Science of the Total Environment 825 (2022) 153798

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

OBOMod - Integrated modelling framework for residents' exposure to pesticides

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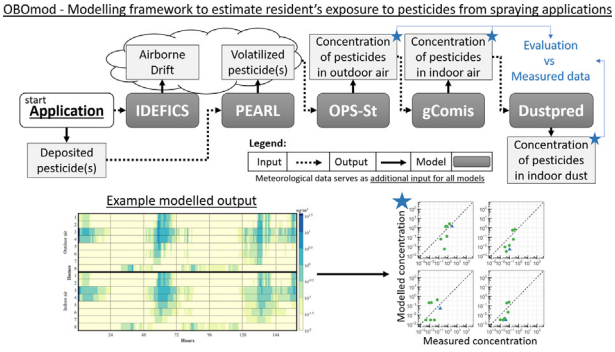
HIGHLIGHTS

- A modelling framework (OBOmod) to estimate pesticide concentrations is proposed.
- In general, estimates remained within one order of magnitude from measured levels.
- OBOmod can be used to estimate residential outdoor and indoor air concentrations.
- Exposure to emissions from volatilization can be higher than exposure to spray drift.
- OBOmod can support policy making and be used in health and epidemiological studies.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 7 October 2021
Received in revised form 21 January 2022
Accepted 7 February 2022
Available online 10 February 2022

Editor: Yolanda Picó

Keywords:
Air
Dust
Plant protection product
Emission
Vapor
Airborne spray drift

ABSTRACT

Background: Pesticides can be transported from the site of application to homes via different routes and lead to exposure of residents, raising concerns regarding health effects. We built a deterministic model framework (OBOmod) to assess exposure of residents living near fields where pesticides are applied.
Methods: OBOmod connects five independent models operating on an hourly timescale and high spatial resolution (meters). Models include descriptions of spray drift, volatilization, atmospheric transport and dispersion, exchange between outdoor and indoor air and exchange between indoor air and dust. Fourteen bulb field applications under different weather conditions and comprising 12 pesticides were simulated. Each simulation included the first seven days after the application. The concentrations computed with OBOmod were compared with those measured in outdoor and indoor air and the amounts measured in indoor dust samples.
Results: Model evaluation indicated suitability of the developed framework to estimate outdoor and indoor air concentrations. For most pesticides, model accuracy was good. The framework explained about 30% to 95% of the temporal and spatial variability of air concentrations. For 20% of the simulations, the framework explained more than 35% of spatial variability of concentrations in dust. In general, OBOmod estimates remained within one order of magnitude from measured levels. Calculations showed that in addition to spray drift during application, volatilization from the field after spraying and pesticides in house dust are important routes for residents' exposure to pesticides.

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http://dx.doi.org/10.1016/j.scitotenv.2022.153798

Conclusions: Our framework covers many processes needed to calculate exposure of residents to pesticides. The evaluation phase shows that, with the exception of the dust model, the framework can be used in support of health and epidemiological studies, and can serve as a tool to support development of regulations and policy making regarding pesticide use.

1. Introduction

1.1. Exposure of residents to pesticides

Farmers, operators, workers, residents, and bystanders may be exposed to pesticides. In the past, assessment of the exposure was focussed on operators, workers, and bystanders (Waheed et al., 2017). In the last 10 years however, residents' exposure has been an increasingly researched topic (Pubmed, 2021). This is likely due to growing concern among the population regarding pesticide usage (Calliera et al., 2019; Schaub et al., 2020; Zeitlin et al., 2021). But also due to efforts of policy makers to study residents' exposure (e.g. Health Council of the Netherlands, 2014). A recent literature review by Dereumeux et al. stresses two important things: "There is evidence that residents living close to agricultural fields are more exposed to pesticides than the general population"; "Some epidemiological studies suggest an association between proximity to agricultural lands and a wide range of associated adverse health outcomes" (Cited from Dereumeaux et al., 2020). Adding to this, it is known that i) part of the exposure of residents to pesticides is attributable to exposure via environmental routes (Cornelis et al., 2009), and ii) contrarily to dietary exposure, much of the environmental exposure is beyond the control of the average individual (Oates and Cohen, 2011). These highlight the importance to understand and quantify the environmental exposure routes and feed this knowledge to regulatory entities.

1.2. Environmental routes contributing to exposure of residents

There are several environmental routes contributing to exposure of residents (Falette et al., 2018). One of the main routes to exposure of residents, living nearby agricultural fields where pesticides are applied, is the spray drift of pesticides through air (Holterman and van de Zande, 2010). During pesticide application, droplets can evaporate, drift and can remain airborne. Besides depositing on the target area, droplets can drift away and deposit in an off-target area (Steward et al., 2019). The percentage of the active ingredients that drift or evaporate before they reach the plants is highly dependent on physiochemical properties of the pesticides, weather conditions and the spraying conditions (Soheilifard et al., 2020).

A fraction of the deposited ingredient can then volatilize (Van den Berg et al., 1999; Langenbach et al., 2021), depending on its vapor pressure and several other parameters (e.g., absorption capacity to soil, penetration into leaves, degradation rates of the active material). The volatilized substance then moves with the wind (Zivan et al., 2016, Taylor et al., 2020).

The pesticide can then be transported via air in the direction of residences (Veludo et al., 2022) and there infiltrate into the house via openings such as open doors, windows, cracks, chimneys. Concentrations outside and inside the residence tends towards an equilibrium and, in theory, the concentration inside the house may reach the air concentration level outside if volatilization and meteorological conditions remain the same (Sangiorgi et al., 2013). However, this might often not be the case at all because of rapid changes of wind direction, source strength, atmospheric mixing, among others. To estimate short term indoor exposure (sometimes with the highest concentrations) it is also needed to have knowledge of the development of these processes in time, explicitly.

Finally, since in the indoor environment pesticides may be present in the gas-phase they can also be adsorbed to indoor dust particles (Butte, 2004). Dust particles can aggregate, and, in this state, pesticides can accumulate in the indoor environment. This can lead to exposure via contact with contaminated surfaces and/or incidental dust ingestion (Tames et al., 2020).

1.3. The need for modelling exposure through the different routes

To combine all of the aforementioned different routes of exposure, for a given spatial scale, models can play an important role: models allow to analyse and quantify exposure to a given pesticide (Butler Ellis et al., 2017), as well as generalize the results of observations and extrapolate results to other places with similar or different settings. Measurements, such as biomonitoring, have proven efficient to understand to what extent residents can be exposed. However, they may suffer from detection problems and limitations to the number of pesticides that can be assessed. In addition, they tend to be very time consuming and costly (Atabila et al., 2017) especially if the aim is to understand exposure of large populations, for many pesticides at the regional or even national scales. Models can be used to make exposure estimates on these scales.

1.4. Aim of the study

The aim of our study was to develop a modelling framework to estimate resident's exposure to pesticides from spraying applications. We focused on pesticide application using a boom sprayer since this is the most used technique in the Netherlands, in Europe and worldwide at large-scale farms (Fujimoto et al., 2016). In our framework, here forth named OBOmod, independent models are used to describe the processes in the causal chain of spraying, droplet drift during application, volatilization of deposited pesticides from vegetation and soil during and after spraying, gas and aerosol dispersion, exchange of pesticides between outdoor and indoor air and sorption of pesticides to house dust. We describe each model individually and how they are connected in the framework. The OBOmod is applied to several case studies on different locations, for different pesticide mixtures and meteorological conditions. This allows us to test the versatility of the framework by simulating distinct real-life scenarios. Case study data with pesticide concentration measurements in outdoor and indoor air, and indoor dust were used to verify model results at different steps along the model chain. The novelty here is the integration of various pathways and evaluation of a framework fit for residential exposure assessment. We show how the OBOmod can be used and provide information so it can be applied in other studies.

2. Methods

2.1. The modelling framework

Five models were selected to build the OBOmod, to quantify contributions from the aforementioned pathways to residents' exposure (Figueiredo et al., 2018). The models are:

- Airborne spray drift: IDEFICS Model. Tests have been reported in several publications, such as Holterman et al. (1997), Holterman et al. (1998), Stallinga et al. (2008).
- Volatilization: PEARL Model. Tests have been reported in several publications, such as Leistra and Wolters (2004), Leistra et al. (2005), Leistra and van den Berg (2007), Van den Berg et al. (2016a).
- Atmospheric short term/short range transport: OPS_Ste Model. Tests have been reported in several publications, such as Van Jaarsveld (2004), Sauter et al. (2018). The potential use of OPS for pesticide transport has been highlighted in van den Berg et al. (2016b) and Butler Ellis et al. (2017).
- Atmospheric transport from outdoors to indoors: gCOMIS Model. Tests have been reported in several publications, such as Phaff (1996), Borchiellini and Furbringer (1999).

- Sorption of pesticides to indoor dust: DUSTPRED Model. Tests have been reported by Weschler and Nazaroff (2010).

These models are well described in literature and a short explanation of each model can be found in Supplementary material A.

As shown in Fig. 1, the OBOmod follows the causal chain. It starts with the application where the IDEFICS model (Holterman et al., 1997), describes spray drift and calculates the deposition and concentration of spray droplets in air, up to five meters downwind of the sprayed area. Input to the model is parameters that were collected prior to the spraying event and based upon measurements (e.g. composition of the pesticide mixture in the tank, spray boom height, nozzle type and size and spray drop characteristics). The minimum set of data needed is i) day of spraying; ii) tank mixture; iii) sprayer and nozzle type. It is possible to run the model with "expert decision" input as long as one it's clear about the quality of the input data. A detailed description of the droplet size distribution for each type of nozzles used in OBOmod can be found in Holterman and van de Zande, 2019.

The PEARL model (van den Berg et al., 2016b), quantifies volatilization from plants after the application. Inputs are physico-chemical properties of the pesticides and an estimate of the amount of deposited material (i.e. applied quantity minus primary drift). Furthermore, the model is driven by meteorological conditions. The computed volatilization is regarded as an emission source strength. Next, the OPS-St (Short term version of the Operational Priority Substances model) advanced Gaussian plume model (Sauter et al., 2015) is used to compute atmospheric transport, dispersion and resulting concentrations at receptor points around the fields. Aerosols already present in the background air were not taken into account. During application, the source strength input to OPS is taken to be the emission of pesticide mass in droplets as computed by IDEFICS at 5 m from the downwind edge of the applied field. IDEFICS can deal with any downwind distance, but larger distances take more computing time. The distance of 5 m was chosen to make sure that larger drops sedimented to the ground and that only the smaller drops remained airborne. After application, the volatilization strength computed by PEARL is used as a source strength from the applied field. Concentrations calculated with the OPS-St model then serve as input to the ventilation model gComis (Feustel and Raynor-Hooson, 1990). This model estimates concentrations in indoor air based on outdoor concentrations, using exchange rates between indoor and outdoor air. These rates are based on building characteristics obtained for each individual home and were assumed to be similar for gas-phase and particle-phase pesticides, given that larger particles will settle before reaching the home. Finally, an equation (Eq. (2) Weschler and Nazaroff, 2010), here called Dustpred, is used to calculate the concentration of pesticides that will be present in indoor dust based on indoor air concentrations. All model inputs are described in more detail in (OBO, 2019).

Wind erosion of particles from agricultural fields (Silva et al., 2019) was not considered in the modelling chain, since in the Netherlands this process is very unlikely to occur, given the high soil moisture content (Wösten et al., 2001) and the average height of the plant canopy, which serves as a barrier for erosion. For countries with low precipitation and low soil moisture content the wind erosion route might be of relevance, so it would need to be added to the OBOmod. The transport of pesticides via shoes, clothes and pets into the indoor environment was not included in the OBOmod either. Although hypothesized as important (Bradman et al., 2009), no model has been reported in the literature for this exposure route (take-home) of pesticides.

The models in the OBOmod are run independently, that is, they are connected solely via input-output information. Output is generated on an hourly basis, allowing us to look at both short (hours) and long-term (weeks) exposures. Spatial resolution is in meters and the model computes values for a-priori defined grids or receptor points.

2.2. General conditions and measured data

Fourteen spraying events under different meteorological conditions and settings and for various pesticide mixtures were simulated (Supplementary material B). All events were real spraying applications planned by the farmer and carried out during the OBO project (OBO, 2019). These events occurred on selected fields described in Figueiredo et al. (2021a).

All planned spraying applications were carried out in the North-western part of the Netherlands, an area with intensive flower bulb growing. Spraying was performed in 2016 and 2017 during all months except September, October, November, and December. Temperatures ranged from 1 °C in January to 32 °C in July but were generally moderate with an average of 14 °C and a median of 16 °C. Relative humidity ranged from 65 to 85%. Wind direction was predominantly from SW. Between hour variability was less than 10 degrees for 75% of all simulated hours. Day-to-day variability was high (usually more than 45 degrees). Wind speed during spraying was low in general and usually below 5 m/s at 10 m height (compliant with Dutch regulations for pesticide spraying). These observations were taken from nearby stations from the Dutch meteorological service KNMI. The observed wind speed, during the periods of application, is quite low for this area. But low wind speeds are, understandably, conditions that are favorable for farmers due to lower losses by drift to neighboring crops and surface water. In total, some 20 different active ingredients were sprayed, 45% being fungicides, 25% herbicides and 30% insecticides. These were applied on nine different flower species including tulips and lilies. Usually, sprayed mixtures contained two or three active ingredients. The choice of active ingredients and doses were made by the farmer.

In this study, 24-hour averaged samples of outdoor air were collected for a period of seven days for every home (total number of homes = 96), during and after the spraying event. Air was sampled through a standard PM10 inlet. We used a combination of a filter and XAD absorbent to collect volatile and particle bound pesticides, these were then pooled for chemical analysis. Also, one air sample was collected inside the homes on the day of spraying (i.e. day 1) (Figueiredo et al., 2021b). In addition, one week after the spraying event, one dust sample was collected in every home using a vacuum cleaner equipped with a special filter (Figueiredo et al., 2021c). It was used on a 4 to 6 m^2 surface depending on the available area (Figueiredo et al., 2019). This, vacuumed floor dust (VFD), is a good representation of pesticides in indoor dust (Curwin et al., 2005) and was used to evaluate the Dustpred model.

All samples were analysed for 46 active ingredients within a few weeks after sampling by using state of the art LC-MS methods (Figueiredo et al.,

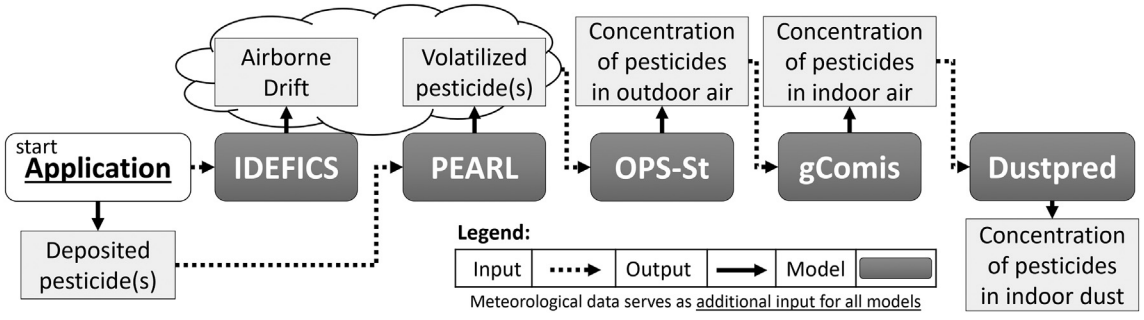


Fig. 1. Integrated modelling framework (adapted from Figueiredo et al., 2021a).

2021a). All experimental data and the results of our simulations can be found in the OBO technical report (OBO, 2019). None of the pesticides were reported as being used indoors in the participating homes (see Supplementary material E from Figueiredo et al., 2021b). All measured pesticides and relevant physico-chemical properties are presented in Table 1.

2.3. Testing model components of the framework

As a first step, we used measured outdoor air concentrations to test the IDEFICS-PEARL-OPS chain. In this step, we included only applications of which we had all required information (i.e. there was no input data missing). It should be noted that the input data for the PEARL model was obtained from data published in the literature and not by independent measurements. In addition, volatilization was assumed to be the only loss process, so the potential contribution of other processes on the plant surface, such as photo-transformation, penetration into the plant tissue and wash-off were not considered. Especially important here is that, apart from the selected field, no other agricultural field within an area of 250 m from homes applied the same pesticide in the same day. We considered contributions from distances above 250 m to be negligible (Gibbs et al., 2017; Figueiredo et al., 2021b) and we only considered volatilization from treated fields, not from off-target areas where pesticides could have been previously deposited after atmospheric dispersion.

Given that these are evaluation steps we used concentrations measured outside close to the homes as input to the gComis model. The Gcomis model was therefore evaluated by comparing indoor concentrations calculated by gComis to concentrations measured indoors to verify the gComis calculations.

As a last step, we used measured indoor air concentrations to predict daily averaged concentrations of different pesticides in indoor dust and compared the average weekly concentration with the VFD. It should be noted that, VFD may also contain pesticides that accumulated before the

Table 1
List of measured pesticides (active ingredients) and relevant physico-chemical characteristics.

Type	Pesticides	CAS RN	DT50 soil[a]	Water solubility[b]	Vapor pressure[c]
Herbicides	Asulam	3337-71-1	[2.1, 39]	962,000	1.9E−01
	Chloridazon	1698-60-8	[3.0, 173.9]	422	6.0E−02
	Chlorpropham	101-21-3	[2.8, 42.8]	110	2.4E+01
	Dimethenamid-P	163515-14-8	[5.0, 31.0]	1499	2.5E+00
	Linuron	330-55-2	[10.1, 168.4]	63.8	1.9E−01
	Metamitron	41394-05-2	[2.2, 44.5]	1770	8.6E−04
	Pendimethalin	40487-42-1	[39.8, 270.0]	0.33	1.3E+00
	S-metolachlor	51218-45-2	[3.6, 221]	530	4.2E+00
	Sulcotrione	99105-77-8	[1.2, 89.7]	165	5.0E−03
	Terbuthylazine	5915-41-3	[6.43, 167.0]	6.6	9.0E−02
Insecticides	Acetamiprid	135410-20-7	[0.8, 5.4]	2950	5.9E+00
	Cyhalothrin-lambda	91465-08-6	[10.1, 1000.0]	< 1	4.5E−04
	Deltamethrin	52918-63-5	[12.5, 231.0]	< 1	1.2E−05
	Flonicamid	158062-67-0	[0.7, 1.8]	5200	9.4E−04
	Fosthiazate	98886-44-3	[9.0, 17.0]	9000	5.6E−01
	Imidacloprid	138261-41-3	[77.0, 425.0]	610	2.1E−01
	Oxamyl	23135-22-0	[0.6, 19.4]	148,100	3.1E+01
	Pirimicarb	23103-98-2	[5.5, 274.0]	3100	9.7E−01
	Pymetrozine	123312-89-0	[2.05, 183.0]	270	1.8E−03*
	Spirotetramat	203313-25-1	[0.05, 1.0]	30	6.0E−06*
	Thiacloprid	111988-49-9	[0.33, 16.8]	184	8.0E−07
Fungicides	Azoxystrobin	131860-33-8	[35.2, 261.9]	7	1.1E−07
	Boscalid	188425-85-6	[27.0, 1214.4]	5	7.2E−04
	Carbendazim[d]	10605-21-7	[11.0, 120.0]	8	1.0E−04
	Cyprodinil	121552-61-2	[11.0, 98.0]	13	4.9E−01
	Difenoconazole	119446-68-3	[20.0, 456.0]	15	3.3E−05
	Dimethomorph	110488-70-5	[17.8, 599.1]	29	9.9E−01
	Fludioxonil	131341-86-1	[8.0, 365.0]	2	3.9E−04
	Fluopicolide	239110-15-7	[77.0, 333.0]	3	8.0E−04
	Fluopyram	658066-35-4	[93.2, 717.0]	16	4.2E−03
	Flutolanil	66332-96-5	[60.4, 1000.0]	8	1.8E+00
	Kresoxim-methyl	143390-89-0	[0.37, 1.85]	2	2.3E−03
	Mepanipyrim	110235-47-7	[34.0, 253.9]	2	2.3E−02
	Prochloraz	67747-09-5	[22.1, 936.1]	27	1.5E−01
	Propamocarb	24579-73-5	14	900,000	7.3E+03
	Prothioconazole	178928-70-6	[0.04, 1.4]	23	4.5E−09*
	Pyraclostrobin	175013-18-0	[4.2, 181.0]	2	2.6E−05
	Tebuconazole	107534-96-3	[25.8, 610.0]	36	1.7E−03
	Thiophanate-methyl	23564-05-8	[0.29, 3.3]	19	9.5E−03
	Toclofos-methyl	57018-04-9	[2.1, 16.4]	<1	5.7E+01
	Trifloxystrobin	141517-21-7	[0.13, 2.83]	<1	3.4E−03
	Fluopyram-benzamide	360-64-5	[6.7, 11.5]	ND	ND
	Metamitron-desamino	36993-94-9	[17.0, 39.7]	400	4.5E−04*
Degradation products	Prothioconazole-desthio	120983-64-4	[4.56, 32.2]	51	1.1E−03*
	Spirotetramat-enol	203312-38-3	[0.02, 10.9]	2700	ND
	Trifloxystrobin-acid	252913-85-2	[21.1, 406.8]	21,000	5.5E−03**

ND (Not determined).
* Estimated using EPI Suite™ (https://www.epa.gov/tsca-screening-tools/epi-suitetm-estimation-program-interface).
** IUPAC Ref: CGA 321113.
[a] Half-life (degradation) in soil (days). Range of values from field and lab studies gathered from Lewis et al., 2016.
[b] Water solubility (at 20 °C [mg l^{-1}], integers presented) gathered from Lewis et al., 2016.
[c] Vapor pressure in milipascal (measured at 20 °C or 25 °C, depending on study) – collected from Pubchem (https://pubchem.ncbi.nlm.nih.gov).
[d] Besides being a fungicide, carbendazim is also an environmental degradation product from thiophanate-methyl.

sampling period. This aspect may lead to significant differences between calculated and observed concentrations in house dust.

2.3.1. The three evaluation steps

In summary, three model evaluation steps were considered:

1) IDEFICS, PEARL and OPS-ST, to calculate, respectively, droplet drift during spraying, volatilization from crop and dispersion of gaseous pesticides on day 1 to 7. The concentrations computed with this model suite are compared with the measured 24-hr (daily) air concentrations outside homes.
2) Gcomis, to calculate concentration inside homes on day 1 based upon concentrations measured outside. The concentrations computed with this model are compared with measured 24-hr average air concentrations inside homes.
3) Dustpred, to calculate daily content (mass) in house dust from measured concentrations in indoor air. The average concentrations computed with this model are compared with the those measured in VFD inside homes.

From the fourteen simulations, only five had specific pesticide mixtures being applied solely in the selected field (see Simulations 1–5, Supplementary material B). Therefore, evaluation for step 1 was solely done using data from these five applications.

In this way, we used concentrations measured outside the homes, concentrations measured inside homes and concentrations measured in indoor dust to test each submodel in the OBOmod. This approach allows us to reduce uncertainty throughout the model evaluation process, by avoiding errors propagated from other model steps.

2.3.2. Metrics used to assess the quality of model estimates

The statistical measure of the quality of model estimates was determined by the coefficient of determination (R^2, or explained variance) between measured and modelled concentrations. Two additional metrics were used to assess the difference between modelled and measured values. We calculated the RMSE (Root Mean Squared Error) and the MAE (Mean absolute error). RMSE is analogous to the standard deviation (SD), since it accounts for the magnitude of the residuals, while MAE takes the average magnitude of the residuals. As proposed by Shmueli et al. (2016), we will use SD of the observations as a benchmark to compare with MAE and RMSE, given that SD is the amount of error that naturally occurs in the measured values. Metrics were computed from non-transformed data. Given the small number of paired samples above LOD for acetamiprid, efficiency metrics could not be calculated for this pesticide.

As additional data analysis, we i) investigated if there were systematic discrepancies between the modelled and the measured values (i.e. proportional bias) via modified Bland-Altman plots and ii) calculated Spearman correlation coefficients for gComis and Dustpred evaluation steps. This was done as a sensitivity analysis of model performance. Both indoor air and indoor dust are known to be influenced by sources and sinks present inside the home (e.g. resuspension, long-term accumulation, dragging in), which may cause the relation between measured and modelled data to be non-monotonic. The results from this sensitivity analysis can be consulted in Supplementary material C.

2.3.3. Temporal and spatial variability in concentrations

The quality of a model to estimate concentrations in outdoor air can be judged by looking at temporal (between days) and spatial (between homes) variability. For temporal variability, R^2, MAE and RMSE were calculated for each pesticide and per home, using daily averages (see Table D.1, Supplementary material D). Scatter plots ($N = 29$) are presented for all measured vs modelled comparisons (see Fig. D.1 in Supplementary material D) and a selection of representative cases ($N = 18$) is discussed here.

For spatial variability, R^2, MAE and RMSE were calculated per pesticide and per day, again using daily averages of concentration (see Table D.2, Supplementary material D). Scatter plots ($N = 48$) were created for all measured vs modelled comparisons and are here discussed.

For indoor air and dust, where we had only one observation per home, metrics were calculated per pesticide but grouping the results from all homes into one single assessment. Here, R^2 refers to the proportion of spatial variability that we can explain with the model. Scatter plots of measured vs modelled data for both indoor air and dust, including all pesticides, can be found in Supplementary material E.

3. Results

3.1. Pesticides - summary of properties and measured concentrations

In the period that the pesticides were used on the selected fields they could be detected in nearly all 24 h outdoor air samples. Pesticide concentrations observed in outdoor air ranged from 0.003 (the detection limit for most pesticides) to 2750 ng/m^3. Concentrations observed in indoor air were lower than those in outdoor air ranging from concentrations below the detection limit (<0.003 ng/m^3) to 25 ng/m^3 on the day of application (Figueiredo et al., 2021b). Concentrations in vacuum cleaner floor dust ranged from 1 ng/g dust to 27000 ng/g dust (Figueiredo et al., 2021c). Based on EFSA guidance documents (EFSA (European Food Safety Authority), 2014), solely chlorpropham and propamocarb are considered highly volatile compounds (see Table 1, vapor pressure > 10 mPa). The remaining pesticides are considered to have low volatility, although pendimethalin can be considered moderately volatile (Lewis et al., 2016). The half-life of the pesticides in soil (DT50 in Table 1) could also be a parameter that plays a role in the outcome of model simulations. It appears however that the reported half-life in soil is quite variable for most compounds, so we did not take this parameter into account when interpreting results. Finally, 13 of the measured pesticides have high water solubility (i.e. amount of chemical substance that can dissolve in water). These group (see Table 1, water solubility >500) have higher volatilization potential if the plant leaves are wet, since volatilization occurs at the water – air interface.

3.2. Example output of simulations

As an example, we show in detail how the OBOmod is setup for one simulation run (Fig. 2) for the pesticide trifloxystrobin, a low volatility fungicide used to control diseases such as mildew and blight in flower bulb growing. In the left panel, a field where application takes place is shown and the eight neighboring study homes. In the right panel, the wind rose, is displayed for each of the seven days after application. These show the frequency of the occurrence of a certain wind direction (degrees) and wind velocity (m/s). Immediately after spraying, volatilization from the field starts and is modelled during the next seven days. Spray drift takes place only during spraying, at Day 1.

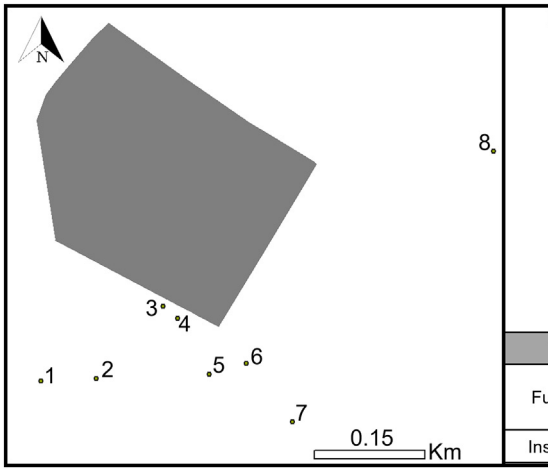
3.2.1. Outdoor and indoor air

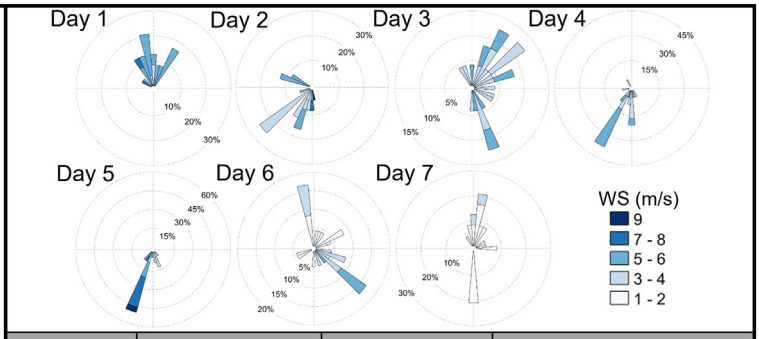
We can see from the modelled concentration in outdoor and indoor air (Fig. 3) that homes downwind of application (homes 1 to 4) are exposed to higher concentrations than other homes during the first hour (when spraying occurs), with homes closer to the field, homes 3 and 4, showing the highest values in outdoor air, 38 and 29 ng/m^3, respectively.

On Day 2, the wind direction shifted and blew mainly from the South-West (Right Panel – Fig. 2), resulting in an increase of concentration for home 8 (average 0.2 ng/m^3, Day 2) and almost no exposure near the other homes. Air concentrations vary within the one-week period, by a factor of six between the lowest and highest modelled concentrations. We can see also that modelled concentrations in outdoor and indoor air are very similar.

3.2.2. Spray drift and volatilization

From the modelled trifloxystrobin data we can infer what would be the most relevant exposure route: spray drift or evaporation of active ingredient deposited (afterwards). If we focus on the homes that are predominantly downwind (Left Panel – Fig. 2, Homes 1 to 4) from the treated area and take the full week into account, it can be seen that total exposure (defined





Type	Active Ingredient	Applied (kg/ha)	Vapor pressure (mPa at 20°C)
Fungicide	Trifloxystrobin	0.075	3.4E-3
	Fluopyram	0.075	1.2E-3
Insecticide	Esfenvalerate	0.01	1.2E-9

Fig. 2. Simulation 4 setup. In the left: The field in grey and the homes as dots. In the upper right: The wind direction (% of the day that was blowing from) and speed (WS – Wind speed) each day during the 7 simulated days. In the bottom right: the applied mixture of pesticides.

here as concentration × hours exposed) via volatilization is higher than the exposure via spray drift (during application). This is illustrated with an example, based on real spraying applications [Supplementary material B, Simulation 4]. In this case, taking cumulative exposure due to volatilization as the multiplication of exposed time (all 7 days) by the arithmetic mean of air concentrations, we end up with concentrations (in ng/m^3) of 20.9, 20.61, 97.2 and 75.1 for homes 1, 2, 3 and 4, respectively. Exposure due to drift, that only happens in the first hour, amounts (in ng/m^3) to 0.26, 0.27, 37.7 and 29.7 for homes 1, 2, 3 and 4, respectively. Thus, for all homes, the exposure caused by drift in the first hour is lower than the cumulative exposure due to volatilization. The result in this example also holds true if we take 7 days median (instead of mean) concentration as hourly exposure.

3.2.3. Indoor dust

In contrast with concentrations in air, indoor dust concentrations are calculated from daily averaged indoor air concentrations instead of hourly values. The modelled concentrations in dust are quite variable. Large differences occur between the concentrations on different days, with values typically varying between about 10 and 100 ng/g of dust. This variability in modelled values is solely caused by the daily variability of indoor air concentrations (Fig. 3), given that the remaining parameters from the Dustpred model are fixed. Modelled trifloxystrobin concentrations in indoor dust can be consulted in Supplementary material F. In short, homes 3 and 4 are consistently more exposed (higher concentrations in dust) than the other homes.

3.3. Testing individual model components of the framework

3.3.1. Step 1: outdoor air

3.3.1.1. Temporal variability. For several homes (28%) the explained temporal variability (R^2) in concentrations by the model was higher than 0.7. For 24% the R^2 ranged between 0.35 and 0.7 and for the remaining homes the R^2 was below 0.35 (for each individual R^2 see Table D.1 Supplementary material D). A selection of representative cases for model and measured data comparison, per pesticide and per home, is presented in Fig. 4. We can see that for some pesticides, such as fluopyram (panel B in Fig. 4) and mepanipyrim (panel E Fig. 4), most of the data points are close to the 1:1 line, indicating good agreement between measured and modelled data. However, for lower concentrations of fluopyram, the model underestimates concentrations (model < measured). For acetamiprid (panel A in Fig. 4)

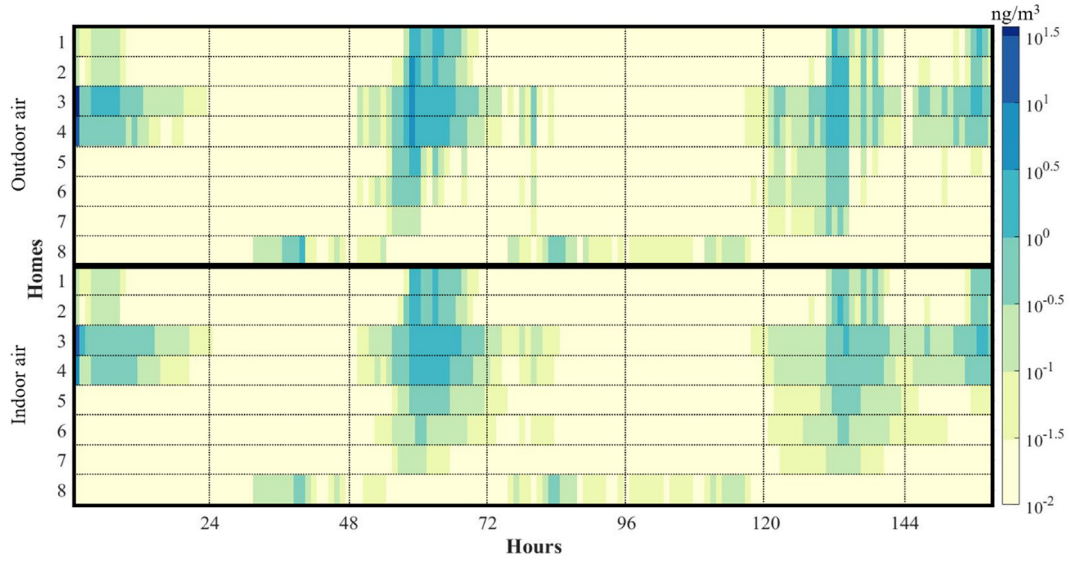


Fig. 3. Modelled Trifloxystrobin, hourly averaged, concentration in outdoor air and indoor air for each home. These are the results of simulation 4, which the setup is shown in Fig. 2. In the x axis, the hour since application (hour = 1). In the y axis the homes. The colour represents the modelled concentration in ng/m^3.

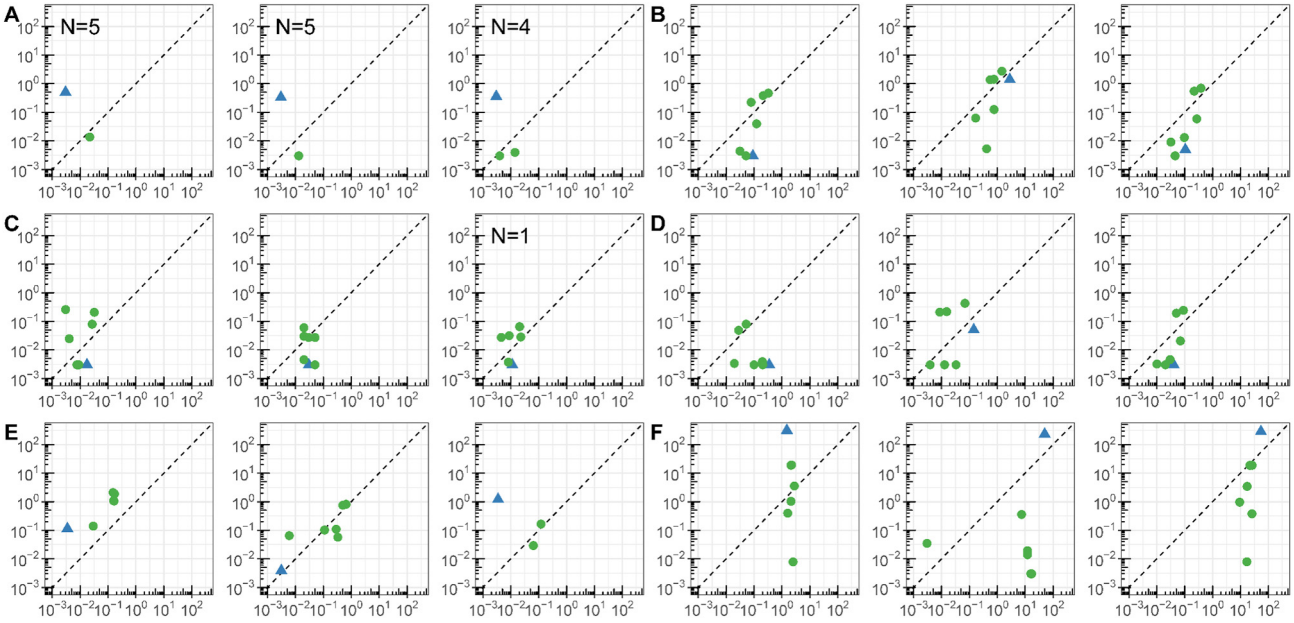


Fig. 4. Measured versus modelled outdoor air concentrations per pesticide and per home – selection of 3 examples per pesticide and only cases for which no other applications in the vicinity were done at the same time as the application on the selected field (simulation 1 to 5). Each panel corresponds to a different pesticide (see legend). Each plot is a different home. For each plot, in the x axis, the measured concentration in ng/m^3 and in the y axis, the modelled concentration in ng/m^3. Both x and y axis are in the logarithmic scale (base 10). The blue triangle is day 1, the day of spraying. The dashed black line is the 1:1 line (i.e. identity line). N = refers to the number of pairs where both the measured and modelled values are below the detection limit of the targeted pesticide (i.e. concentrations < 0.003 ng/m3). Legend: A = Acetamiprid, B = Fluopyram, C = Tebuconazole, D = Trifloxystrobin, E = Mepanipyrim, F = Chlorpropham. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

there is also good agreement, but most of the modelled and measured concentrations are below the detection limit (LOD). The exception here is an overestimation of acetamiprid concentrations in the day of spraying (day 1).

For chlorpropham (panel F in Fig. 4), the model overestimates concentrations in day 1 and underestimates concentrations for all homes in most of the consequent days. Finally, for both tebuconazole (panel C in Fig. 4) and trifloxystrobin (panel D in Fig. 4) we have a similar outcome. For some homes, most of the dots are close to the 1:1 line, whereas for other homes there is a systematic underestimation of concentrations (values below 1:1 line in panel D). All scatter plots and a detailed summary of model quality metrics calculated per pesticide and per home can be consulted in Supplementary material D.

3.3.1.2. Spatial variability. Modelled and measured data comparisons, per pesticide and per day, are presented in Fig. 5. There is a lot of variability

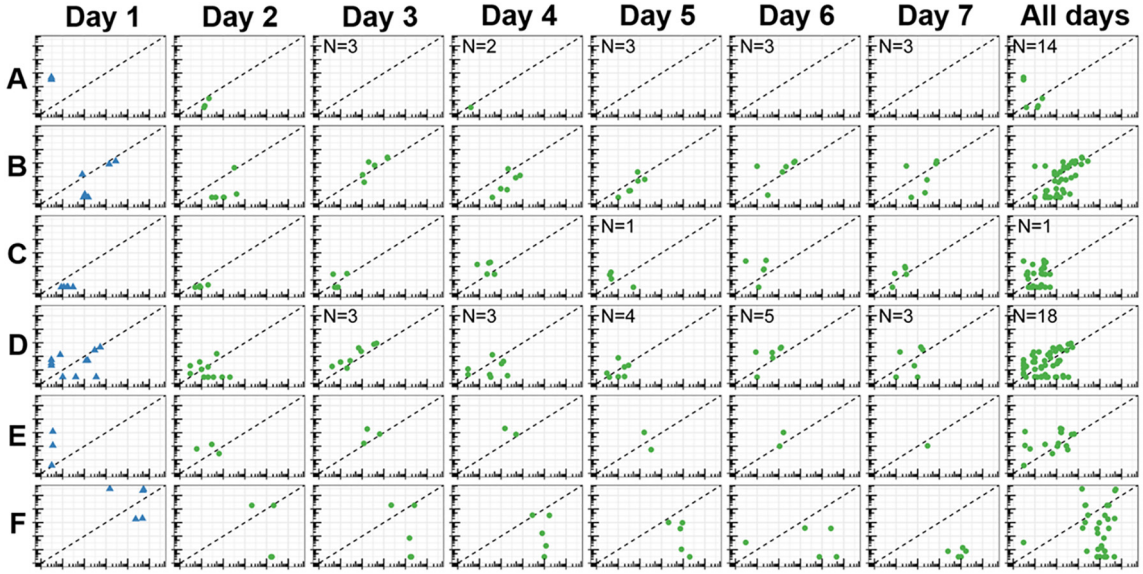


Fig. 5. Modelled vs measured concentration in outdoor air, per pesticide and per day. Each row is a different pesticide (see legend). Each column is a different day. Last column includes all days (day 1 to day 7). For each plot, in the x axis the measured concentration and in the y axis the modelled concentration. Both x and y axis are in the logarithmic scale (base 10). Each point within a plot is a different home. The dashed black line is the 1:1 line (i.e. identity line). N = refers to the number of pairs where both the measured and modelled values are below the detection limit of the targeted pesticide (concentrations < 0.003 ng/m3). Legend: A = Acetamiprid, B = Fluopyram, C = Tebuconazole, D = Trifloxystrobin, E = Mepanipyrim, F = Chlorpropham.

on model agreement between the different days, with the R^2 values ranging from 0.05 to 0.995 (see Table D.2 Supplementary material D, for all calculated R^2). There is also quite some variation in model performance between the different pesticides (A to F in Fig. 5). For acetamiprid (A in Fig. 5), we cannot draw any conclusions on model performance. Although most of the times both modelled and measured values are below the LOD, we do not know how the model performs for very low concentrations (< LOD).

For fluopyram (B in Fig. 5), the model explains between 60% to 70% of the spatial variability in concentrations for the first 5 days, but only 15% on the last two days of measurements. The model often underpredicts fluopyram concentrations when measured values are below 10^{-1} ng/m^3 (B – All days, Fig. 5). For tebuconazole and trifloxystrobin (C and D in Fig. 5, respectively) we see that the explained variance is high for some days, such as days 3 and 6. Whereas for other days the R^2 is quite low, such as days 1 and 4. For mepanipyrim (E in Fig. 5), when comparing Day 1 with all days, the model seems to perform reasonably well with the exception on day 1. Finally, for chlorpropham the model seems to explain on average 40% of the spatial variability in concentrations.

Regarding accuracy (i.e. how close the modelled value is to the measured value), the model performs well for fluopyram and trifloxystrobin. Most values are within one SD from the true mean (i.e. MAE < SD). However, for both of those pesticides (B and D in Fig. 5), the RMSE indicates that the deviation between modelled and measured values is not similar on all days (RMSE > MAE). For tebuconazole (C in Fig. 5), most of the predicted values are in the same order of magnitude as the measured values and are on average around one SD from the mean, except for days 4 and 6. The RMSE indicates that there is not much difference in the magnitude of the residuals (RMSE ≈ MAE). For both mepanipyrim and chlorpropham (E and F in Fig. 5) most of the predicted values are within the same order of magnitude as the measured values, but the average of residuals is for most cases quite high (MAE ≈ between 2 and 4 times the SD). The RMSE indicates that there is not much difference in the magnitude of the residuals (RMSE ≈ MAE).

3.3.2. Step 2: indoor air

Model performance metrics for the gComis evaluation step are presented in Table 2. Here, we can see that for 32% of pesticides the R^2 was above 0.35, while for the remaining 68% was below this value. The average R^2 was 0.3. For most cases (N = 15 pesticides) model accuracy is good, given that MAE < SD.

For the remaining 10 pesticides, MAE is 2 × SD as a maximum, except for chlorpropham and pendimethalin, where MAE is much higher than SD. For almost all cases (80%) where MAE < SD, RMSE is either lower or quasi-equal to SD. Finally, for almost all pesticides, the RMSE was slightly higher than MAE, indicating that for some homes the predicted values are further away from the measured value compared to other homes (RMSE > MAE). Here, the difference between RMSE and MAE was often lower for less volatile pesticides, such as deltamethrin, pyraclostrobin and difenoconazole.

3.3.3. Step 3: indoor dust

Results of the Dustpred evaluation step are presented in Table 3. Overall, the capacity to explain spatial variability of pesticide concentrations in indoor dust was low (average R^2 of 0.2). The model explained more than 35% of spatial variability for 4 pesticides out of the 18. Here, $R^2 > 0.75$ for difenoconazole and kresoxim-methyl. For some pesticides (N = 8) model accuracy is good, given that MAE < SD. However, for other pesticides, specially more volatile ones such as pendimethalin, s-metolachlor and chlorpropham, MAE can be more than 1 order of magnitude higher than SD. Like the indoor air evaluation step, the difference between RMSE and MAE was often lower for less volatile pesticides, such as azoxystrobin, fludioxonil and flonicamid.

4. Discussion

In this study, we developed and tested a new modelling framework to estimate residents' exposure to pesticides resulting from boom sprayer applications. Results show the suitability of the framework to estimate concentrations in outdoor and indoor air for different pesticide mixtures and meteorological conditions. Estimating concentrations in indoor dust remains challenging.

Table 2
Estimating indoor air concentrations - model efficiency per pesticide. Pesticides are ordered by vapor pressure (higher to lower).

Pesticide	Paired-N	Measured		Model efficiency		
		Mean	SD	R^2	MAE	RMSE
Propamocarb	4	0.513	0.734	0.968	0.259	0.453
Chlorpropham	16	5.251	6.425	0.018	21.45	37.781
S-Metolachlor	16	0.531	1.045	0.032	0.944	1.396
Fluopyram-benzamide	13	0.056	0.059	0.69	0.103	0.158
Pendimethalin	16	1.49	1.865	0.006	7.496	10.059
Dimethenamid-P	14	0.092	0.075	0.294	0.127	0.208
Tolclofos-methyl	15	1.067	2.758	0.152	0.305	0.523
Prochloraz	12	0.06	0.1	0.348	0.011	0.02
Carbendazim	13	0.372	0.67	0.223	0.176	0.359
Linuron	14	0.027	0.027	0.483	0.047	0.09
Mepanipyrim	6	0.062	0.077	<0.01	0.047	0.088
Trifloxystrobin	6	0.009	0.006	0.155	0.01	0.011
Trifloxystrobin-acid	4	0.073	0.106	0.814	0.173	0.213
Kresoxim-methyl	9	0.062	0.059	0.433	0.042	0.06
Tebuconazole	9	0.132	0.171	0.088	0.1	0.171
Fluopyram	11	0.155	0.26	0.476	0.409	0.803
Flonicamid	10	0.077	0.058	0.251	0.149	0.268
Metamitron	8	0.17	0.216	0.051	0.137	0.233
Boscalid	9	0.031	0.019	0.009	0.015	0.023
Prothioconazole-desthio	14	0.066	0.084	0.261	0.078	0.095
Lambda-cyhalotrin	3	0.079	0.048	0.356	0.035	0.05
Difenoconazole	5	0.075	0.068	0.047	0.086	0.104
Pyraclostrobin	13	0.112	0.217	0.14	0.021	0.03
Deltamethrin	6	0.015	0.009	0.168	0.007	0.009
Azoxystrobin	7	0.069	0.108	<0.01	0.029	0.061

Paired-N – Number of modelled and measured paired values, for the homes with quantifiable samples for the given pesticides (first column). SD – Standard deviation (ng/m^3); R^2 – coefficient of determination (−); MAE – Mean absolute error (ng/m^3); RMSE – Root mean square error (ng/m^3).

Table 3
Estimating indoor dust concentrations - model efficiency per pesticide. Pesticides are ordered by vapor pressure (higher to lower).

Pesticide	Paired-N	Measured		Model efficiency		
		Mean	SD	R^2	MAE	RMSE
Chlorpropham	9	104	84	0.015	580	1272
S-Metolachlor	5	12	8	0.572	1305	2187
Pendimethalin	27	33	52	0.039	852	1453
Tolclofos-methyl	9	20	14	0.011	18	22
Prochloraz	27	23	29	0.009	24	38
Carbendazim	27	157	140	0.002	134	190
Linuron	6	8	3	0.325	6	9
Kresoxim-methyl	8	29	44	0.769	7	9
Tebuconazole	27	20	30	0.05	13	21
Fluopyram	27	4	9	0.178	15	21
Flonicamid	27	4	7	0.077	7	12
Metamitron	6	75	99	0.149	140	282
Boscalid	27	26	35	0.158	17	27
Prothioconazole-desthio	27	6	12	0.001	23	27
Fludioxonil	4	10	10	0.439	28	32
Difenoconazole	5	13	12	0.911	25	32
Pyraclostrobin	27	44	50	0.04	45	63
Azoxystrobin	12	6	10	0.04	14	22

Paired-N – Number of modelled and measured paired values, for the homes with quantifiable samples for the given pesticides (first column). SD – Standard deviation (ng/m^3); R^2 – coefficient of determination (−); MAE – Mean absolute error (ng/m^3); RMSE – Root mean square error (ng/m^3).

4.1. Atmospheric transport and dispersion of pesticides

In the first step of the framework evaluation we explained a large portion of the variance in outside air concentrations due to vapor drift during application and vaporization after application. The model seems to tackle well both spatial and temporal variability (day to day variations) for some pesticides, such as fluopyram and mepanipyrim. For other pesticides, like chlorpropham, the explained spatial variance was lower (~40%). Unexplained variance might be related to factors that were not included in the modelling chain, such as: processes on the plant surface, such as phototransformation, penetration into the plant tissue and wash-off; the effect of the formulation of the pesticide on the vapor pressure of the pesticide, pesticide volatilization from fields planted and sprayed in the periods before or during our study; the influence of obstacles (e.g. built environment); particle-bound pesticides that travel long distances (Sanusi et al., 1999); emissions related to pesticide use in bulb disinfection in the area (Brouwer et al., 1994) or residential use (Deziel et al., 2017).

However, we hypothesize that from these, effects on volatilization from fields and bulb disinfection are the most likely factors influencing air concentration differences between homes at local scale (<250 m) in our setting. Bulb disinfection contribution is also postulated by Figueiredo et al. (2021c) who focused on pesticide concentrations in indoor dust. The other factors are more likely contributors to the overall background of pesticide concentrations (Degrendele et al., 2016) and will hardly affect variance in this study. Hence, most of these compounds will show small concentration gradients outside source areas and consequently will not show large differences in concentration between homes located relatively close to each other (few hundred meters).

Regarding accuracy, the model seems to overall perform reasonably well. Interestingly, within the pesticides sprayed, model accuracy was lower for the more volatile ones, such as chlorpropham. But as mentioned above, this could be related to bulb disinfection in the area. Larger residuals for this group might be explained by many of the aforementioned factors that have direct influence on the volatilization processes and therefore have greater effect on the calculations for more volatile pesticides.

4.2. Exchange of pesticides between outside and inside air

In the second evaluation step, going from concentrations in outdoor to indoor air, we explained more than 40% of the variance for 8 different pesticides concentrations in the homes. Figueiredo et al. (2021b) showed that, in the studied locations, measured outdoor and indoor concentrations correlated moderately. Therefore, it does not come as a surprise that for several other pesticides we cannot explain more than 30% of the variability in indoor air concentrations between homes. Additionally, some unexplained variability is to be expected, given that not all indoor sources and sinks of pesticides were accounted for. As a source, resuspension of particle-attached pesticides should be included in gComis model and as a sink (i.e. indoor loss processes) deposition and absorption to surfaces, such as walls (Wei et al., 2019) should also be included.

Independently of the above missing sources and sinks, model accuracy was good for nearly all pesticides, with predicted concentrations often lower than one SD from the true mean. A few large deviations were found between modelled and measured concentrations, that could not be explained. However, one hypothesis here is that our model is not predicting the total measured fraction. Our measured samples include pesticides in the gas-phase and absorbed to particles smaller than 10 μm. It is possible that the difference between measured and modelled could be explained by the contribution of particle-attached pesticides that were already present indoors. Apart from those, almost all modelled concentrations were in the same order of magnitude as the ones measured.

In a recent study, Pelletier et al. (2017) showed that exposure to semi-volatile organic compounds was mainly driven by indoor concentrations. Therefore, an increase in the precision of estimates of concentration inside homes will reduce the uncertainty in exposure estimates of persons. Here, we show that modelling this step is relevant because concentrations were not equal between indoor and outdoor. The differences varied per pesticide. This step becomes especially important for exposure routes that are largely influenced by indoor air concentrations, such as inhalation and dermal skin uptake (Shi et al., 2014). Modelling indoor air concentrations might be less relevant for areas with no local sources, where concentrations are more or less constant and both outdoor and indoor concentrations are equal to background levels.

To the best of our knowledge this is the first time a ventilation model is integrated in an approach to estimate concentrations of pesticides in indoor air. We see that the use of this model is important when estimating quantitative levels, and it is more relevant when looking at a small time-windows of exposure (i.e. finer time resolution), when balance between both environments is not yet reached. This phenomenon is shown in other studies (e.g. Table 3 - Raeppel et al., 2016). Nevertheless, for epidemiological purposes where relative ranking is the norm, using outdoor air concentrations as a proxy for indoor concentrations could still be valid. This may be concluded from comparisons done between indoor and outdoor concentrations (Figueiredo et al., 2021b) as well as the model simulations done in our study.

4.3. Estimating concentrations in indoor dust

In the last evaluation step, when comparing the modelled and measured data, the model could explain only a very small part of the spatial variability in the measured concentrations in VFD. Contrarily from the previous two steps, explained variability was only high for two pesticides, difenoconazole and kresoxim-methyl. Model accuracy was also not as good as the previous two evaluation steps, especially for more volatile pesticides. We found that the main advantage of the model is its simplicity and limited number of input parameters when compared to other more complex models for semi-volatile compounds such as the ones presented by Liang et al. (2019) or Wei et al. (2019).

Concentrations in indoor dust are largely driven by the dust-air partition coefficient (used in the Dustpred model). There are however other factors that influence the pesticide presence in VFD. These were not considered due to lack of available data. These include the half-life of the pesticide in the indoor environment (Li et al., 2019), which is known to be quite variable and the influence of the take-home pathway (i.e. pesticides in clothing, shoes and brought by pets) (Teysseire et al., 2020). The relevance of the latter process is still an unknown. However, by choosing VFD over dust from doormats as evaluation, we minimized the influence of the take home pathway, assuming that most particles get trapped in the doormat.

The main limitation however is not related to these factors, but to the complexity of the dust matrix. Pesticide levels in indoor dust are not just a reflection of current nearby applications but also i) applications done in the past and ii) pesticide use in other areas and transported through air across longer distances (Fuhrimann et al., 2020). These will eventually deposit (settle) indoors (Quirós-Alcalá et al., 2011) and accumulate in indoor dust (Rull and Ritz, 2003; Rothlein et al., 2006).

The influence of past applications and degradation on pesticides present in the indoor environment remains one of the most challenging problems. It is key to better understand lifetime exposure of residents via the dermal pathway and inhalation of small contaminated particles.

4.4. Estimating residents' exposure to pesticides

Several studies have shown that during spraying, pesticides will drift outside the application area (e.g. Coronado et al., 2011, van de Zande et al., 2012). However, the observation that volatilization leads to detectable concentrations, even seven days after application, also observed by van den Berg et al. (1995), emphasizes the need to include this route in exposure studies. At the same time, it illustrates the necessity to study differences in exposure resulting from primary drift following application and the exposure resulting from volatilization lasting for days (and perhaps much longer for some pesticides). The latter may as such be a larger contributor to residential exposure than droplet drift during application. This

will depend on the pesticide persistence in the environment (Socorro et al., 2016) and physicochemical properties, notably the potential to volatilize. As we have seen in the example provided here, there is a possibility that long-term cumulative exposure to volatilization is higher than cumulative short-term exposures to drift. In a future study we will investigate differences in exposure resulting from primary and secondary drift more systematically by including more pesticides, simulate for more days, multiple applications and different climate scenarios.

The above is an indication that both routes may at least be equally important in evaluation frameworks. Regulation however is focused on reducing spray drift as a means to reduce exposure of surface water and ground level close to fields. Volatilization is not directly affected by these regulations. Therefore, to limit the contribution of volatilization, we would need to reduce the amount sprayed/used or/and use pesticide that degrade faster in the environment, always taking into account possible transformation products. The modelling framework can support the development of guidelines considering not just spray drift but also volatilization (e.g. Boesten et al., 2021).

In general, the modelling framework seems capable of simulating spray drift, vaporization and atmospheric transport and dispersion of different pesticide mixtures for different meteorological conditions reasonably well. The framework explained about 30% to 95% of the temporal and spatial variability of air concentrations, respectively. Environmental concentration estimates given here are likely a better exposure proxy and closer to reality than earlier exposure proxies used for health studies, such as buffers of agricultural fields surrounding homes (e.g. Ward et al., 2006), proximity to fields (e.g. Bukalasa et al., 2017), remote sensing (e.g. Wan, 2015) and many others. These exposure proxies do not include the impact of meteorological conditions and physico-chemical properties.

The results of the first two evaluation steps (i.e. estimating outdoor and indoor air concentrations) show moderate to good accuracy of the model for most pesticides, except chlorpropham. This part of the framework currently can be used for estimating exposure of residents to pesticides. This may be done for individual pesticides or mixtures and can include single or multiple fields, depending on existing spraying applications and availability of input data. However, estimating concentrations of pesticides in indoor dust proved to be difficult. This could be related to the adequacy of the Dustpred model for the situations we tried to simulate here. It could be that accumulation of pesticides in dust from previous applications or applications further away plays a large role. So, at this stage, evaluation showed weaknesses in the model for this step and the Dustpred model should only be used for pesticides that degrade rapidly in the environment, thus not being affected by “historical” use, residential use (Meftaul et al., 2020) and long-range transport.

4.5. Modelling framework - strengths and limitations

Our modelling framework has several strengths. Firstly, it was developed by collecting already built and verified models and each step was evaluated independently, therefore avoiding possible errors propagated between models. Secondly, the fact that the models are connected solely by input and output gives a great flexibility for improvement and adjustments in future research, as well as for running only parts of the framework if needed. Finally, simplifications can be done in all models to reduce the number of input parameters, adjust resolution (i.e. m to km) and use the model to estimate pesticide exposures for both local and national scales.

Regarding the use of the framework, the model with the largest limitation is the Dustpred model. It is likely related to the inherent complexity of the dust matrix and all factors influencing concentrations in this medium. Nevertheless, as dust can be an important exposure source, additional steps need to be taken to improve indoor dust models through either deterministic or empirical modelling. We know that improvement may be achieved by adding information regarding historical pesticide use, residential use (Glorennec et al., 2017) and the direct influence of indoor sources and sinks, as mentioned by Sukiene et al. (2017), when it becomes available. One of the main problems to also tackle is the take-home pathway (such as drag) (Figueiredo et al., 2021c).

Other limitations of the framework are related to the volatilization model. In the current version of this model, the effect of the formulation on the behavior of the pesticide on the plant surface is not taken into account. To remedy this, model concepts for the description of this effect would have to be developed and tested.

In many cases, the required model input is incomplete. Missing values need to be estimated (e.g. Leistra, 2011) or obtained from measurements published in the scientific literature or reported in EFSA peer reviews of the active substances of the plant protection product. However, there can be uncertainty in the value for a substance property, such as the vapor pressure, as sometimes different values are reported in the literature. In addition, the lack of information of the effect of the formulation on the actual vapor pressure adds to this uncertainty. It is important to establish what are possible ranges for the input parameters, so as to know how much uncertainty there is in the model chain. The temporal and or spatial resolution of input data, such as meteorological data, needs also further attention as the impact of some variables may not be possible to assess when using data at lower resolution. A sensitivity analysis considering all relevant input variables at different spatial or temporal resolution would help to identify the scale at which these variables should be measured preferably and to define the parts of the model that need further improvement. These steps will be included in future OBOmod iterations.

The use of the model framework depends also on the availability of relevant data. Inclusion of data on these properties, e.g. on other relevant processes on the plant surface, such as penetration of the substance into the plant tissue, photo-transformation and wash-off in the dossiers submitted for registration in the EU would be needed to further improve the assessment of the exposure of residents as a result of agricultural use of pesticides. In a next step, dedicated field experiments on the emission and atmospheric transport of agricultural pesticides are needed to evaluate the improvement of the OBO model framework using the improved set of input values. Also, uncertainty (individual and propagation) as well as sensitivity analysis on certain input parameters will be studied. This hopefully leads to further improvements of the OBOmod.

5. Concluding remarks

An integrated framework based on different existing deterministic models is used to estimate residents' exposure to pesticides.

We estimated the exposure to pesticides in homes near agricultural sites using models describing air-borne drift, volatilization, atmospheric transport and dispersion, exchange between outdoor and indoor air in residential areas close to treated fields.

From the comparison between modelled and measured concentrations in air we conclude that, in general, the predicted 24-h exposure concentrations of residents to pesticides in air were in the same order of magnitude as those measured. Some studies showed that house dust seems to be an important exposure route. However, predictions of concentrations in this medium remain difficult. Another important finding is that especially for the volatile compounds considered in this study, the cumulative exposure due to volatilization after application may be larger than exposure to droplet drift during application.

The framework can be used in local settings at a range of a few kilometres away from the source to quantitatively estimate exposure via air. The framework can be used for different purposes. It can be used to link to different health outcomes and improve epidemiological studies. It can be used to help public health policy makers by simulating worst case scenarios or integrate with toxicology data to allow for a more complete assessment of human health risk from pesticides. Finally, it can also be used to quantify relative contributions from exposure pathways, and so support development of regulations (e.g. quantity applied) regarding pesticide application.

CRediT authorship contribution statement

Daniel Figueiredo: Conceptualization, Methodology, Validation, Formal analysis, Software, Visualization, Supervision, Data curation, Writing - original draft, Writing - review & editing, Formal analysis, Investigation. **Jan Duyzer**: Conceptualization, Methodology, Resources, Supervision, Writing - Review & Editing. **Cor Jacobs**: Software, Conceptualization, Methodology, Writing - Review & Editing. **Henk Jan Holterman**: Software, Conceptualization, Methodology, Writing - Review & Editing. **Esmeralda Krop**: Conceptualization, Methodology, Data curation, Supervision, Writing - Review & Editing, Project administration. **Jan C. van de Zande**: Conceptualization, Methodology, Resources, Writing - Review & Editing. **Frederik van den Berg**: Conceptualization, Methodology, Resources, Writing - Review & Editing. **Daan Buijtenhuijs**: Investigation, Writing - Review & Editing. **Gooijer, Y.M.**: Investigation, Writing - Review & Editing. **Luuk Lageschaar**: Investigation, Writing - Review & Editing. **Anke Huss**: Conceptualization, Writing - Review & Editing. **Roel Vermeulen**: Conceptualization, Methodology, Resources, Writing - Review & Editing, Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This study was funded by the Dutch Ministry of Infrastructure and Water Management and the Ministry of Economic Afairs & Climate Policy. This work was commissioned by the Dutch National Institute for Public Health and the Environment (RIVM). We would like to thank all participants and growers for their willingness to participate in this study, the stakeholders for continuous feedback given and all the parties involved in the OBO study group.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.153798.

References

Atabila, A., Phung, D.T., Hogarh, J.N., Osei-Fosu, P., Sadler, R., Connell, D., Chu, C., 2017. Dermal exposure of applicators to chlorpyrifos on rice farms in Ghana. Chemosphere 178, 350–358. https://doi.org/10.1016/j.chemosphere.2017.03.062.

Boesten, J.J.T.I., Adriaanse, P.I., Holterman, H.J., ter Horst, M.M.S., Tiktak, A., van der Zande, J.C., Wipfler, L., 2021. Scenarios for exposure of aquatic organisms to plant protection products in the Netherlands: part 2: sideways and upward spraying in Dutch fruit crops (final report). Wageningen Environmental Research report; No. 3100. Wageningen Environmental Research https://doi.org/10.18174/549658.

Borchiellini, R., Furbringer, J., 1999. An evaluation exercise of a multizone air flow model. Energy and Buildings 30 (1), 35–51.

Bradman, A., Salvatore, A., Boeniger, M., et al., 2009. Community-based intervention to reduce pesticide exposure to farmworkers and potential take-home exposure to their families. J. Expo. Sci. Environ. Epidemiol. 19, 79–89. https://doi.org/10.1038/jes.2008.18.

Brouwer, D.H., Brouwer, E.J., van Hemmen, J.J., 1994. Estimation of long-term exposure to pesticides. Am. J. Ind. Med. 25 (4), 573–588. https://doi.org/10.1002/ajim.4700250411.

Bukalasa, J.S., Brunekreef, B., Brouwer, M., Vermeulen, R., de Jongste, J.C., van Rossem, L., Vonk, J.M., Wijga, A., Huss, A., Gehring, U., 2017. Proximity to agricultural fields as proxy for environmental exposure to pesticides among children: the PIAMA birth cohort. Sci. Total Environ. 595, 515–520. https://doi.org/10.1016/j.scitotenv.2017.03.269.

Butler Ellis, M.C., van de Zande, J.C., van den Berg, F., Kennedy, M.C., O'Sullivan, C.M., Jacobs, C.M., Fragkoulis, G., Spanoghe, P., Gerritsen-Ebben, R., Frewer, L.J., Charistou, A., 2017. The BROWSE model for predicting exposures of residents and bystanders to agricultural use of plant protection products: an overview. Biosyst. Eng. 154, 92–104. https://doi.org/10.1016/j.biosystemseng.2016.08.017.

Butte, W., 2004. Sources and impacts of pesticides in indoor environments. Handbook of Environmental Chemistry. 4, pp. 89–116. https://doi.org/10.1007/b94832.

Calliera, M., Luzzani, G., Sacchettini, G., Capri, E., 2019. Residents perceptions of non-dietary pesticide exposure risk. Knowledge gaps and challenges for targeted awareness-raising material in Italy. Sci. Total Environ. 685 (June), 775–785. https://doi.org/10.1016/j.scitotenv.2019.06.223.

Cornelis, C., Schoeters, G., Kellen, E., Buntinx, F., Zeegers, M., 2009. Development of a GIS-based indicator for environmental pesticide exposure and its application to a Belgian case–control study on bladder cancer. Int. J. Hyg. Environ. Health 212 (2), 172–185. https://doi.org/10.1016/j.ijheh.2008.06.001.

Coronado, G.D., Livaudais, J., Hanisch, R., Tekeste, T., 2011. Take-home route of pesticide exposure. Encyclopedia of Environmental Health. 1, pp. 312–324. https://doi.org/10.1016/B978-0-444-52272-6.00641-3.

Curwin, B.D., Hein, M.J., Sanderson, W.T., Nishioka, M.G., Reynolds, S.J., Ward, E.M., Alavanja, M.C., 2005. Pesticide contamination inside farm and nonfarm homes. J. Occup. Environ. Hyg. 2 (7), 357–367. https://doi.org/10.1080/15459620591001606.

Degrendele, C., Okonski, K., Melymuk, L., Landlová, L., Kukučka, P., Audy, O., Klánová, J., 2016. Pesticides in the atmosphere: a comparison of gas-particle partitioning and particle size distribution of legacy and current-use pesticides. Atmos. Chem. Phys. 16 (3), 1531–1544. https://doi.org/10.5194/acp-16-1531-2016.

Dereumeaux, C., Fillol, C., Quenel, P., Denys, S., 2020. Pesticide exposures for residents living close to agricultural lands: a review. Environ. Int. 134 (November 2019), 105210. https://doi.org/10.1016/j.envint.2019.105210.

Deziel, N.C., Beane Freeman, L.E., Graubard, B.I., Jones, R.R., Hoppin, J.A., Thomas, K., Friesen, M.C., 2017. Relative contributions of agricultural drift, Para-occupational, and residential use exposure pathways to house dust pesticide concentrations: meta-regression of published data. Environ. Health Perspect. 125 (3), 296–305. https://doi.org/10.1289/EHP426.

EFSA (European Food Safety Authority), 2014. Guidance on the assessment of exposure of operators, workers, residents and bystanders in risk assessment for plant protection products. EFSA Journal 12 (10), 3874. https://doi.org/10.2903/j.efsa.2014.3874 55 pp.12.

Falette, N., Kiss, A., Bayle, M.-L., Bouaoun, L., Scalbert, A., Fervers, B., 2018. Seasonal variations of exposure to agricultural pesticides in residents proximate to vineyards: SIGEXPOSOME study. Rev. Epidemiol. Sante Publique 66, S340. https://doi.org/10.1016/j.respe.2018.05.279.

Feustel, H.E., Raynor-Hooson, A., 1990. COMIS Fundamentals. Air Infiltration and Ventilation Centre, Technical Note 29, Lawrence Berkeley Laboratory Report, LBL-28560. https://www.aceee.org/files/proceedings/1998/data/papers/0509.PDF.

Figueiredo, D.M., Vermeulen, R.C., Duyzer, J., 2018. An integrated modelling framework to estimate residents exposure to pesticides from boom sprayer applications. ISEE Young Conference https://doi.org/10.1136/oemed-2018-iseeabstracts.84.

Figueiredo, D.M., Nijssen, R., Krop, E., Buijtenhuijs, D., Duyzer, J., Huss, A., Mol, H., Vermeulen, R.C.H., 2019. Pesticides in indoor dust – a possible important route for residents' exposure. Environ. Epidemiol. 3 (October 2019), 122. https://doi.org/10.1097/01.EE9.0000607072.50712.0a.

Figueiredo, D.M., Krop, E.J., Duyzer, J., Gerritsen-Ebben, R.M., Gooijer, Y.M., Holterman, H.J., Huss, A., Jacobs, C.M., Kivits, C.M., Kruijne, R., Mol, H.J., Oerlemans, A., Sauer, P.J., Scheepers, P.T., van de Zande, J.C., van den Berg, F., Wenneker, M., Vermeulen, R.C., 2021a. Research on exposure of residents to pesticides in the Netherlands: study protocol (preprint). JMIR Research Protocols. 10. https://doi.org/10.2196/27883.

Figueiredo, D.M., Duyzer, J., Huss, A., Krop, E.J.M., Gooijer, Y., Vermeulen, R.C.H., 2021b. Spatio-temporal variation of outdoor and indoor pesticide air concentrations in homes near agricultural fields. Atmos. Environ. 262 (June), 118612. https://doi.org/10.1016/j.atmosenv.2021.118612.

Figueiredo, D.M., Nijssen, R., Krop, E.J.M., Buijtenhuijs, D., Gooijer, Y., Lageschaar, L., Duyzer, J., Huss, A., Mol, H., Vermeulen, R.C.H., 2021. Pesticides in Doormat and Floor Dust from Homes Close to Treated Fields: Spatio-temporal Variance and Determinants of Occurrence and Concentrations. https://doi.org/10.2139/ssrn.3951730.

Fuhrimann, S., Klánová, J., Přibylová, P., Kohoutek, J., Dalvie, M.A., Röösli, M., Degrendele, C., 2020. Qualitative assessment of 27 current-use pesticides in air at 20 sampling sites across Africa. Chemosphere 258, 127–333. https://doi.org/10.1016/j.chemosphere.2020.127333.

Fujimoto, A., Satow, T., Kishimoto, T., 2016. Simulation of spray distribution with boom sprayer considering effect of wind for agricultural cloud computing analysis. Engineering in Agriculture, Environment and Food 9 (4), 305–310. https://doi.org/10.1016/j.eaef.2016.04.001.

Gibbs, J.L., Yost, M.G., Negrete, M., Fenske, R.A., 2017. Passive sampling for indoor and outdoor exposures to chlorpyrifos, azinphos-methyl, and oxygen analogs in a rural agricultural community. Environ. Health Perspect. 125, 333–341. https://doi.org/10.1289/ehp425.

Glorennec, P., Serrano, T., Fravallo, M., Warembourg, C., Monfort, C., Cordier, S., Chevrier, C., 2017. Determinants of children's exposure to pyrethroid insecticides in western France. Environ. Int. 104 (April), 76–82. https://doi.org/10.1016/j.envint.2017.04.007.

Health Council of the Netherlands, 2014. Crop Protection and Local Residents. Health Council of the Netherlands, The Hague publication no. 2014/02E.

Holterman, H.J., van de Zande, J.C., 2010. The Cascade Drift Model: First results of a realistic study on regional pesticide deposition. International advances in pesticide application 99, 367–374.. https://edepot.wur.nl/169059.

Holterman, H.J., van de Zande, J.C., 2019. Spray drift simulations to estimate the exposure of residents to pesticides close to a sprayed field. Wageningen Research, Report WPR-878 https://doi.org/10.18174/467149.

Holterman, H.J., van de Zande, J.C., Porskamp, H.A.J., Huijsmans, J.F.M., 1997. Modelling spray drift from boom sprayers. Comput. Electron. Agric. 19 (1997), 1–22.

Holterman, H.J., Michielsen, J.M.G.P., Van de Zande, J.C., 1998. Spray drift in crop protection: validation and usage of a drift model. Amsterdam Centre for European Studies Research Paper No. 2021/03, Available at SSRN:Paper Presented at the International Conference on Agricultural Engineering (AgEng), Oslo (Norway), August 24-27, 1998, Paper No. 98-A-012, 9 pp.

Langenbach, T., de Campos, T.M.P., Caldas, L.Q., 2021. Why Airborne Pesticides Are So Dangerous [Online First]. IntechOpen https://doi.org/10.5772/intechopen.95581.

Leistra, M., 2011. Methods for estimating the vapour pressure of organic chemicals. Report 2215. Alterra, Wageningen, the Netherlands.
Leistra, M., van den Berg, F., 2007. Volatilization of parathion and chlorothalonil from a potato crop simulated by the PEARL model. Environ. Sci. Technol. 41, 2243–2248. https://doi.org/10.1021/es0627242.
Leistra, M., Wolters, A., 2004. Computations on the volatilisation of the fungicide fenpropimorph from plants in a wind tunnel. Water Air Soil Pollut. 157, 133–148. https://doi.org/10.1023/B:WATE.0000038883.86688.83.
Leistra, M., Smelt, J.H., van den Berg, F., 2005. Measured and computed volatilisation of the fungicide fenpropimorph from a sugar beet crop. Pest Manag. Sci. 61, 151–158. https://doi.org/10.1002/ps.964.
Lewis, K.A., Tzilivakis, J., Warner, D., Green, A., 2016. An international database for pesticide risk assessments and management. Hum. Ecol. Risk Assess. Int. J. 22 (4), 1050–1064. https://doi.org/10.1080/10807039.2015.1133242.
Li, L., Qiu, Y., Gustafsson, Å., Krais, A.M., Weiss, J.M., Lundh, T., Bergman, Å., 2019. Characterization of residential household dust from Shanghai by particle size and analysis of organophosphorus flame retardants and metals. Environ. Sci. Eur. 31 (1). https://doi.org/10.1186/s12302-019-0279-9.
Liang, Y., Bi, C., Wang, X., Xu, Y., 2019. A general mechanistic model for predicting the fate and transport of phthalates in indoor environments. Indoor Air 29 (1), 55–69. https://doi.org/10.1111/ina.12514.
Meftaul, I.M., Venkateswarlu, K., Dharmarajan, R., Annamalai, P., Megharaj, M., 2020. Pesticides in the urban environment: a potential threat that knocks at the door. Sci. Total Environ. 711, 134612. https://doi.org/10.1016/j.scitotenv.2019.134612.
Oates, L., Cohen, M., 2011. Assessing diet as a modifiable risk factor for pesticide exposure. Int. J. Environ. Res. Public Health 8 (6), 1792–1804. https://doi.org/10.3390/ijerph8061792.
OBO, 2019. Research on exposure of residents to pesticides in the Netherlands: OBO flower bulbs. https://www.rijksoverheid.nl/binaries/rijksoverheid/documenten/rapporten/2019/04/10/bijlage-1-onderzoeksrapport-obo/bijlage-1-onderzoeksrapport-obo.pdf.
Pelletier, M., Bonvallot, N., Ramalho, O., Mandin, C., Wei, W., Raffy, G., Glorennec, P., 2017. Indoor residential exposure to semi-volatile organic compounds in France. Environ. Int. 109 (September), 81–88. https://doi.org/10.1016/j.envint.2017.08.024.
Phaff, J.C., 1996. Final Report Annex 23 - Multizone Ventilation Models: Participation of TNO Bouw. Examples. Report 96-BB1-R1086. TNO.
Pubmed, 2021. https://pubmed.ncbi.nlm.nih.gov/?term=pesticide+exposure+resident [accessed on 30-09-2021].
Quirós-Alcalá, L., Bradman, A., Nishioka, M., Harnly, M.E., Hubbard, A., Mckone, T.E., Ferber, J., Eskenazi, B., 2011. Pesticides in house dust from urban and farmworker households in California: an observational measurement study. Environ. Health (2011) https://doi.org/10.1186/1476-069X-10-19.
Raeppel, C., Salquèbre, G., Millet, M., Appenzeller, B.M.R., 2016. Pesticide detection in air samples from contrasted houses and in their inhabitants' hair. Sci. Total Environ. 544, 845–852. https://doi.org/10.1016/j.scitotenv.2015.12.020.
Rothlein, J., Rohlman, D., Lasarev, M., Phillips, J., Muniz, J., McCauley, Linda, 2006. Organophosphate pesticide exposure and neurobehavioral performance in agricultural and non-agricultural Hispanic workers. Environ. Health Perspect., 114:5 https://doi.org/10.1289/ehp.8182.
Rull, R.P., Ritz, B., 2003. Historical pesticide exposure in California using pesticide use reports and land-use surveys: an assessment of misclassification error and bias. Environ. Health Perspect. 111 (13), 1582–1589. https://doi.org/10.1289/ehp.6118.
Sangiorgi, G., Ferrero, L., Ferrini, B.S., Lo Porto, C., Perrone, M.G., Zangrando, R., Bolzacchini, E., 2013. Indoor airborne particle sources and semi-volatile partitioning effect of outdoor fine PM in offices. Atmos. Environ. 65, 205–214. https://doi.org/10.1016/j.atmosenv.2012.10.050.
Sanusi, A., Millet, M., Mirabel, P., Wortham, H., 1999. Gas-particle partitioning of pesticides in atmospheric samples. Atmos. Environ. 33 (29), 4941–4951. https://doi.org/10.1016/S1352-2310(99)00275-7.
Sauter, F., van Zanten, M., van der Swaluw, E., Aben, J., de Leeuw, F., van Jaarsveld, H., 2015. The OPS-model. Description of OPS 4.5.0. Bilthoven, Rijksinstituut voor Volksgezondheid en Milieu. http://www.rivm.nl/media/ops/OPS-model.pdf.
Sauter, F., Van Zanten, M., Van der Swaluw, E., Aben, J., De Leeuw, F., Van Jaarsveld, H., 2018. The OPS-Model. Description of OPS 4.5.2. RIVM (National Institute for Public Health and the Environment), Bilthoven. https://www.rivm.nl/media/ops/v4.5.2/OPS-model-v4.5.2.pdf.
Schaub, S., Huber, R., Finger, R., 2020. Tracking societal concerns on pesticides – a Google Trends analysis. Environ. Res. Lett. 15, 084049. https://doi.org/10.1088/1748-9326/ab9af5.
Shi, S., Li, Y., Zhao, B., 2014. Deposition velocity of fine and ultrafine particles onto manikin surfaces in indoor environment of different facial air speeds. Build. Environ. 81, 388–395. https://doi.org/10.1016/j.buildenv.2014.07.017.
Shmueli, G., Bruce, P.C., Stephens, M., Patel, N.R., 2016. Data Mining for Business Analytics: Concepts, Techniques, and Applications With JMP Pro. 3rd edition. Wiley.
Silva, V., Mol, H.G.J., Zomer, P., Tienstra, M., Ritsema, C.J., Geissen, V., 2019. Pesticide residues in european agricultural soils – a hidden reality unfolded. Sci. Total Environ. 653, 1532–1545. https://doi.org/10.1016/j.scitotenv.2018.10.441.
Socorro, J., Durand, A., Temime-Roussel, B., Gligorovski, S., Wortham, H., Quivet, E., 2016. (2016) the persistence of pesticides in atmospheric particulate phase: an emerging air quality issue. Sci. Rep. 6, 33456. https://doi.org/10.1038/srep33456.
Soheilifard, F., Afshin, M., Raini, M.G., Taki, M., van Zelm, R., 2020. Chemical footprint of pesticides used in citrus orchards based on canopy deposition and off-target losses. Sci. Total Environ. 732, 118–139. https://doi.org/10.1016/j.scitotenv.2020.139118.
Stallinga, H., Holterman, H.J., Michielsen, J.G.P., van Velde, P., 2008. A two-year experimental study on airborne drift using active and passive sampling. In: Alexander, L.S., Carpenter, P.I., Cooper, S.E., Glass, C.R., Andersen, P.G., Magri, B., Robinson, T.H., Stock, D., Taylor, W.A., Thornhill, E.W., Zande, J.C. (Eds.), Applied Biology. 84. Association of Applied Biologists, Warwick, UK, pp. 1–8.
Steward, B.L., Hanna, H.M., Dixon, P.M., Mompremier, R.K., 2019. Measuring and modelling the movement of spray droplets into off-target areas. 2019 ASABE Annual International Meeting, p. 1901496 https://doi.org/10.13031/aim.201901496.
Sukiene, V., von Goetz, N., Gerecke, A.C., Bakker, M.I., Delmaar, C.J., Hungerbuhler, K., 2017. Direct and air-mediated transfer of labeled SVOCs from indoor sources to dust. Environ. Sci. Technol. 51 (6), 3269–3277.
Tames, F., Miglioranza, K.S.B., Nuñez, M.R., Carreras, H., 2020. Indoor persistent organic pollutants in agricultural areas from Argentina. Indoor Air (January), 1–10 https://doi.org/10.1111/ina.12649.
Taylor, M., Lyons, S.M., Davie-Martin, C.L., Geoghegan, T.S., Hageman, K.J., 2020. Understanding trends in pesticide volatilization from agricultural fields using the pesticide loss via volatilization model. Environmental Science & Technology 54 (4), 2202–2209. https://doi.org/10.1021/acs.est.9b04762.
Teysseire, R., Manangama, G., Baldi, I., Carles, C., Brochard, P., Bedos, C., et al., 2020. Assessment of residential exposures to agricultural pesticides: a scoping review. PLoS ONE 15 (4), e0232258. https://doi.org/10.1371/journal.pone.0232258.
van den Berg, F., Bor, G., Smidt, R.A., van de Peppel-Groen, A.E., Smelt, J.H., Müller, T., Maurer, T., 1995. Volatilization of Parathion and Chlorothalonil After Spraying Onto a Potato Crop. DLO Winand Staring Centre, Wageningen 59 p.
Van den Berg, F., Kubiak, R., Benjey, W.G., Majewski, M.S., Yates, S.R., Reeves, G.L., Smelt, J.H., Van der Linden, A.M.A., 1999. Emission of pesticides into the air. Water Air Soil Pollut. 115, 195–218. https://doi.org/10.1023/A:1005234329622.
van den Berg, F.C.M.J., Jacobs, M.C., Butler-Ellis, P., Spanoghe, P., Doan Ngoc, K., Fragkoulis, G., 2016a. Modelling exposure of workers, residents and bystanders to vapour of plant protection products after application to crops. Sci. Total Environ. 573, 1010–1020. https://doi.org/10.1016/j.scitotenv.2016.08.180.
van den Berg, F., Tiktak, A., Boesten, J.J.T.I., van der Linden, A.M.A., 2016b. PEARL Model for Pesticide Behaviour and Emissions in Soil-Plant System: Description of Processes. Statutory Research Tasks Unit for Nature & the Environment, Wageningen, p. 134 (WOt-technical report 61).
Van Jaarsveld, J.A., 2004. Description and Validation of OPS-Pro 4.1, RIVM report 500045001/2004. https://www.rivm.nl/bibliotheek/rapporten/500045001.pdf.
Veludo, A.F., Figueiredo, D.M., Degrendele, C., Masinyana, L., Curchod, L., Kohoutek, J., Kukučka, P., Martiník, J., Přibylová, P., Klánová, J., Dalvie, M.A., Röösli, M., Fuhrimann, S., 2022. Seasonal variations in air concentrations of 27 organochlorine pesticides (OCPs) and 25 current-use pesticides (CUPs) across three agricultural areas of South Africa. Chemosphere 289, 133–162. https://doi.org/10.1016/j.chemosphere.2021.133162.
Waheed, S., Halsall, C., Sweetman, A.J., Jones, K.C., Malik, R.N., 2017. Pesticides contaminated dust exposure, risk diagnosis and exposure markers in occupational and residential settings of Lahore, Pakistan. Environ. Toxicol. Pharmacol. 56 (November), 375–382. https://doi.org/10.1016/j.etap.2017.11.003.
Wan, N., 2015. Pesticides exposure modeling based on GIS and remote sensing land use data. Appl. Geogr. 56, 99–106. https://doi.org/10.1016/j.apgeog.2014.11.012.
Ward, M.H., Lubin, J., Giglierano, J., Colt, J.S., Wolter, C., Bekiroglu, N., Camann, D., Hartge, P., Nuckols, J.R., 2006. Proximity to crops and residential exposure to agricultural herbicides in Iowa. Environ. Health Perspect. 114 (6), 893–897.
Wei, W., Ramalho, O., Mandin, C., 2019. A long-term dynamic model for predicting the concentration of semi-volatile organic compounds in indoor environments: application to phthalates. Build. Environ. 148 (October 2018), 11–19. https://doi.org/10.1016/j.buildenv.2018.10.044.
Weschler, C.J., Nazaroff, W.W., 2010. SVOC partitioning between the gas phase and settled dust indoors. Atmos. Environ. 44 (30), 3609–3620. https://doi.org/10.1016/j.atmosenv.2010.06.029.
Wösten, J.H.M., Veerman, G.J., De Groot, W.J.M., Stolte, J., 2001. Waterretentie- en doorlatendheidskarakteristieken van boven- en ondergronden in Nederland: de Staringreeks - Vernieuwde uitgave 2001. Alterra, Wageningen Report no. 153. 86 pp https://www.wur.nl/nl/Publicatie-details.htm?publicationId=publication-way-333133353539.
Zande, J.C.van de, Holterman, H.J., Huijsmans, J.F.M., 2012. Spray drift assessment of exposure of aquatic organisms to plant protection products in the Netherlands. Part 1: Field crops and downward spraying. 84pWageningen UR Plant Research International, Plant Research International Report 419, Wageningen . https://edepot.wur.nl/243240.
Zeitlin, Jonathan, Weimer, Maria, van der Duin, David, Kuhn, Theresa, Jensen, Martin Dybdahl, 2021. Reforming EU Pesticides Regulation, Rebuilding Public Support: Evidence From Survey Experiments in Six Member States. Amsterdam Centre for European Studies Research Paper No. 2021/03, Available at SSRN: https://doi.org/10.2139/ssrn.3862421. https://ssrn.com/abstract=3862421.
Zivan, O., Segal-Rosenheimer, M., Dubowski, Y., 2016. Airborne organophosphate pesticides drift in Mediterranean climate: the importance of secondary drift. Atmos. Environ. 127, 155–162. https://doi.org/10.1016/j.atmosenv.2015.12.003.