0.150	0.010	0.010	0.003
Rose and gerbera	0.827	0.150	0.010	0.010	0.003
Very small young plants	0.000	0.977	0.010	0.010	0.003

As described before, pesticides are in Dutch greenhouses not applied anymore by fogging (personal communication Erik van Os, 2018) so we recommend to exclude this application method for GEM-B (and also for GEM-C).

6.4 The concentrations in the greenhouse air and the condensation water

We propose to simulate the concentrations in the air and in the condensation water separately in order to be able to include dissolution of pesticide that was deposited onto the roof surface. Furthermore also volatilisation of this deposited pesticide should be included in view of the measurements by Crum *et al.* (1991) who found that only about 40% of the deposited pesticide methomyl ended up in the condensation water.

The proposed mass balance equation for the mass per surface area present in the condensation water is

$$\frac{dA_w}{dt} = -J_{a/w} + J_{dis} - J_{circ} - H_w \; k_{w,cds} \; c_{w,cds} \qquad \text{Eqn 57}$$

where J_{dis} is the mass flux of dissolution of pesticide that is deposited onto the roof surface (kg m^{-2} d^{-1}), $k_{w,cds}$ is the degradation rate coefficient of the substance in the condensation water (d^{-1}) and $J_{a/w}$ is the mass flux for exchange between water and air (kg m^{-2} d^{-1}) which is described by

$$J_{a/w} = GAI \frac{\left(K_H \, c_{w,cds} - c_{a,g}\right)}{r_a} \qquad \text{Eqn 58}$$

where GAI is the Glass Area Index (-) defined as the surface area of roof divided by the surface area of the greenhouse. The resistance r_a is set to 200 s/m = 2.32 $\times 10^{-3}$ d/m. This formulation has the consequence that the value of $J_{a/w}$ is positive if the flux is from the condensation water to the air. This convention was chosen because it seems most appropriate to consider the air compartment as the central compartment, so fluxes to this compartment have a positive sign (consistent with Eqn 11 of Appendix D of van der Linden *et al.*, 2015).

For the flux for the dissolution of pesticide deposited onto the roof we propose

$$J_{dis} = \alpha \, q_{cds} \frac{A_r}{A_{r,i}} \left(c_{w,sol} - c_{w,cds}\right) \qquad \text{Eqn 59}$$

where α is a proportionality factor (-), $c_{w,sol}$ is the water solubility of the pesticide (kg m^{-3}), A_r is the mass per surface area deposited onto the roof surface (kg m^{-2}), and $A_{r,i}$ is the initial value of A_r. The ratio $A_r/A_{r,i}$ is included because it can be expected that the surface area covered with deposits

decreases with decreasing A_r. The dimensionless factor α takes care of two reduction phenomena: (1) it is unlikely that there will be full equilibrium between the solid phase of pesticide deposited (in its formulation) onto the roof surface and the condensation water present on this roof (equilibrium would have the consequence that the pesticide concentration in the condensation water should approach the water solubility shortly after application), and (2) the currents of condensation water will not be in contact will all the pesticide deposition spots. Bor *et al.* (1994) measured daily average maximum concentrations of parathion-ethyl in the condensation water of 0.4, 1.0 and 1.2 mg/L shortly after application for three different application methods (with a measured q_{cds} in the order of 0.1 mm/d). Parathion-ethyl has a water solubility of 12 mg/L. So we set the default value of α tentatively at the average 0.9/12 = 0.07 and to consider in a sensitivity analysis a range from 0.4/12 = 0.03 to 1.2/12 = 0.10.

A_r is calculated as:

$$\frac{dA_r}{dt} = -J_{dis} - J_{v,r} \qquad \text{Eqn 60}$$

where $J_{v,r}$ is mass flux of pesticide volatilisation rate from deposit on roof (kg m^{-2} d^{-1}) which is described by

$$J_{v,r} = g\ GAI\ \frac{A_r}{A_{r,i}}\left(\frac{c_{a,sat} - c_{a,g}}{r_a}\right) \qquad \text{Eqn 61}$$

where g is a proportionality factor (-) to account for the fact that probably only a very small fraction of the roof is covered by this deposit and $A_{r,i}$ is the initial value of A_r immediately after application. We propose to set g tentatively at 0.01 and to check via sensitivity analysis whether this estimate requires further attention.

The mass flux of pesticide to the recirculation water is simply calculated as:

$$J_{circ} = q_{cds}\ c_{w,cds} \qquad \text{Eqn 62}$$

The relationship between A_a and $c_{a,g}$ is simply

$$A_a = H_a\ c_{a,g} \qquad \text{Eqn 63}$$

Similarly the relationship between A_w and $c_{w,cds}$ is

$$A_w = H_{w,cds}\ c_{w,cds} \qquad \text{Eqn 64}$$

The mass balance equation for A_a is based on Eqn 11 of Appendix D of van der Linden *et al.* (2015) plus the consideration that the volatilisation rate from the deposits on the roof has to be added:

$$\frac{dA_a}{dt} = +J_{a/w} + J_{v,p} + J_{v,f} + J_{v,r} - J_{vent} \qquad \text{Eqn 65}$$

6.5 Processes at the plant surface

As described in Section 3.8, there is ample evidence that the concentration in the air at the plant surface is considerably below the saturated vapour concentration shortly after spray applications. The concept of a saturated vapour pressure assumes that pesticide deposits on a leaf surface behave as deposits on an inert surface (e.g. such as stainless steel or perhaps glas). However, a leaf surface is not an inert surface; its surface consists of a wax layer and the cells in the leaf contain water.

As described in this section, initial concentrations in greenhouse air are available for many pesticides while the analysis of Section 3.7 showed that the volatilisation rate is so fast that initial concentrations in greenhouse air are close to those at the leaf surfaces. The most simple approach to account for this would be to introduce a reduction factor *F* as described by Eqn 39.

One might hypothesize that the reduction in the concentration in the air at the plant surface increases with increasing octanol-water partition coefficient and with increasing water solubility. We recommend to develop a regression model for this reduction factor *F* based on all available measurements in the literature. A first try could be to assume that the reduction factor is a linearly decreasing function of the logarithms of the octanon-water partition coefficient and of the water solubility. It is also an option to assess first the sensitivity of the GEM-B model to this reduction factor as a first step.

6.6 Fluxes in the gas phase

Combining the considerations in Section 3.3.1 with those in Section 6.5 we recommend to simulate volatilisation from the plant surface by

$$J_{v,p} = b \ \ LAI \ \frac{A_p}{A_{p,i}} \ \ \frac{F \ c_{a,sat} - c_{a,g}}{r_a} \qquad \text{Eqn 66}$$

with b = 2 for spray applications and b = 1 for LVM applications because spray applications lead to pesticide present at the underside of the leaves whereas this is not the case for LVM applications. Furthermore we recommend F = 0.2 based on the data shown in Table 3. Eqn 66 applies with the restriction that only positive values of the flux are considered acceptable, so if $c_{a,g} < F \ c_{a,sat}$ then the flux is set to zero.

We recommend to simulate volatilisation from the floor by

$$J_{v,f} = \frac{A_f}{A_{f,i}} \frac{c_{a,sat} - c_{a,g}}{r_a} \qquad \text{Eqn 67}$$

We recommend to use r_a = 200 s/m = 2.32 $\times 10^{-3}$ d/m for both processes.

We recommend not to include fluxes from the air to the plants and from the air to the floor because there is no clear mechanism that could provide a driving force for the flux from the air to the floor and because it is unlikely that both types of fluxes have a significant impact on peak concentrations in the condensation water.

Based on measurements by Stanghellini (1987) on heat exchange between plant leaves and greenhouse air we recommend to use r_a = 200 s/m = 2.32 $\times 10^{-3}$ d/m for all exchange processes between surfaces (leaves, floor, roof, pots) and the air (with the consequence that the diffusion coefficient in the air of the pesticide has no effect anymore on the flux). This estimate has a weak empirical basis for the floor, the roof and the pots. Therefore we recommend to include this r_a in the sensitivity analysis not as a single parameter but by considering r_a values for the different surfaces separately.

7 Proposed changes in GEM-C

7.1 Application methods

As indicated before, we propose to limit the application methods in GEM-B to (1) spraying and (2) room treatments with the LVM.

7.2 Initial distribution

In analogy to GEM-B we propose to assume that immediately after a spray application, the air and condensation water are free of substance and that 0.1% is deposited on the roof. Based on these assumptions and following the approach in Section 4.2 we propose to set f_p at 0.80 × 100/92.1 = 0.869 for spray applications. So then $A_{pot,i} + A_{tab,i} = 0.131\ A_i$ which gives $A_{pot,i}/A_i = f_{pot} \times 0.131$ and $A_{tab,i}/A_i = (1\text{-}f_{pot}) \times 0.131$. Considering that f_{pot} ranges usually from about 0.1 to 0.8 (see Figure 14), leads usually to a direct contamination of the recirculation water of 3-12% of the dose and a direct deposition onto the pots of 1-10% of the dose. This direct contamination of the recirculation water will probably dominate the emissions to the surface water strongly.

Direct contamination of troughs is no issue because there are no troughs in the ebb/flood system.

In analogy to GEM-B we propose to assume for LVM applications (1) that immediately after an application the concentration in the air is equal to the saturated vapour concentration, (2) that immediately after an application the concentration in the condensation water is free of pesticide, (3) that 1% of the dose is deposited on the roof. Following the approach in Section 4.2 we propose to calculate the fraction in the air, f_a, by

$$f_a = \frac{H_a\ c_{a,sat}}{A_i} \qquad \text{Eqn 68}$$

with the restriction that f_a cannot exceed 1 (this restriction is insignificant because a pesticide with such a volatility is unlikely to lead to any efficacy). In the current GEM-C version, 35% stays in the air, 55% is deposited on the crop and 10% is deposited on the pots plus the tables (see Section 4.2). So the ratio between the deposition onto the crop and the deposition onto the pots plus the tables was 55:10. Let us define the fraction of the dose deposited on pots plus table, $f_{pot+tab}$ and the fraction deposited on the roof by f_r. It then follows that

$$f_a + f_p + f_{pot+tab} + f_r = 1 \qquad \text{Eqn 69}$$

Furthermore we know:

$$f_{pot+tab} = \frac{10}{55} f_p \qquad \text{Eqn 70}$$

Combining these two equations gives

$$f_p = \frac{55}{65}\left(1 - f_a - f_r\right) \qquad \text{Eqn 71}$$

This gives the following expressions for $A_{pot,i}$ and $A_{tab,i}$:

$$A_{pot,i} = \frac{10}{65} \left(1 - f_a - f_r\right) f_{pot} \; A_i \qquad \text{Eqn 72}$$

$$A_{tab,i} = \frac{10}{65} \left(1 - f_a - f_r\right)\left(1 - f_{pot}\right) A_i \qquad \text{Eqn 73}$$

Usually f_a will be close to zero (see Figure 7) and f_r is only 0.01. If f_a = 0, the fraction deposited on the pots ($A_{pot,i}/A_i$) becomes 0.15 × f_{pot} and the fraction deposited on the tables ($A_{tab,i}/A_i$) becomes 0.15 × (1 - f_{pot}). Using again a range of f_{pot} of 0.1 to 0.8, GEM-C will usually calculate 3-14% direct deposition of pesticide on the water on the tables for LVM applications (and 3-12% for spray applications as described in the first paragraph of this section). This direct deposition is thus likely to be a much larger source of contamination of the recirculation water in GEM-C than the condensation water. We recommend to underpin these numbers by measurements for a few pot crops and for both spray and LVM applications.

In view of the probably large contribution of the direct deposition of pesticide on the water on the tables, we recommend to introduce a crop-specific f_{pot} value in GEM-C.

7.3 Description of processes

With respect to the description of the concentrations in the air and the condensation water and with respect to the volatilisation flux from the plants, we recommend to follow the proposals made for GEM-B in Sections 6.4, 6.5 and 6.6. As for GEM-B we propose to ignore deposition onto the plants via diffusion.

We propose to omit degradation in greenhouse air from GEM-C in view of the analysis in Section 3.6; this analysis applies also to GEM-C because the ventilation rate coefficient for greenhouses with crops in pots is similar to that for greenhouses with crops on slabs.

Based on the considerations in Section 4.4.1 we propose to replace the approach of a fixed 2-mm layer for diffusion into the soil in the pots by the following approach:

- a 5-cm top layer in which diffusion in both the liquid and the gas phase takes place and in which the water flow rate is set to zero;
- the diffusion flux at the bottom of this layer is set to zero mimicking in a simplified way the upward flow resulting from the irrigation events; this is more or less consistent with the current GEM-C model which assumes a 15-cm pot height of which the bottom 10 cm is wetted by the recirculation water during irrigation events (see Appendix E of van der Linden *et al.*, 2015);
- within this 5-cm top layer a linear sorption isotherm is assumed and first-order degradation (as did van der Linden *et al.*, 2015);
- in this layer the organic matter content is set at 10%, the dry bulk density at 1 kg/L and the porosity at 0.6 (as in the current version of GEM-C) but the volume fraction of liquid is set at 0.3 instead of the 0.06 assumed by van der Linden *et al.* (2015) because we consider this 0.06 far too dry for humic soil in pots whose bottoms are every two or three days inundated in a 5-cm water layer;
- all fluxes in this top 5-cm are described as mass rates per surface area of pots, so they do not contain the factor f_{pot} ; however, in the rate equation for A_a the flux from or to the pots is multiplied with the factor f_{pot} to ensure an adequate mass balance of A_a.

With respect to the interaction between the recirculation water on the tables and the pots, we recommend to assume that the water that is taken up by the pots as a result of an irrigation event contains a pesticide concentration that is equal to the concentration in the water on the tables during the irrigation event.

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Annex 1 List of symbols

Symbol	Quantity	Unit
b	parameter describing the increase of the available leaf area surface in case application occurs both on top and underside of the leaves	-
$c_{a,g}$	mass concentration of pesticide in greenhouse air	kg m^{-3}
$c_{a,g,0}$	mass concentration of pesticide in greenhouse air at start	kg m^{-3}
$c_{a,g,\infty}$	mass concentration of pesticide in greenhouse air after infinite time	kg m^{-3}
$c_{a,sat}$	mass concentration of pesticide in air corresponding with saturated vapour pressure (saturated vapour concentration)	kg m^{-3}
$c_{a,f}$	mass concentration of pesticide in air at floor surface	kg m^{-3}
$c_{a,pot}$	mass concentration of pesticide in air at surface of the pots	kg m^{-3}
$c_{a,tab}$	mass concentration of pesticide in air at surface of water on the tables	kg m^{-3}
$c_{a,p}$	mass concentration of pesticide in air at plant surface	kg m^{-3}
c_w	mass concentration of pesticide in water	kg m^{-3}
$c_{w,cds}$	mass concentration of pesticide in condensation water	kg m^{-3}
$c_{w,sol}$	mass concentration of pesticide in water corresponding with the water solubility of the pesticide	kg m^{-3}
d_{lam}	equivalent thickness of laminar air boundary layer	m
d_{min}	minimal thickness of pesticide layer at plant surface	m
d_{pot}	thickness of the top 2-mm layer of the pots	m
f_a	fraction of dose that stays in the air	-
f_f	fraction of dose applied to floor	-
f_p	fraction of dose applied to plants	-
f_{pot}	fraction of surface area that is covered with pots	-
f_r	fraction of dose applied to roof	-
f_{red}	factor for reduction of mass flux of pesticide to recirculation water	-
g	factor to account for coverage of roof by initial deposition on roof	-
k_{air}	rate coefficient for degradation in greenhouse air	d^{-1}
k_f	rate coefficient for decline of pesticide on floor	d^{-1}
k_p	rate coefficient for decline of pesticide on plants	d^{-1}
k_{pot}	rate coefficient for degradation in pots	d^{-1}
$k_{w,cds}$	rate coefficient for degradation in condensation water	d^{-1}
m_{mol}	molar mass of pesticide	kg mol^{-1}
m_{om}	mass fraction of organic matter in pots	-
m_{sub}	mass of pesticide in substrate tank	kg
m_{sys}	mass of pesticide in system of water and plastic tubes	kg
p	mass of plastic tube divided by length of plastic tube	kg m^{-1}
q_{cds}	volume flux of condensation water per surface area of greenhouse	m^3 m^{-2} d^{-1}
r_a	boundary layer resistance	d m^{-1}
t	time	d
A_a	mass of pesticide in air per surface area of greenhouse	kg m^{-2}
$A_{a,i}$	mass of pesticide initially in air per surface area of greenhouse	kg m^{-2}
$A_{a,sat}$	mass of pesticide in air per surface area of greenhouse when concentration in air equals saturated vapour concentration	kg m^{-2}
A_{a+w}	mass of pesticide in air plus condensation water per surface area of greenhouse	kg m^{-2}
A_f	mass of pesticide on floor per surface area of greenhouse	kg m^{-2}
A_i	mass of pesticide applied per surface area of greenhouse	kg m^{-2}
A_p	mass of pesticide on plants per surface area of greenhouse	kg m^{-2}

$A_{p,crit}$	mass of pesticide on plants per surface area of plants required to cover the surface area of the plants completely	kg m^{-2}
$A_{p,i}$	mass of pesticide applied to plants per surface area of greenhouse	kg m^{-2}
A_{pot}	mass of pesticide in the pots per surface area of greenhouse	kg m^{-2}
$A_{pot,i}$	mass of pesticide applied to the substrate in the pots per surface area of greenhouse	kg m^{-2}
A_r	mass of pesticide per surface area of greenhouse on roof	kg m^{-2}
$A_{r,i}$	mass of pesticide per surface area of greenhouse initially deposited on roof	kg m^{-2}
A_{ref}	reference mass of pesticide per surface area of greenhouse	kg m^{-2}
$A_{tab,i}$	mass of pesticide initially deposited on recirculation tables per surface area of greenhouse	kg m^{-2}
A_w	mass of pesticide in condensation water per surface area of greenhouse	kg m^{-2}
D_a	diffusion coefficient of pesticide in air	m^2 d^{-1}
$D_{a,ref}$	diffusion coefficient in air at reference temperature	m^2 d^{-1}
$D_{a,pot}$	diffusion coefficient in gas phase in pots	m^2 d^{-1}
F	factor for decrease of pesticide concentration at plant surface	-
GAI	Glass Area Index, i.e. surface area of roof divided by surface area of greenhouse	-
H_a	volume of air per surface area of greenhouse	m^3 m^{-2}
H_w	volume of condensation water per surface area of greenhouse	m^3 m^{-2}
$J_{a/w}$	mass flux for exchange of pesticide between water on roof and air in greenhouse	kg m^{-2} d^{-1}
J_{circ}	mass flux of pesticide from condensation water to recirculating water	kg m^{-2} d^{-1}
$J_{d,f}$	mass flux[6] of pesticide deposition onto floor	kg m^{-2} d^{-1}
$J_{d,p}$	mass flux of pesticide deposition onto plants	kg m^{-2} d^{-1}
$J_{d,pot}$	mass flux of pesticide deposition onto pots	kg m^{-2} d^{-1}
$J_{d,tab}$	mass flux of pesticide deposition onto water on tables	kg m^{-2} d^{-1}
$J_{dif,pot}$	mass flux of pesticide for diffusion in pots	kg m^{-2} d^{-1}
J_{dis}	mass flux of dissolution of pesticide on roof surface	kg m^{-2} d^{-1}
J_p	mass flux of pesticide at plant surface	kg m^{-2} d^{-1}
$J_{v,f}$	mass flux of pesticide volatilisation from floor	kg m^{-2} d^{-1}
$J_{v,p}$	mass flux of pesticide volatilisation from plants	kg m^{-2} d^{-1}
$J_{v,pot}$	mass flux of pesticide volatilisation from pots	kg m^{-2} d^{-1}
$J_{v,r}$	mass flux of pesticide volatilisation rate from deposit on roof	kg m^{-2} d^{-1}
$J_{v,tab}$	mass flux of pesticide volatilisation from water on tables	kg m^{-2} d^{-1}
J_{vent}	mass flux of pesticide leaving the greenhouse by ventilation	kg m^{-2} d^{-1}
K_{foil}	linear sorption coefficient for sorption of pesticide to foil	m^3 kg^{-1}
K_H	air-water partitioning coefficient	-
K_{om}	organic-matter/water distribution coefficient	m^3 kg^{-1}
K_{ow}	octanol-water partitioning coefficient	-
K_{sub}	linear sorption coefficient for sorption of pesticide to substrate	m^3 kg^{-1}
K_{tube}	linear sorption coefficient for sorption of pesticide to plastic tube	m^3 kg^{-1}
L	length of plastic tube	m
LAI	Leaf Area Index	-
M_{foil}	mass of foil	kg
M_{roots}	mass of wet roots	kg
M_{sub}	mass of dry substrate	kg
M_{tube}	mass of plastic tube	kg
N_{vent}	ventilation rate coefficient	d^{-1}
P	saturated vapour pressure of pesticide	Pa
Q	quotient for decrease of concentration due to sorption to plastic tube	-
R	gas constant	J mol^{-1} K^{-1}
RCF	root concentration factor	m^3 kg^{-1}
S	inner surface area of plastic tube	m^2

[6] In this list mass flux is defined as mass rate per surface area of greenhouse unless stated otherwise.

T	temperature	K
T_{ref}	reference temperature	K
V_{res}	volume of water in reservoir	m^3
V_{sub}	volume of water in substrate tank	m^3
X_{sub}	mass of pesticide sorbed per mass of dry substrate	kg kg^{-1}
X_{tube}	mass of pesticide sorbed per mass of plastic tube	kg kg^{-1}
X_{foil}	mass of pesticide sorbed per mass of foil	kg kg^{-1}
α	proportionality factor describing the efficiency of dissolution of pesticide deposited on roof surface	-
ε	volume fraction of gas in pots	-
θ	volume fraction of water in pots	-
λ	rate coefficient for volatilisation from plant surfaces	d^{-1}
μ	mass of pesticide in wet roots divided by mass of wet roots	kg kg^{-1}
ρ	mass of liquid or solid pesticide per volume of liquid or solid pesticide	kg m^{-3}
ρ_{pot}	mass of dry soil per volume of soil in pots	kg m^{-3}
φ_p	fraction of plant surface covered with pesticide	-
τ_{vc}	time constant for system with ventilation and flow of condensation water	d
τ_{vv}	time constant for system with ventilation and volatilisation	d
Φ	fraction lost by ventilation	-

Annex 2 Volatilisation of spray drift deposits from plant surfaces

Authors: H.J. Holterman & J.J.T.I. Boesten

Introduction

Leistra & Wolters (2004) proposed to assume that the volatilisation rate from spray drift deposits on leaf surfaces is proportional to the remaining mass of pesticide on the leaf surface based on the following argumentation: "The pesticide is assumed to be deposited on the leaves in spots of variable thickness. The thinner the deposit at a certain place, the sooner that place will be depleted by volatilisation. The concept is that the volatilising surface decreases in proportion to the decrease in mass of pesticide in the deposit."

The consequence of this assumption is that the remaining mass of pesticide on the leaf surface (assuming no other loss processes than volatilisation) under constant atmospheric conditions should decrease exponentially with time. The purpose of the exercise in this annex is to test whether the concept of Leistra & Wolters (2004) is valid using realistic distributions of the diameters of the spray droplets.

Model

A distribution of spray droplets of different sizes falling on a plant surface will transform into a distribution of droplets at the plant surface which have all the same shape (but different size). It can be assumed that the diameter of the droplets on the plant surface (d_p) is proportional to the diameter of the droplets in the air (d_a):

$$d_p = \alpha\, d_a \qquad \text{(A1-1)}$$

where α is a proportionality constant (-) that depends on the contact angle between the droplet and the plant surface. This contact angle is related to the hydrophobicity of the plant surface.

So the surface area of such a droplet at the plant surface (S_d) is given by:

$$S_d = \frac{\pi}{4} {d_p}^2 = \frac{\pi}{4} \alpha^2 {d_a}^2 \qquad \text{(A1-2)}$$

The volume of the droplet equals $\pi\ (d_a)^3/6$. Let us assume that the volume fraction of the pesticide in the droplet equals β (-). Then after evaporation of the spray solvent a cylindrical spot of pesticide residue remains with diameter $\alpha\, d_a$ and a height, h_s, as given by:

$$h_s = \frac{\frac{\pi}{6} \beta {d_a}^3}{S_d} = \frac{2}{3} \frac{\beta\, d_a}{\alpha^2} \qquad \text{(A1-3)}$$

Thus h_s is proportional to the diameter of the spray droplets. So large droplets do not only lead to large spots but also to thick spots.

The volume of pesticide in a spot equals the product of $h_s\ S_d$ and this volume is proportional to the mass of pesticide in a spot. So by considering the time course of this volume divided by the corresponding initial volume we obtain also the time course of the remaining fraction of the mass of pesticide on the plant surface.

Simulations were made with a model with three droplet size distributions as shown in Figure A2-1. These represent very fine, medium and very coarse droplet size distributions. The parameter α was set arbitrarily to 1 and β was set to 0.003. So a spray droplet with d_a = 100 µm will give h_s = 200 nm

with Eqn A1-3. Constant volatilisation conditions were assumed corresponding with a decrease rate of h_s of 1 nm/min. The droplet size distributions were discretized into classes with a width of 25 µm and the decrease in h_s was simulated using a time step of 1 min.

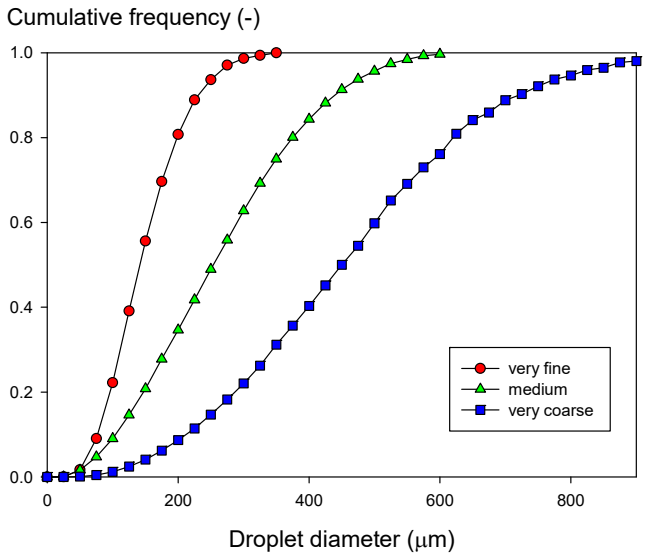

Figure A2-1 *Cumulative frequency distribution of the diameters of the three classes of droplets. The distribution is based on the volume of the droplets.*

Results and discussion

Resulting decrease curves of the remaining fraction of the dose in Figure A2-2 show that the droplet size distribution has a considerable effect on the speed of decrease. Coarse droplets lead to a slower decline than fine droplets. This is understandable because coarse droplets lead to thicker residue spots than fine droplets.

The purpose of this exercise was to check whether the decline proceeds exponential, i.e. giving a straight line in Figure A2-2. This is clearly not the case: the slope of the lines increases steadily with time.

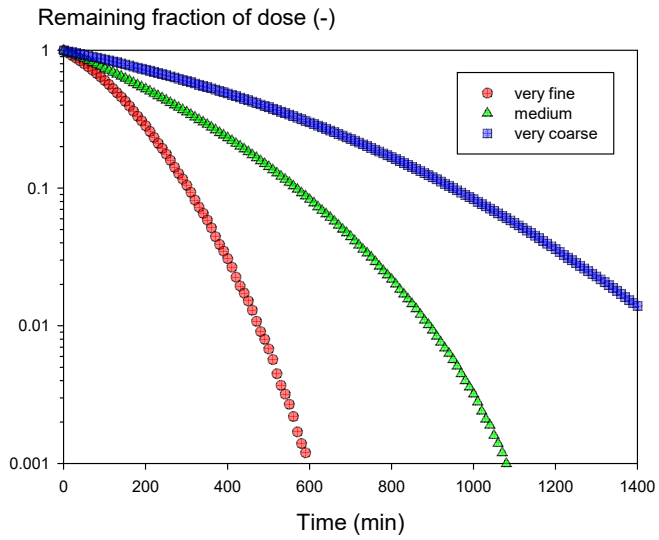

Figure A2-2 *Remaining fraction of dose (on a logarithmic scale) as a function of time simulated for the three distributions of droplet size as indicated.*

An alternative would be to assume that the volatilisation rate is not proportional to the remaining mass of pesticide on the plant surface. The consequence would be that the decline curve would be linear. Figure A2-3 shows that this is no good alternative: the decline curves are also clearly non-linear.

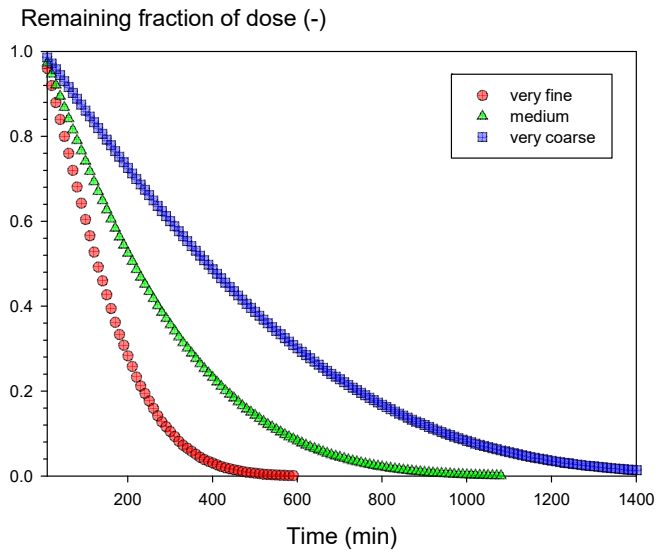

Figure A2-3 *Remaining fraction of dose (on a linear scale) as a function of time simulated for the three distributions of droplet size as indicated.*

The above model is of course a simplified representation of the deposition of spray drift and subsequent volatilisation from a plant surface. The model does not include overlap between droplets which may occur if high water volumes are used. It is of course also possible that Eqn A1-1 is too simple, e.g. due to local differences in roughness, coat of hair or wettability of the plant surface. Furthermore, the impact of the droplets on the plant surface is a dynamic process (e.g. large droplets may fall to pieces thus generating a number of small droplets).

Annex 3 Use of OECD-309 studies for estimating degradation rates in water in soilless cultivations in greenhouses

Introduction

The degradation of pesticides in the water in soilless cultivations in greenhouses may reduce emissions of these pesticides to surface water. It is therefore relevant to estimate this degradation rate as realistically as possible. The lower limit of this degradation rate can be based on the hydrolysis rate at the prevailing pH and temperature of the cultivation system. However, hydrolysis rates of most pesticides are slow. Pesticide dossiers may contain also OECD-309 studies, i.e. studies with freshly sampled surface water incubated in the dark (OECD, 2004). This OECD-309 guideline prescribes that such studies should be conducted in flasks that are filled with a water volume that is at most 1/3 of the total volume so leaving at least 2/3 of the volume for air (to keep the system aerobic, *i.e.* above 0.5 mg O_2/L). It recommends further that studies should preferably start within 1 day after collection of the surface water (allowing storage for at most 4 weeks at 4°C). The OECD-309 guideline offers two options for performing the test: a 'pelagic test' or a 'suspended-sediment test'. In a pelagic test no suspended sediment is added and in a suspended-sediment test sediment is added at concentrations ranging from 0.01 to 1 g/L. The intention of adding the sediment is to increase the microbial activity in the system. There may be considerable microbial activity in the water of soilless culture systems (Alsanius *et al.*, 2013). Thus it is worthwhile to assess whether suspended-sediment OECD-309 studies can provide a better estimate of the degradation rate in water in soilless cultivations than the studies on the hydrolysis rate.

Therefore an inventory was made of studies on the degradation rate in water in soilless cultivations in greenhouses. This inventory is reported below. This work was started at the end of 2016. Then, it was the intention to compare in a next step these degradation rates to the rates obtained in OECD-309 studies. However, recently it appeared that these studies are not yet available in pesticide dossiers. So the comparison to the rates obtained in OECD-309 is not yet possible.

Measurements of degradation rates in water in soilless cultivations in greenhouses available in literature

An overview of measurements available in the literature is given in Table A3-1. Data for some eight substances are available. The studies for azinphos methyl and cyromazine are labelled as unreliable because also other loss process than degradation may have contributed significantly to the observed decline. About half of the studies were performed with systems containing the roots and the other half were conducted with water that was sampled from the cultivation system. Only for metalaxyl, reliable DegT50 values are available. For most other pesticides only lower limits of the DegT50 are available. However, these are useful as well if the OECD-309 studies will generate lower DegT50 values than these lower limits.

For metalaxyl the DegT50 in a water-sediment system is 56 d (PPDB pesticide properties database). Metalaxyl is only moderately sorbed so the degradation rate in the water-sediment was much slower than the rates reported in Table A3-1. This indicates low microbial activity in the water-sediment studies. Adriaanse *et al.* (2012) reported that prosulfocarb degraded much faster in an outdoor mesocosm than in a water-sediment system and also attributed this difference to the comparatively low microbial activity in the water-sediment system.

Table A3-1 *Overview of measurements of degradation of pesticides in soilless growing cultures available in literature. DegT50 values with superscript 'u' are considered unreliable.*

substance	reference	substrate system			DegT50 (d)	initial	plant
		type	with roots ?	pH	in substrate system	concen-tration (mg/L)	
azinphos methyl	Flocco *et al.* (2004)	hydroponic culture	Y	5-6	11u	10	alfalfa
cyromazine	Karras *et al.* (2007)	pumice	Y		10u	not clear	gerbera
cyromazine	Patakioutas *et al.* (2007)	pumice	Y	5-7	16-20u	80-120	bean
etridiazole	Crum *et al.* (1985)	rockwool	N		>> 28	100	tomato
fenhexamid	Alsanius *et al.* (2013)	pumice	N	7-8	>> 21	8	tomato
imidacloprid	Alsanius & Bergstrand (2014)	pumice	N		>> 21	100	tomato
metalaxyl	Dunsing *et al.* (1988)	rockwool	Y		6	2	tomato
metalaxyl	Matser & Leistra (1997)	pumice	Y		9	25	tomato
		rockwool	Y		5	23	
metalaxyl	Karras *et al.* (2005)	pumice	Y		5-7	20-30	gerbera
oxamyl	Matser & Leistra (1997)	pumice	Y		> 22 d	27	tomato
		rockwool	Y		> 22 d	27	
pyrimethanil	Alsanius & Bergstrand (2014)	pumice	N		>> 21	10	tomato

Recent Dutch measurements of degradation rates in water in soilless cultivations in greenhouses

In 2012, 2014 and 2016 experiments were conducted in the Netherlands in rockwool systems to test the GEM model (van der Maas *et al.*, 2015; van der Linden *et al.*, 2017; Wipfler *et al.*, 2019). From these experiments estimates of the DegT50 were obtained based on inverse modelling or via considering the sensitivity to the DegT50. Table A3-2 gives the overview of the currently available information. Probably it is possible to estimate lower limits for most of the experiments. This would be useful because Table A3-2 contains four pesticides that do not occur in Table A3-1.

Table A3-2 *Overview of recent Dutch measurements of degradation of pesticides in soilless growing cultures. DegT50 values with superscript 'u' are considered unreliable. Roots were present in all systems.*

substance	reference	substrate		DegT50 (d)	initial	plant
		type	pH	in substrate system	concen-tration (mg/L)	
dimethomorph	van der Linden *et al.* (2017)	rockwool		>>6	22	cucumber
fluopyram	van der Linden *et al.* (2017)	rockwool		>>6	24	cucumber
imidacloprid	van der Linden *et al.* (2017)	rockwool		>>6	30	cucumber
imidacloprid	Wipfler *et al.* (2019)	rockwool		?		sweet pepper
propamocarb	van der Maas *et al.* (2015)	rockwool		1u	952	gerbera
pymetrozine	van der Maas *et al.* (2015)	rockwool		2u	52	gerbera
pymetrozine	van der Maas *et al.* (2015)	rockwool		0.5u	62	sweet pepper
pymetrozine	Wipfler *et al.* (2019)	rockwool		?		sweet pepper

Table A3-3 *Properties of studied pesticides taken from the PPDB pesticide properties database unless stated otherwise. The symbol σ indicates that the substance was stable in hydrolysis studies.*

substance	molar mass (g/mol)	water solubi-lity (mg/L) at 20oC	saturated vapour pressure (mPa) at 25oC	Henry coef-ficient KH (-)	log Kow	Kom (L/kg)	DegT50 hydrolysis (d) at 25oC and pH			photo-lysis ?
							5	7	9	
azinphos methyl	317	28	0.0005	0.0002	3.0	250-740	38	37-50	4-7	yes
cyromazine	166	13000	0.0004		0.1	20-1000	σ	σ	σ	
dimethomorph	388	29	0.0010	8E-09	2.7	180-300	-1	70	σ	slow
ethoprophos	242	1300	78	6E-06	3.0	20-100	σ	σ	83	
etridiazole	248	89	1430	0.0016	3.4	100-200	92	98	88	
fenhexamid	302	24	0.0004	2E-09	3.5	200-700	σ	σ	σ	
fluopyram	397	16	0.0012	1E-08	3.3	140-230	σ	σ	σ	yes
imidacloprid	256	610	4E-07	7E-14	0.6	60-240	σ	σ	σ	yes
metalaxyl	279	8400	0.75	1E-08	1.8	16-160	-2	106	115	no
oxamyl	219	148100	0.051	3E-11	-0.4	5-23	σ	8	0.1	yes
propamocarb	188	900000	730	6E-08	0.8	-				
pymetrozine	217	270	0.0042	1,3E-09	-0.2	140-1800	9	σ	σ	yes
pyrimethanil	199	121	1.1	7E-07	2.8	50-300	σ	σ	σ	

1) stable at pH 4

2) no decline after storing nutrient solution at 5°C for 7 months or after storing both aerated and non-aerated aqueous solutions at room temperature for 42 d (Dunsing *et al.*, 1988)

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Annex 4 Density of plants growing in Dutch greenhouses

The data below are based on a personal communication of Daniel Ludeking (2015).

This table is for pot plants.

Crop (with between brackets the pot size)	Type of crop	Number of plants per m^2	Fraction of surface area covered by the pots (-)	Table (T) or floor (F)
Seedlings Lisianthus	young plants	750	0.2	F
Seedlings summer flowers	young plants	500	0.29	F
Cuttings Chrysanthemum (blocks of 5 cm)	young plants	400	1	F
Cuttings rose	young plants	25	0.25	F
Seedlings vegetables	young plants	15	0.15	F
Seedlings, young pot plants	young plants	150	0.29	F
Cutting, young pot plants	young plants	150	0.29	F/T
Ficus (21 cm)	pot plant	7	0.24	F
Kalanchoe (10.5 cm)	pot plant	48	0.42	T
Phalaenopsis (12 cm)	pot plant	35	0.395	T
Bromelia, Guzmania (10.5 cm)	pot plant	40	0.34	T
Begonia (13 cm)	pot plant	17	0.23	T
Cyclamen (12 cm)	pot plant	32	0.36	T
Anthurium (17 cm)	pot plant	11	0.24	T
Euphorbia pulcherima (13 cm)	pot plant	10	0.13	T
Zamioculcas (17 cm)	pot plant	32	0.72	F
Spathiphyllum (10.5 cm)	pot plant	25	0.22	T/F
Calathea/ Marantha (19 cm)	pot plant	10	0.28	T/F
Chamedoria (13 cm)	pot plant	35	0.46	F
Dracaena (13 cm)	pot plant	40	0.53	F
Yucca (17 cm)	pot plant	20	0.45	F
Dieffenbachia (13 cm)	pot plant	30	0.4	F
Pot rose (13 cm)	pot plant	23	0.31	T
Pot chrysanthemum (13 cm, 3 cuttings)	pot plant	22	0.29	T
Bulb chrysanthemum (17 cm)	pot plant	4	0.09	F/T
Pelargonium zonale (Geranium) (10.5 cm)	pot plant	35	0.3	T
Impatiens (12 cm)	pot plant	17	0.19	T
Petunia (10.5 cm)	pot plant	40	0.34	T
Saint paulia (12 cm)	pot plant	35	0.4	T
Fuchsia (19 cm)	pot plant	9	0.26	T
Hortensia (Hydrangea) (13 cm)	pot plant	7	0.09	T
Schefflera (13 cm)	pot plant	30	0.4	F/T

This table is for crops growing on substrates such as stone wool.

Crop	Type of crop	Number of plants per m^2	Fraction of surface area covered by the pots (-)
Aubergine	vegetable on substrate	1.6	0.016
Cucumber	vegetable on substrate	1.5	0.015
Cucumber, high wire	vegetable on substrate	1.75	0.0175
Tomatoes	vegetable on substrate	2.5	0.025
Courgette	vegetable on substrate	1.3	0.013
Paprika	vegetable on substrate	2.3	0.023
Vine tomatoes 'cocktail'	vegetable on substrate	2.5	0.025
Cherry tomatoes	vegetable on substrate	2.5	0.025
Lettuce	vegetable on substrate	16	0.16
Rose	cut flowers on substrate	7	0.07
Gerbera	cut flowers on substrate	5.5	0.055

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