www.soilprom.eu

Organisation Wageningen Research
Main Authors Louise Wipfler

SOILPROM

Plan for models' upgrade and integration

Deliverable 1.4

Date: 28.04.2025
Doc. Version: 2

This project has received funding from the European Union's Horizon Europe Research and Innovation program under Grant Agreement No. 101156589. This output reflects only the author's view, and the European Union cannot be held responsible for any use that may be made of the information contained therein.

SOILPROM

Document Control Information

Settings	Value
Deliverable Title	Plan for models upgrade and integration
Work Package Title	Setting the scene for soil pollution processes modelling
Deliverable number	1.4
Description	Plan for upgrade and integration of the models used in the SOILPROM project
Lead Beneficiary	Wageningen Research
Lead Authors	Louise Wipfler (WR)
Contributors	Jan Vanderborght (FZJ), Lutz Weihermüller (FZJ), Jian Liu (NIBIO), Adam Szymkiewicz (GUT), Ingeborg Joris (VITO), Jose Alvarez Rogel (UPTC), Agatha Zamuner (WU), Bart van Stratum (WU) Jantiene Baartman (WU), Jos van Dam (WU), Mahrooz Rezaei (WU), Steven Droge (WR), Michael Deligiannis (WR), Pavan Cornelissen (WR)
Submitted by	Wageningen University
Doc. Version (Revision number)	2
Sensitivity (Security):	Public
Date:	28/04/2025

Document Approver(s) and Reviewer(s):

NOTE: All Approvers are required. Records of each approver must be maintained. All Reviewers in the list are considered required unless explicitly listed as Optional.

Name	Role	Action	Date
Lutz Weihermüller (FZJ)	Reviewer	Reviewed (partly)	14/4/2025
Nick Jarvis (SLU)	External Reviewer	Reviewed	13/4/2025
Yonggen Zhang (TJU)	External Reviewer	reviewed	24/4/2025

Document history:

The Document Author is authorized to make the following types of changes to the document without requiring that the document be re-approved:

- Editorial, formatting, and spelling
- Clarification

To request a change to this document, contact the Document Author or Owner.
order (latest version first).

Revision	Date	Created by	Short Description of Changes
2	28/4/2025	Louise Wipfler	Correction of the report based on the reviewers comments

Configuration Management: Document Location

The latest version of this controlled document is stored in the project SharePoint Folder

'Submitted Deliverables' and in the Document Library of the EU Funding and Tenders Portal.

Nature of the deliverable		
R	Report	

Dissemination level		
PU	Public	

ACKNOWLEDGEMENT

This report is part of the deliverables from the project "SOILPROM" which has received funding from the European Union's Horizon Europe Research and Innovation program under grant agreement No 101156589.

We thank the external review by SOILPROM modelling board members, Nicholas Jarvis from the Swedish University of Agricultural Sciences (SLU) and Yonggen Zhang from Tianjin University (TJU), for their willingness to provide their feedback to the plans as presented in this report.

More information on the project can be found at: http://www.soilprom.eu.

SOILPROM

TABLE OF CONTENTS

EXECUTIVE SUMMARY

SOILPROM objective is to improve the understanding of 13 soil related processes that are currently under-represented in modelling pollutant transport. This will be done through the upgrading/integrating and validation of a selection of models across different environmental compartments. The activity as reported in this deliverable aimed to:

- Revisit and finetune the list of the models preselected by the partners to be further upgraded, validated, and applied in WP2, and included in the Modelling Platform in WP4.
- Provide plans for models upgrading/integrating and validation.

In the Annexes all SOILPROM model upgrading/integrating and validation plans are collected. The Deliverable will feed into Task 2.2 on the upgrade of the models and T2.3 on the integration across environmental compartments.

I. INTRODUCTION AND BACKGROUND

SOILPROM objective is to improve the understanding of 13 prime soil related processes that are currently under-represented in modelling pollutant transport (Figure 1). SOILPROM will improve the understanding and the description of these processes through upgrading/integrating and validation of a selection of models across different environmental compartments.
To ensure smooth developments, the SOILPROM consortium preselected models in the proposal writing phase to be used for further development in the project. These models cover diffuse pollution processes for metals, pesticides, microplastics, PFAS, and nutrients. The preselected models focus on the following processes: (1) Adsorption and desorption of ionizable substances, as PFAS, (2) Colloidal transport of microplastics in soil, (3) Metal biogeochemistry in soil, (4) Sorption and desorption of phosphorus, (5) Wind erosion and atmospheric transport processes of pesticides, (6) Wind erosion and atmospheric transport processes of microplastics, (7) Atmospheric deposition of PFAS to soil, (8) Wind erosion and atmospheric transport processes of metals, (9) Water erosion and runoff of dissolved and sediment-bounded pesticides, (10) Metal transport by infiltration, water erosion, and runoff, (11) Plant uptake of PFAS, (12) Transport of pesticides in soil and groundwater, (13) Transport of nutrients in soil towards surface water.

The activity as reported in this deliverable aimed to:

- Revisit and finetune the list of the models preselected by the partners to be further upgraded, validated, and applied in WP2, and included in the Modelling Platform in WP4.
- Provide plans for models upgrading/integrating and validation.

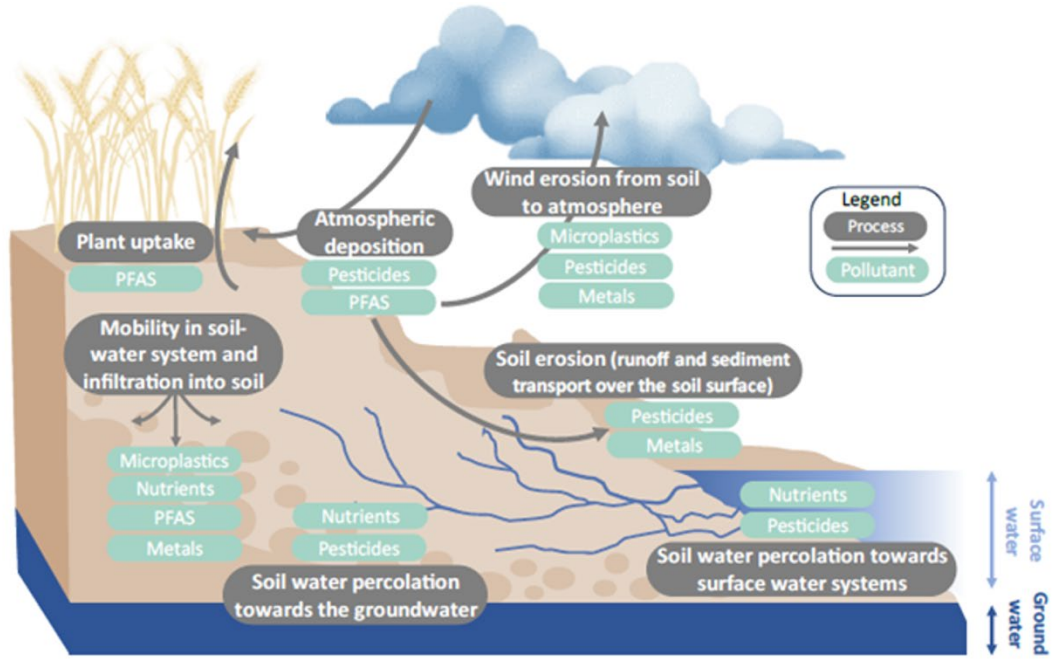

Figure 1 Soil-related processes studied in SOILPROM.

Upgraded/integrated models will be applied in 9 use-cases as presented in Table 1. The report builds on D1.1 in which the use-cases are further specified as well as the requirements regarding the modelling platform (MP) and the decision support tool (DST), and D1.2 in which the knowledge base is given and gaps are identified. It feeds into Task 2.2 on the upgrade of the models and T2.3 on the integration across environmental compartments.

Table 1. Overview of SOILPROM use-cases.

Pollutant	Use-Cases		Use-Case Leader Organisation	Location
Microplastics	1	Colloidal transport of microplastics in soil	WU	Wageningen, The Netherlands
	2	Wind erosion and atmospheric transport and deposition of dust and microplastics	WU	Valthermond, The Netherlands
Pesticides	3	Water erosion and runoff of dissolved and sediment-bounded pesticides	WU	Limburg, The Netherlands
	4	Wind erosion and atmospheric transport and deposition of dust-bounded pesticides	WU	Valthermond, The Netherlands
	5	Flow of water and transport of pesticides in soils and groundwater	FZJ	Jülich, Germany

PFAS	6	Atmospheric PFAS deposition to soil, and plant uptake of PFAS	VITO	Willebroek, Belgium
Heavy metals	7	Biogeochemistry of metals, and fate and transport of metals due to infiltration and water and wind erosion processes	UPCT	Cartagena-La Unión Mining District, Spain
Nutrients	8	Transport of nutrients in soil, groundwater, and surface water with marine discharge	GUT	Puck bay, Poland
	9	Sorption and desorption of phosphorus in soil, and transport of phosphorus to surface water	NIBIO	Vandsemb, Norway

II. APPROACH

The preliminary list of models was revisited by the SOILPROM project team, as well as the link between the identified soil pollution processes to be studied and the envisioned model improvements and validation.

Plans for model upgrading/integrating and validation were developed based upon addressing the following questions. Model improvement and validation plans have been added as annexes to this report.

- What aspects needs to be updated/upgraded/validated? And why?
- Are there approaches available (e.g. for other pollutants) which can be used as example?
- Are there other models available that may be better suitable, or which can be used for inspiration?
- How will the model be improved? Which solutions are foreseen?
- Which steps will be taken?
- What is the validation plan of the improvements and how is this linked to the use case?
- Which data are needed? How will be made use of public data (maps?), such as those from e.g. JRC?

III. LIST OF MODELS TO BE UPGRADED/INTEGRATED AND VALIDATED IN SOILPROM

Revisiting the list of models, resulted in more targeted combination of model-processes as compared to the preliminary list. Decisions were taken which model to use to describe a specific process best (e.g. it was decided to use GeoPEARL for the coupling of the atmospheric deposition of PFAS to soil-transport instead of HYDRUS). It was further decided to focus the model upgrade and testing of the HYDRUS-HPx model on microplastics and nutrients. Metals will not be the focal substance for this model. Improvement of the description of transport of metals

due to water erosion and wind erosion was given priority above the further elaboration of the geochemical processes in soil.

The final list and the link to the processes and use-cases is given in Table 2. Based on this consolidated list in a series for model upgrade plans that are drafted and included as annexes to this report.

Table 2. Overview of the models to be used in SOILPROM, including the links with soil-related processes, pollutant types and use-cases. The plans for upgrade of the process-model combinations are referred to in the last column of the Table.

	Model	Soil related process simulation that are currently underrepresented and for which the model will be used	Pollutant	Use-case no.	Plan described in
1	HYDRUS, HYDRUS-HPx	12. Transport of pesticides in soils and groundwater	Pesticides	5	Annex 1
		13. Transport of nutrients in soil	Phosphorus and nitrogen	8	Annex 2
		2. Colloidal transport of microplastics	Microplastics	1	Annex 3
2	MODFLOW/MT3D	12. Transport of pesticides in soil and groundwater	Pesticides	5	Annex 1
		13. Transport of nutrients in soil	Nutrients	8	Annex 2
3	TSMP	12. Transport of pesticides in soils and groundwater	Pesticides	5	Annex 1
4	SWAT	13. Transport of nutrients in soil	Phosphorus and nitrogen	8	Annex 2
		4. Sorption and desorption of phosphorus	Phosphorus	9	Annex 2
5	SWAP-PEARL	1. Adsorption and desorption PFAS	PFAS	6	Annex 4
		11. Plant uptake of PFAS	PFAS	6	Annex 5
6	Cplantbox	11. Plant uptake of PFAS	PFAS, pesticides	5, 6	Annex 6
7	IFDM	7. Atmospheric deposition of PFAS to soil	PFAS	6	Annex 7
8	ICECREAM	4. Sorption and desorption of phosphorus	Phosphorus	9	Annex 8
9	MicroHH	5. Wind erosion and atmospheric transport processes of pesticides	Pesticides	4	Annex 9
		6. Wind erosion and atmospheric transport processes of microplastics	Microplastics	2	Annex 9
		8. Wind erosion and atmospheric transport processes of metals	Metals	7	Annex 9
10	OpenLISEM	9. Water erosion and runoff of dissolved and sediment-bounded pesticides	Pesticides	3	Annex 10
		10. Transport of metals by infiltration, water erosion, and runoff	Metals	7	Annex 10

List of plans for model upgrading/integrating and validation:

ANNEX 1: HYDRUS-MODFLOW-MT3DMS / HYDRUS-TSPM PESTICIDE
ANNEX 2: HYDRUS- SWAT-MODFLOW-MT3DMS- NUTIENTS
ANNEX 3: HYDRUS-COLLOIDAL TRANSPORT OF MICROPLASTICS
ANNEX 4: PEARL- ADSORPTION-DESORPTION OF PFAS
ANNEX 5: PEARL-PLANT UPTAKE OF PFAS
ANNEX 6: CPLANTBOX- PLANT UPTAKE OF PFAS AND PESTICIDE
ANNEX 7: IFDM-(GEO-)PEARL- ATMOSPHERIC DEPOSITION OF PFAS
ANNEX 8: ICECREAM-PHOSPHORUS
ANNEX 9: MICROHH-WIND EROSION AND ATMOSPHERIC TRANSPORT OF PESTICIDES, MICROPLASTIC and METALS
ANNEX 10: OPENLISEM- WATER EROSION AND RUNOFF OF PESTICIDES AND METALS

ANNEX 1: HYDRUS-MODFLOW-MT3DMS / HYDRUS-TSPM PESTICIDES

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**HYDRUS-MODFLOW-MT3DMS / HYDRUS-TSPM**
Process	Transport of pesticides in soil and groundwater
Pollutant	Pesticides
Lead partner organisation	FZJ

UPGRADING-INTEGRATING NEEDS

According to the EU Water Framework Directive (WFD, Dir. 2000/60/EC) and the Pesticide Directive (Dir. 2009/128/EC) the reduction of the groundwater pollution by pesticides is an important goal. Once pesticides reach the soil surface, they are transported mainly by the water phase downwards undergoing complex physical, chemical and biological transformations while moving through the unsaturated zone (Yaron, 1989). In groundwater, pesticide concentrations are measured mainly if the water is used for drinking water purposes. Within the EU a strict limit of 0.1 µg l-1 (Directive (EU) 2020/2184, 2020) is allowed as maximum pesticide concentration in drinking waters for single compounds and 0.5 µg l^{-1} for the sum of all pesticides (de Oliveira et al., 2023). Pesticide contamination of groundwater is generally assessed through pesticide monitoring programs (e.g., Worral et al., 2005; Thapinta et al., 2003; Papadopoulou-Mourkidou et al., 2004) and once the information about the pesticide concentrations in the groundwater is obtained, to assess the potential to mitigate the contaminant, it is crucial to analyze the spatiotemporal relations between surface applications of pesticides and their groundwater concentrations. Classically, the potential inflow of pesticides into the groundwater is predicted by the pesticide leaching leaving the root zone (FOCUS, 2000), whereas the vadose zone can be much deeper than the rooted zone and varying groundwater tables will directly impact the timing of pesticides reaching the groundwater. Coupled soil and groundwater models (Franke and Teutsch, 1994, Kupferberg et al., 2018, Beegum et al., 2020) can help to identify the source areas and also to predict the long-term pollution of the groundwater by pesticides sprayed on the soil surface. In these coupled soil and groundwater models the fluctuations of the groundwater table need to be provided as boundary conditions as the modelling domain is not independent from outside inflow of groundwater. This is classically done using piezometer data, although those data are not always available at the sites of interest. Therefore, coupling the soil and groundwater models with existing land-surface-models (LSM) which simulate also dynamic groundwater tables at high spatial resolution such as those presented by Belleflamme et al. (2023) seems to be a solution.

However, the accuracy of the predicted groundwater fluctuations at the site of interested simulated by the LSM-groundwater model is largely unknown and the impact of fluctuating groundwater tables on the pesticide masses entering the groundwater remains unexplored.
Therefore, the aim of this subproject is to analyse the impact of fluctuating groundwater tables on the pesticide masses reaching the groundwater using a coupled unsaturated/saturated model suit (HYDRUS-MODLOW-MT3DMS) in spatial (200 m vertical and cm horizontal for the unsaturated zone) and temporal high resolution. Thereby, we aim to explore the sensitivity of model boundary and input parameters such as management (e.g., crop rotation), soil maps and upscaling, climate input data and groundwater fluctuations on the pesticide masses entering the groundwater. As for a general use (transferability to any site within EU) no exact information on the groundwater fluctuations might be available, we also aim to couple the high resolution model TSMP (Terrestrial Systems Modelling Platform), which is currently operational at 600 m for the German domain and at 3 km at the EU domain, with the unsaturated zone model HYDRUS-1D for a seamless model suite.

In general, the model exercise proposed and the new model coupling of HYDRUS with TSMP can be used to analyse also other pollutants such as PFAS, heavy metals, colloids, and microplastics as all those pollutants have been already integrated into the HYDRUS software.

Review of related models

To assess pesticide leaching to groundwater at regional scales, 1D pesticide fate models have been coupled with geographical information about soil properties, climate, and land use (e.g. GeoPearl, EuroPearl). In some studies, also groundwater table depth information and annual groundwater fluctuations were included in the approach. With these approaches, the spatial distribution of pesticide leaching to groundwater could be assessed. However, pesticide concentrations in groundwater are not simulated. Other models like Hydrus-3D or HydroGeoSphere simulate the variable saturated flow in soil-aquifer systems in 3D and couple this with 3D transport simulations so that also pesticide concentrations in groundwater is simulated. However, these models are very computationally expensive at the scale of interest and therefore not easy to run, compared to the computationally less demanding approached proposed here.

Plan for model improvement

In the project, two major improvements are envisioned. The first improvement is to use TSMP modelled groundwater levels as inputs for the HYDRUS-MODFLOW-MT3DMS simulations to better represent groundwater fluctuations in the model and to cope with changes in vadose zone thickness, which is of special importance if the groundwater table is shallow. Therefore, the simulated groundwater tables

from TSMP will be used as inputs into the HYDRUS-MODFLOW-MT3DMS model suite as a kind of database and no direct coupling between the two models will be done. Secondly, the HYDRUS model will be also coupled to TSMP to simulate seamlessly the impact of fluctuating groundwater tables on pesticide transport through the vadose zone.

Model validation

As the pesticide transport simulations cannot be easily validated since only sparse data of pesticide contamination of the aquifer are available, we will first concentrate on the correct representation of the groundwater tables. Therefore measured data from the local water authorities will be used. Secondly, selected pesticides will be simulated, which can provide information on the long-term fate of such pesticides. Therefore, banned pesticides such as Atrazine are suited as the ban is documented and the atrazine concentrations in the groundwater have been monitored over the last 2 decades.
As the model results strongly depend on the choice of input parameters such as soil hydraulic properties and atmospheric forcing as well as the up or downscaling of such data, different publicly available datasets such as SoilGrids @ 250 m scale for the soil information (soil texture, bulk density, horizonation) are used and up-scaled to the model grid of 200x200 m. As the upscaling poses an additional source of uncertainty different upscaling approaches are tested and a functional sensitivity study will be performed. Additionally, different publicly available data on atmospheric forcing (e.g., ERA5 or ERA5-Land) are compared with long-term records from locally available climatic stations. As the up-scaling of soil properties and the selection of appropriate atmospheric forcing is not only crucial for the Zwischenscholle use-case, but we will also extend the analysis to all 7 use-cases within SOILPROM.

References

Beegum, S.; Vanderborght, J.; Šimůnek, J.; Herbst, M.; Sudheer, K.P.; & Nambi, I.M. (2020) Investigating Atrazine Concentrations in the Zwischenscholle Aquifer Using MODFLOW with the HYDRUS-1D Package and MT3DMS. Water: 12, 1019. https://doi.org/10.3390/w12041019

Belleflamme, A., Goergen, K., Wagner, N., Kollet, S., Bathiany, S. Zohbi, J.E, Rechid, D., Vanderborght, J. & Vereecken, H. (2023). Hydrological forecasting at impact scale: the integrated ParFlow hydrological model at 0.6 km for climate resilient water resource management over Germany. Front. Water. 5: https://doi.org/10.3389/frwa.2023.1183642

de Oliveira, D.M. Agostinetto, L., & Siegloch, A.E. (2023). Comparison of the drinking water standard for pesticides of the Brazil with other countries. Heliyon, 9 (3): e13783, https://doi.org/10.1016/j.heliyon.2023.e13783.

FOCUS (2000). FOCUS groundwater scenarios in the EU review of active substances" Report of the FOCUS Groundwater Scenarios Workgroup, EC Document Reference Sanco/321/2000 rev.2, 202pp

Franke, H.-J. & Teutsch, G. (1994). Stochastic simulation of the regional pesticide transport including the unsaturated and the saturated zone. Ecol. Model. 75, 529–539.

Kupfersberger, H.; Klammler, G.; Schumann, A.; Brückner, L., & Kah, M. (2018). Modeling subsurface fate of s-metolachlor and metolachlor ethane sulfonic acid in the Westliches Leibnitzer Feld aquifer. Vadose Zone J. 17, 170030.

Papadopoulou-Mourkidou, E.; Karpouzas, D.G.; Patsias, J.; Kotopoulou, A.; Milothridou, A.; Kintzikoglou, K.; & Vlachou, P. (2004). The potential of pesticides to contaminate the groundwater resources of the Axios river basin in Macedonia, Northern Greece. Part I. Monitoring study in the north part of the basin. Sci. Total Environ. 321, 127–146.

Thapinta, A. & Hudak, P.F. (2003). Use of geographic information systems for assessing groundwater pollution potential by pesticides in Central Thailand. Environ. Int. 29, 87–93.

Worrall, F.& Besien, T. (2005). The vulnerability of groundwater to pesticide contamination estimated directly from observations of presence or absence in wells. Journal of Hydrology. 303, 92–107.

Yaron, B. (1989). General principles of pesticide movement to groundwater. Agriculture, Ecosystems & Environment, 26(3-4): 275-297. https://doi.org/10.1016/0167-8809(89)90016-9.

ANNEX 2: HYDRUS - SWAT-MODFLOW-MT3DMS- NUTRIENTS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**HYDRUS- SWAT-MODFLOW-MT3DMS**
Process	Transport of nutrients with marine discharge
Pollutant	Nutrients
Lead partner organisation	GUT

UPGRADING-INTEGRATING NEEDS

The model upgrade activities are based on the existing modelling platform for the Puck Bay area, developed in the framework of earlier project WaterPUCK (https://waterpuck.pl/). The modelling system consists of a SWAT hydrological model for surface water and soil water flow, a MODFLOW-NWT model for groundwater flow, a MT3DMS model for nitrate transport in groundwater and 3D CEMBS hydrodynamic and ecological model of the Puck Bay. Details of the modelling platform are provided by Dzierzbicka-Głowacka et al. (2022), Wielgat et al. (2021) and Szymkiewicz et al. (2020). An overview of connections between models is shown in Fig. 2.

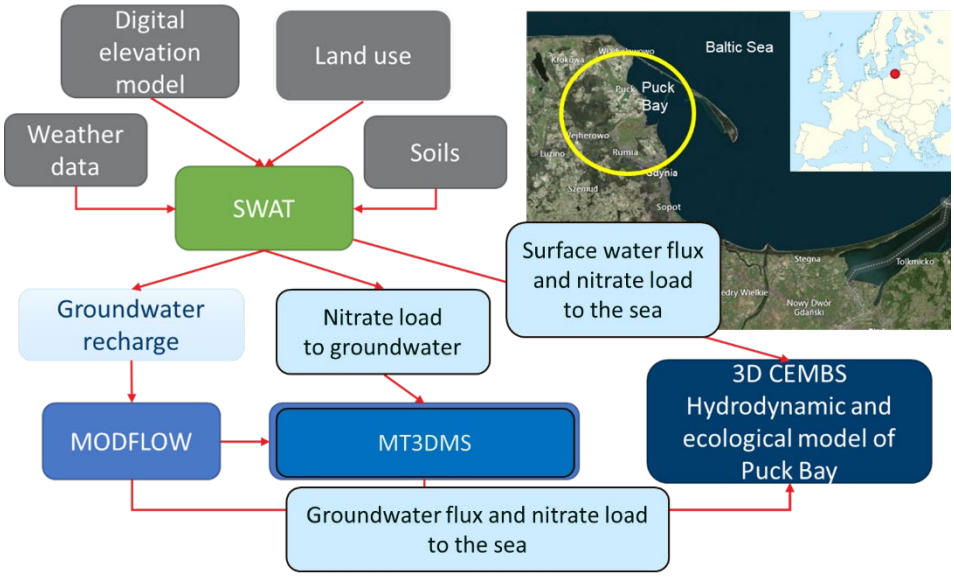

Fig. 2. Model connections in the existing modelling platform WaterPUCK.

The existing modelling system has several shortcomings, which were described in SOILPROM Deliverable D1.2 Report on soil pollution processes knowledge gaps and will be addressed in the project. An overview of the considered model upgrades is presented in Fig. 3. Specifically, we plan to:

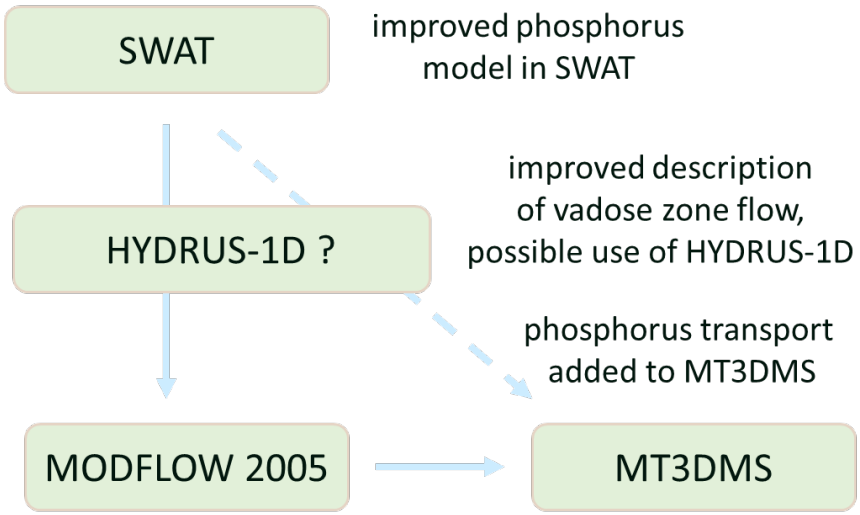

Fig. 3. Overview of planned model upgrades. The improved vadose zone description includes improved nitrate transport.

1. Improve the description of nitrate through deep vadose zone towards the groundwater table. The current model (SWAT) describes the movement of water through deep vadose with a relatively simple exponential transfer function (Neitsch et al. 2011):

$$w_{rch,i} = \left(1 - exp\left[-1/\delta_{gw}\right]\right) \cdot w_{seep,i} + exp\left[-1/\delta_{gw}\right] \cdot w_{rch,i-1} \quad (1)$$

where $w_{rch,i}$ is the recharge on day i, $w_{rch,i-1}$ is the recharge on day i-1, $w_{seep,i}$ is the water percolation from the root zone and d_{gw} is the model parameter related to transfer time through the unsaturated zone. Similarly, the movement of nitrate below the root zone is described as (Neitsch et al. 2011):

$$NO3_{rch,i} = \left(1 - exp\left[-1/\delta_{gw}\right]\right) \cdot NO3_{seep,i} + exp\left[-1/\delta_{gw}\right] \cdot NO3_{rch,i-1} \quad (2)$$

where $NO3_{rch,i}$ is the nitrate load to groundwater transported by recharge on day i, $NO3_{rch,i-1}$ is the load on day i-1 and $NO3_{seep,i}$ is the nitrate load transported by water seeping from the root zone on day i. While the above approach is very efficient computationally, comparisons with a more comprehensive model based on the Richards' equation showed that it does not accurately represent time lags of water flow and pollutant transport, especially for deeper groundwater tables and soils with lower permeability, such as sandy loams or loams. In SOILPROM we will seek for an alternative approach, representing a compromise between accuracy and computational efficiency, as described in the following sections. We plan to test the following modelling approaches: (i) 1D Richards' equation + advective-dispersion equation (HYDRUS-1D), (ii) kinematic wave equation, as implemented in MODFLOW-UZF/UZT packages (Niswonger et al. 2006), (iii) transfer functions.

2. Upgrade the phosphorus model (vadose zone and groundwater) by adding phosphorus leaching from soil to groundwater, phosphorus transport in groundwater and an improved description of phosphorus sorption to soil components. The improved description will be developed by NIBIO. In the current model (SWAT) phosphorus can leach from soil to surface water only, not to groundwater. Consequently, phosphorus transport in groundwater is not modelled. However, elevated concentrations of phosphorus were found in groundwater in some locations of the study area. Moreover, there is evidence that the models of phosphorus sorption should consider increased sorption capacity due to Fe, Al and Ca presence in soil (a more detailed discussion is provided in another chapter of this report, i.e. on the ICECREAM model). In SOILPROM we plan to introduce the following upgrades to the existing model components: (i) adding a model for phosphorus transport through the deep vadose zone analogous to the model for nitrate, (ii) adding phosphorus transport in groundwater to the MT3DMS model, (iii) implementing an improved model for phosphorus sorption developed by NIBIO in the SWAT program.

REVIEW OF RELATED MODELS

Several existing models can be used to describe transport of nitrogen and phosphorus species through the vadose zone. The most comprehensive models for nitrogen transport include both water flow and air flow. They use either the full two-phase flow formulation, i.e. water and air flow described by two coupled differential equations (Akhavan et al. 2013) or an alternative formulation based on Richards' equation for water flow and a simplified model for air flow (Ben Moshe et al. 2021). The fluid flow equations are coupled with advection-dispersion equations describing movement of different forms of nitrogen (typically organic N, ammonia and nitrate), as well as organic carbon and oxygen (Akhavan et al. 2013, Ben Moshe et al. 2013). Microbial biomass growth is also included.

The next group of models does not consider air flow and uses Richards' equation to describe water flow through the vadose zone and the advection-dispersion equation to describe the movement of contaminants, as implemented e.g. in HYDRUS-1D (Šimůnek et al. 2013) or SWAP (Kroes et al. 2017) computer programs. Some models from this group include several N species (organic N, ammonia, nitrate) and account for the effects of various environmental factors (such as water content or temperature) on reaction rates (Murphy et al. 2024). Other models focus on ammonia and nitrate and use constant reaction rates (e.g. Lahjouj et al. 2023, Liu et al. 2013). The simplest models from this group assume non-reactive (conservative) contaminant transport (e.g. Vero et al. 2017) and are applicable to nitrate leaching from the root zone to the water table under the assumption of negligible denitrification.

The third group consists of models which replace the numerical solution of advection-dispersion equations with a simplified representation of contaminant movement along the soil profile. For example, the ANIMO model (Groenendijk et al. 2005) is based on a semi-analytical solution to the advection-dispersion

equations for nitrogen and phosphorus species. It can be coupled with the SWAP agro-hydrological model (Kroes et al. 2017) based on the Richards equation, to provide input data on water content and water fluxes in the soil profile (e.g. Sabzzadeh and Alimohammadi 2023). Alternatively, the ANIMO model can be coupled with a simplified model for water movement in soil given by the kinematic wave approximation, as implemented in the UZF package for MODFLOW groundwater model (Niswonger et al. 2006, De Filippis et al. 2021). The kinematic wave approximation offers a significant advantage over the Richards equation in terms of computational efficiency.
We plan to focus our efforts on the models based on Richards' equation, the kinematic wave equation and alternative transfer functions (Jury and Roth 1990) as possible replacements to the transfer function currently implemented in SWAT. Considering phosphorus transport, Wei and Bailey (2021) described a coupled SWAT-MODFLOW-RT3D model, which is capable of simulating phosphorus leaching from soil to groundwater and its further transport in the subsurface. This model may be a helpful reference for the development of phosphorus transport model in SOILPROM. An improved formulation of phosphorus sorption will be developed by NIBIO and is described in another section of this report.

Plan for model improvement

Upgrade of the model for water flow and nitrogen transport in deep vadose zone will be carried out in the following steps:

1. Literature review on transfer functions and selection of the functions with the largest application potential,
2. Numerical simulation of deep vadose zone flow and transport with HYDRUS-1D, based on the Richards equation, for a wide range of soil conditions typical of the field site. Due to the comprehensiveness of the Richards' equation, these simulations will be considered as reference to evaluate other methods,
3. Comparisons of Richards' equation-based simulations with kinematic wave model and transfer functions, in terms of accuracy and computational efficiency. Selection of the most appropriate model for vadose zone transport taking into account accuracy, efficiency and data requirements (models with lower data requirements are preferred),
4. Implementation of the selected vadose zone model in the coupled SWAT-MODFLOW model. Depending on which model will be chosen, this can be achieved either by modifying the source code of SWAT (which is open and freely accessible) or by linking an external program (e.g. HYDRUS-1D) via Python scripts.

Implementation of the phosphorus model will be performed in the following steps:

1. Modification of SWAT source code to include dissolved phosphorus leaching from the root zone to groundwater (Wei and Bailey 2021),

2. Adding dissolved phosphorus transport to the model for deep vadose zone flow and transport. The choice of the vadose zone model will be consistent with the model for nitrogen and will depend on the inter-model comparison described above,
3. Adding dissolved phosphorus transport in groundwater to MT3DMS model. The phosphorus transport will be described by the advection-dispersion equation:

$$R\frac{\partial c}{\partial t} = -\frac{\partial}{\partial x}(v_i c) + \frac{\partial}{\partial x}\left(D_{ij}\frac{\partial c}{\partial x}\right) + S \quad (3)$$

where c is the dissolved phosphorus concentration, R is the retardation factor accounting for sorption of phosphorus on the soil/rock matrix, v_i are the components of groundwater velocity (obtained from MODFLOW model), D_{ij} are the components of hydrodynamic dispersion tensor, S is the source term representing leaching from the soil and other processes. The source term will be specified using the results of the simulation with the chosen vadose zone model. The transfer of data between models will be implemented by Python script in similar way as it is done for nitrate in the current model.

Furthermore, we will attempt to implement the modified model for phosphorus sorption developed by NIBIO in the SWAT model. This will require modification of the SWAT source code and possibly including additional input data, such as Fe and Al content in soil. In case of difficulties with implementation in SWAT we will look for alternatives, such as the HP1 model (Jacques and Simunek 2005), which is a combination of HYDRUS-1D and PHREEQC and offers a large flexibility in defining hydrogeochemical reactions.

MODEL VALIDATION

The current set of coupled models was developed in the earlier project WaterPUCK and an extensive set of input data is available, including weather data, land management practices, plant growth parameters, soil and aquifer parameters. The models were calibrated against measured groundwater levels, nitrate concentrations, crop yield and other variables (Szymkiewicz et al. 2020). The new data needs of the upgraded models are as follows:

Models for water flow and nitrate transport in the deep vadose zone. Data requirements depend on the choice of specific form of the model which will be decided at a later stage of the project, as described above. Models based on the Richards' equation and advection-dispersion equation require data on soil water retention curve, hydraulic conductivity curve and hydrodynamic dispersion coefficients. Soil hydraulic data can be inferred from the data currently used in SWAT model and the dispersion coefficient can be estimated using available empirical formulas. The models based on kinematic wave equation also require

data about soil hydraulic properties, which can be estimated using the data from the SWAT model, soil maps or pedotransfer functions. The transfer functions typically require a limited number of parameters, which might be physically based or empirical. These parameters will be either estimated from available soil data or calibrated by comparing the results with the simulations using other models performed as part of the inter-model comparison described in the previous section.

Phosphorus transport. The main new data requirement for the phosphorus model is the phosphorus sorption coefficient, describing P sorption to soil and aquifer matrices. This parameter is needed for the MT3DMS model and possibly also for the deep vadose zone transport model. We plan to estimate the sorption coefficient based on literature data and eventually refine it by calibrating the groundwater model to the measured phosphorus concentrations.
Additionally, we plan to collect local-scale data on three sites within our research area (Fig. 4). On each site we will perform measurements in soil and shallow groundwater. Groundwater data include water table depth, EC, dissolved forms of nitrogen (NO3, NO2, NH4) and dissolved phosphorus. Soil data include water retention curves, saturated hydraulic conductivity, total nitrogen, total phosphorus, and possibly also NO3, NH4, plant-available phosphorus and Fe, Al and Ca content. Groundwater data will be collected monthly and soil data every 3 months. This data will be used to validate the upgraded models for vadose zone transport and phosphorus sorption at the local scale.

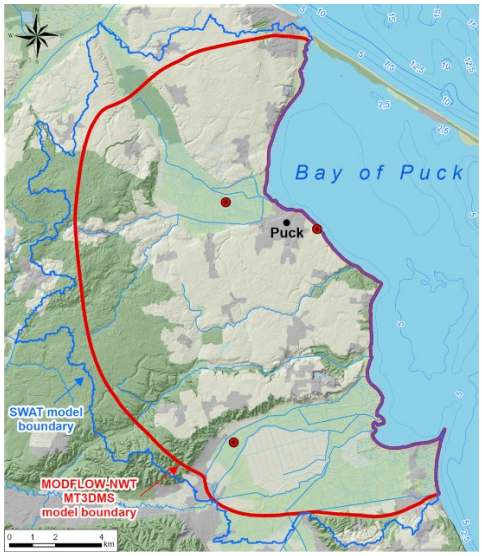

Fig. 4 GUT use-case area with 3 sites for local-scale measurements.

An alternative set of results will be obtained by performing simulations at the local scale using data available in open databases, described in SOILPROM Deliverable D1.3. Specifically, we plan to use data on soil properties (ISRIC and ESDAC

databases), land cover (CORINE database) and weather (COPERNICUS database) and compare the results with simulations based on locally measured parameters.

REFERENCES

Akhavan, M., Imhoff, P. T., Andres, A. S., & Finsterle, S. (2013). Model evaluation of denitrification under rapid infiltration basin systems. Journal of contaminant hydrology, 152, 18-34.

Bancheri, M., Coppola, A., & Basile, A. (2021). A new transfer function model for the estimation of non-point-source solute travel times. Journal of Hydrology, 598, 126157.

Ben Moshe, S., Weisbrod, N., & Furman, A. (2021). Optimization of soil aquifer treatment (SAT) operation using a reactive transport model. Vadose Zone Journal, 20(1), e20095.

De Filippis, G., Ercoli, L., & Rossetto, R. (2021). A spatially distributed, physically-based modeling approach for estimating agricultural nitrate leaching to groundwater. Hydrology, 8(1), 8.

Dzierzbicka-Glowacka, L., Dybowski, D., Janecki, M., Wojciechowska, E., Szymczycha, B., Potrykus, D., ... & Puszkarczuk, T. (2022). Modelling the impact of the agricultural holdings and land-use structure on the quality of inland and coastal waters with an innovative and interdisciplinary toolkit. Agricultural Water Management, 263, 107438.

Jacques, D., & Simunek, J. (2005). User manual of the multicomponent variably-saturated flow and transport model HP1 (No. BLG--998). SCK-CEN.

Jury, W. A., & Roth, K. (1990). Transfer functions and solute movement through soil: theory and applications (pp. 226-pp).

Lahjouj, A., El Hmaidi, A., M'hamed, B., & Karima, B. (2023). Simulation of soil water and nitrate transport in wheat field under various nitrogen fertilizer rates and rainfed conditions using HYDRUS-1D. Soil Science Annual, 74(1), 1-14.

Liu, X., Sun, S., Ji, P., & Šimůnek, J. (2013). Evaluation of historical nitrate sources in groundwater and impact of current irrigation practices on groundwater quality. Hydrological Sciences Journal, 58(1), 198-212.

Murphy, N. P., Furman, A., Moshe, S. B., & Dahlke, H. E. (2024). Comparison of reactive transport and non-equilibrium modeling approaches for the estimation of nitrate leaching under large water application events. Journal of Hydrology, 628, 130583.

Neitsch, S.L., Arnold, J.G., Kiniry J.R., Williams, J.R. (2011) Soil and Water Assessment Tool - Theoretical Documentation, Version 2009. Texas Water Resources Institute Technical Report No. 406

Niswonger, R. G., Prudic, D. E., & Regan, R. S. (2006). Documentation of the Unsaturated-Zone Flow (UZF1) Package for modeling unsaturated flow between the land surface and the water table with MODFLOW-2005 (No. 6-A19).

Šimůnek, J., Šejna, M., Saito, H., Sakai, M., & van Genuchten, M. T. (2013). The HYDRUS-1D software package for simulating the one-dimensional movement of water. Heat, and Multiple Solutes in Variably-Saturated Media, 4.

Szymkiewicz, A., Potrykus, D., Jaworska-Szulc, B., Gumuła-Kawęcka, A., Pruszkowska-Caceres, M., & Dzierzbicka-Głowacka, L. (2020). Evaluation of the influence of farming practices and land use on groundwater resources in a coastal multi-aquifer system in Puck region (Northern Poland). Water, 12(4), 1042.

Wei, X., & Bailey, R. T. (2021). Evaluating nitrate and phosphorus remediation in intensively irrigated stream-aquifer systems using a coupled flow and reactive transport model. Journal of Hydrology, 598, 126304.

Wielgat, P., Kalinowska, D., Szymkiewicz, A., Zima, P., Jaworska-Szulc, B., Wojciechowska, E., ... & Dzierzbicka-Glowacka, L. A. (2021). Towards a multi-basin SWAT model for the migration of nutrients and pesticides to Puck Bay (Southern Baltic Sea). PeerJ, 9, e10938.

ANNEX 3: HYDRUS–COLLOIDAL TRANSPORT OF MICROPLASTICS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**HYDRUS**
Process	Colloidal transport in soils
Pollutant	Microplastics
Lead partner organisation	WU

UPGRADING–INTEGRATING NEEDS

Taking into account current knowledge gaps in microplastic transport as identified in deliverable D1.2 and feasible research activities within SOILPROM, 5 innovations were identified:

- Effect of microplastic size and shape on their transport in soil
- Long-term field experiments for monitoring microplastics in soil
- Considering different plastic types, including biodegradable ones
- Preferential flow microplastics in macropores
- Integration with atmospheric deposition model

For colloidal transport in porous media, we can learn much from approaches used to simulate the transport of conventional colloids, such as microbeads and bacteria (Figure 5). While these types of colloids range in size from 0.001 to 1 µm and have a typical spherical shape, microplastics range in size from 0.1 to 5000 µm and have the shape of films, fibres, or granules. As pore sizes in natural soils range from 0.05 to 30 µm (micropores), 30 to 75 µm (mesopores), and 75 to 20,000 µm (macropores), the transport of microplastics is much more constrained by the physical size of the pores than conventional colloids. Therefore, the effect of microplastic size and shape is an important research item. Current measurement techniques for small plastic particles are limited to the size range 10 - 700 µm. Therefore, we focus on microplastics and don't include nano-plastics.

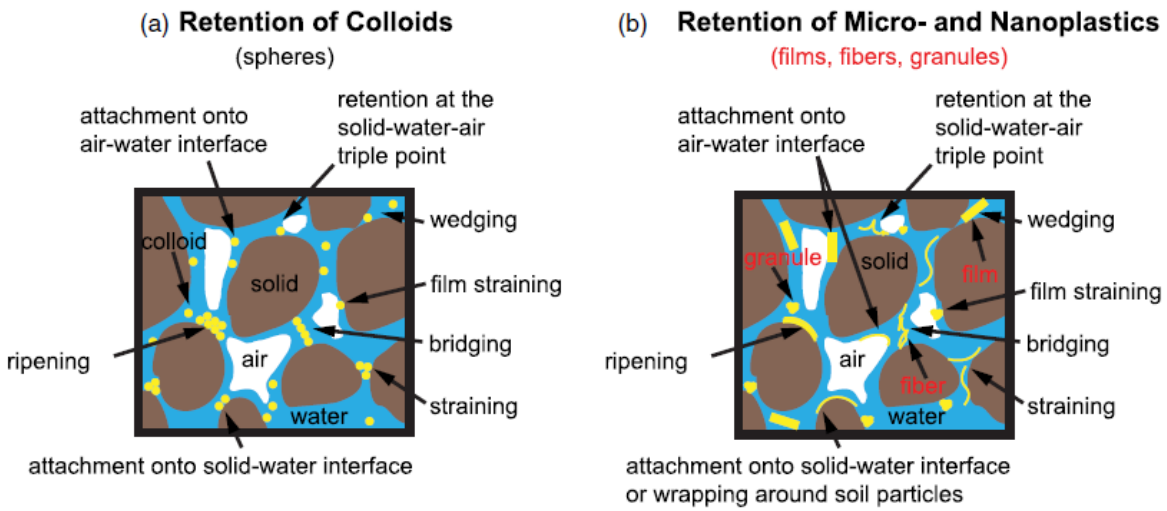

Figure 5. Retention mechanisms of (a) conventional colloids and (b) micro- and nanoplastics (after Yu and Flury, 2021).

Until now most microplastic transport experiments were performed on soil columns in the controlled conditions of a laboratory (Xu et al., 2020; Yu and Flury, 2021; Lwanga et al., 2022; Guo and Fei, 2023; Li et al., 2024). The duration of these experiments was limited to a few days or weeks. In SOILPROM, we investigate long-term transport (3 years) with 6 measurement campaigns under natural field conditions. These experiments will include different types of microplastics, including biodegradable plastics. The plastic decomposition rate depends much on soil environmental conditions such as moisture content, temperature, and soil microbes. Therefore, the long-term field experiment will yield valuable information beyond the existing information.

We hypothesize that the transport of microplastics in soils mainly occurs in macropores. However, most models for soil microbial transport assume uniform soil properties and make no distinction in the soil matrix between fine pores and macropores. Within SOILPROM, we will explore transport concepts tailored to the transport in both fine pores and macropores.

The 5th innovation relates to the integration of the atmospheric deposition model of microplastics with microplastic transport in the soil. This makes it possible to evaluate the environmental spreading of microplastics in areas with atmospheric microplastic deposition sourced from agricultural fields.

Review of related models

Guo and Fei (2023) provide a comprehensive and up-to-date review of experimental methodologies and theoretical models for microplastic transport in soils. Commonly, the one-dimensional convection-dispersion equation is used to fit the experimental results of column experiments. To better accommodate both the breakthrough and retention profiles of microplastics along the transport direction in soils, it is common to adopt a kinetic model for microplastic attachment and detachment to interfaces. Models differ in the number of kinetic

sites. Some models limit themselves to one kinetic site. Other models apply dual-site kinetic models. The two sites refer to the water-solid and air-water interfaces in unsaturated soils. Further models differ in the number of fitting parameters. The available data on microplastic concentrations and fluxes determine which fitting parameters can be optimized. Guo and Fei (2023) distinguished a total of 6 mainstream theoretical models. All 6 models are available within HYDRUS-1D. We will take advantage of the data in the extensive review of Guo and Fei (2023).
As HYDRUS-1D focuses on water flow in soils and incorporates different microplastic transport models, this model has been used in various scientific studies on microplastic transport in the years 2020-2024: Hu et al., 2020; Dong et al., 2021; Hu et al., 2021; Ren et al., 2021; Liang et al., 2022; Xu et al., 2022; Wang et al., 2022; Guo et al., 2023; Wang et al., 2023; Ameen et al., 2024; Feng et al., 2024; Yang et al., 2024.

As described earlier, most listed studies relate to soil columns in laboratory conditions and are limited to a few days or weeks. Within SOILPROM, we intend to simulate microplastic transport under field conditions for 3 years. We will thus build further on the model experiences gained thus far.

Plan for model improvement

In SOILPROM use-case 1, three types of microplastics (LDPE[1], PBAT[2], and Starch-based) are added to the soil surface of a field in a layout with 5 replicas (Figure 6). In the 3 years 2024-2026, we plan 6 measurement campaigns in which we collect soil samples at different depths. In these soil samples, we determine the microplastic concentrations of different sizes and shapes. In addition, we collect data on weather conditions (rainfall and evapotranspiration), soil hydraulic functions (retention function and conductivity function), and groundwater levels.
We plan to test several theoretical transport concepts for microplastics in soils:

1. Kinetic model for attachment and detachment of microplastics to interfaces
2. Simulate attachment to water-solid and water-air interfaces separately or together
3. Effect of size and shape of microplastics and macropores on transport

The model HYDRUS-1D includes modelling concepts for the above points. We will not add extra concepts but apply the concepts to the field data of use-case 1. We expect that the long-term field experiment will yield valuable extra information and insight in addition to the currently reported laboratory studies. This will enable the further underpinning and quantification of parameter values used in the different types of model concepts and for different types of microplastics. HYDRUS-1D includes an optimization shell, which allows parameter calibration. This

[1] Low-density polyethylene
[2] polybutylene adipate terephthalate (biodegradable)

yields valuable information on the optimal parameters, their confidence regions, and the reliability of the simulation.

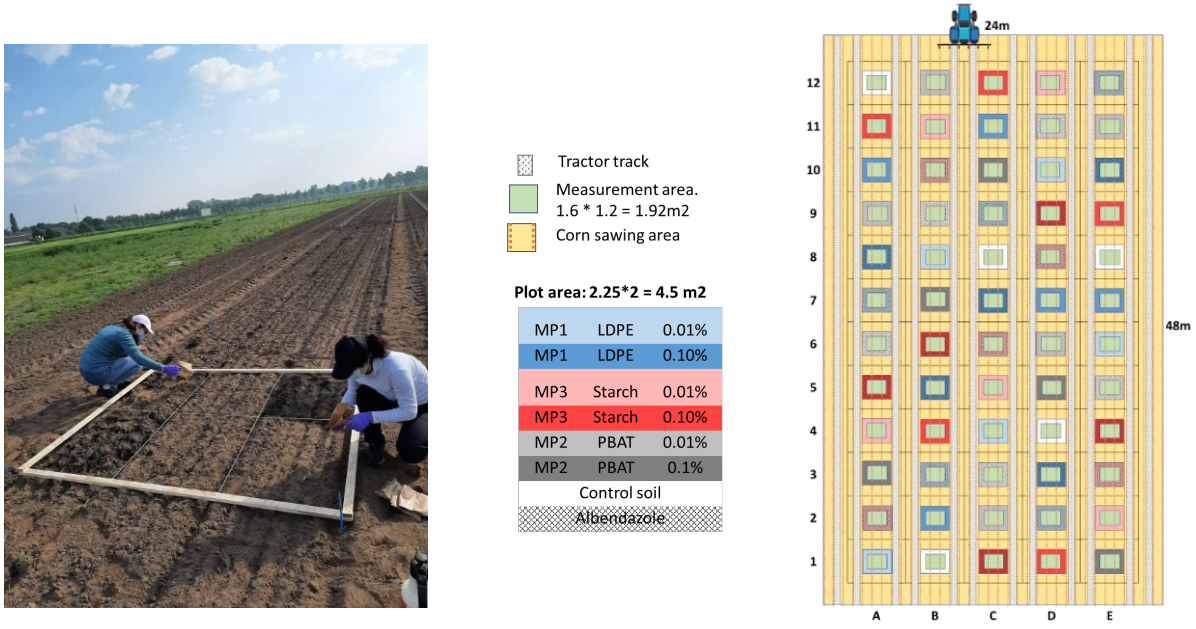

Figure 6. Setup of a long-term field experiment to measure transport of 3 types of microplastics near Wageningen. Data will be collected over a period of 3 years.

Model validation

The long-term field experiment allows for splitting the data in two periods. The data of the first period will be used for model calibration. The data of the second period will be used for model validation. Both calibration and validation will be performed with the different models that are available within HYDRUS on colloid transport and preferential flow. This will show the improvement with the model that is ultimately selected.

HYDRUS-1D is a process-based model. Therefore, the model is able to simulate soil water flow at any location in Europe if input data are available on weather (rain, evapotranspiration), land use, soil type (to derive soil hydraulic functions) and drainage conditions. Deliverable D1.3 specifies which databases can be used for HYDRUS as input for water flow, which is the main driver of microplastic transport. Within SOILPROM, similar input will be used for HYDRUS-1D to simulate pesticide transport in soils.

In SOILPROM, we will make the connection between an atmospheric model that simulates microplastic transport due to wind erosion in agricultural fields and the soil model that simulates microplastic transport from the soil surface downwards. In this way, the spreading of microplastics via the air and soil from hotspots in Europe with high microplastic concentrations can be analyzed. In addition, the validated model can be used at other locations where the microplastic concentration in the topsoil is known.

REFERENCES

Guo, Y., and Fei, X., 2023. Insights on experimental methodologies and theoretical models for microplastics transport in soils and sediments based on meta-analysis. Acta Geotechnica 18:4477–4492. https://doi.org/10.1007/s11440-022-01767-3

Li W., Brunetti G., Zafiu C., Kunaschk M., Debreczeby M., Stumpp C., 2024. Experimental and simulated microplastics transport in saturated natural sediments: Impact of grain size and particle size. Journal of Hazardous Materials, 468, https://doi.org/10.1016/j.jhazmat.2024.133772

Liu B., Shen Z., Zhou Q., Hu L., Zeng G., Wang X., Zhang M., Zhang D., Pan X., 2024. Microplastics in soil aggregates: Analytical methods, occurrence patterns, impact analyses and removal approaches. Trends in Analytical Chemistry, 178, https://doi.org/10.1016/j.trac.2024.117855

Lwanga E.H., Beriot N., Corradini F., Silva V., Yang X., Baartman J., Rezaei M., van Schaik L., Riksen M., Geissen V., (2022). Review of microplastic sources, transport pathways and correlations with other soil stressors: a journey from agricultural sites into the environment. Chemical and Biological Technologies in Agriculture, 9, https://doi.org/10.1186/s40538-021-00278-9

Xu, B. et al., 2020. Microplastics in the soil environment: occurrence, risks, interactions and fate–a review. Critical Reviews in Environmental Science and Technology, 50(21): 2175-2222. https://doi.org/10.1080/10643389.2019.1694822

Yu, Y. and Flury, M., 2021. Current understanding of subsurface transport of micro- and nanoplastics in soil. Vadose Zone Journal, 20, https://doi.org/10.1002/vzj2.20108

Hu, E., S. Shang, Z. Fu, X Zhao, X. Nan, Y. Du, and X. Chen, Cotransport of naphthalene with polystyrene nanoplastics (PSNP) in saturated porous media: Effects of PSNP/naphthalene ratio and ionic strength, Chemosphere, 245, 125602, 2020.

Dong, S., J. Xia, L. Sheng, W. Wang, H. Liu, and Bin Gao, Transport characteristics of fragmental polyethylene glycolterephthalate (PET) microplastics in porous media under various chemical conditions, Chemosphere, 276, 130214, 9 p., 2021.

Hu, E., H. Yuan, Y. Du, and X. Chen, LDPE and HDPE microplastics differently affect the transport of tetracycline in saturated porous media, Materials, 14, 1757, 10 p., 2021.

Ren, Z., X. Gui, Y. Wei, X. Chen, X. Xu, L. Zhao, H. Qiu, and X. Cao, Chemical and photo-initiated aging enhances transport risk of microplastics in saturated soils: Key factors, mechanisms, and modeling, Water Research, 202, 117407, 2021.

Liang, Y., Y. Luo, C. Shen, and S. A. Bradford, Micro-and nanoplastics retention in porous media exhibits different dependence on grain surface roughness and clay coating with particle size, Water Research, 221, 118717, 2022.

Xu, L., Y. Liang, C. Liao, T. Xie, H. Zhang, X. Liu, Z. Lu, and D. Wang, Cotransport of micro- and nano-plastics with chlortetracycline hydrochloride in saturated porous media: Effects of physicochemical heterogeneities and ionic strength, Water Research, 209, 117886, 2022.

Wang, X., Y. Dan, Y. Diao, F. Liu, H. Wang, and W. Sang, Transport and retention of microplastics in saturated porous media with peanut shell biochar (PSB) and MgO-PSB amendment: Co-effects of cations and humic acid, Environmental Pollution, 305, 119307, 9 p., 2022.

Guo, Y., and X. Fei, Insights on experimental methodologies and theoretical models for microplastics transport in soils and sediments based on meta-analysis, Acta Geotechnica, 16 p., doi: 10.1007/s11440-022-01767-3, 2023.

Wang, L., Z. Hu, H. Yin, S. A. Bradford, J. Luo, and D. Hou, Aging of colloidal contaminants and pathogens in the soil environment: Implications for nanoplastic and COVID-19 risk mitigation, Soil Use and Management, 39(1), 70-91, 2023.

Ameen, A., M. E. Stevenson, A. K. T. Kirschner, S. Jakwerth, J. Derx, and A. P. Blaschke, Fate and transport of fragmented and spherical microplastics in saturated gravel and quartz sand, Journal of Environmental Quality, 16 p., 2024.

Feng, Q., Z. Chen, G. Huang, C. An, X. Yang, and Z. Wang, Prolonged drying impedes the detachment of microplastics in unsaturated substrate: Role of flow regimes, Water Research, 2024.

Yang, Z., Y. Ma, Q. Jing, and Z. Ren, Comigration behavior of Cr(VI) and microplastics and remediation of microplastics facilitated Cr(VI) transportation in saturated porous media, Polymers, 16, 3271, 17 p., 2024.

ANNEX 4: PEARL- ADSORPTION-DESORPTION OF PFAS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	SWAP-PEARL
Process	Soil adsorption and mobility
Pollutant	PFAS and other ionizable substances
Lead partner organisation	WR

UPGRADING-INTEGRATING NEEDS

The PEARL model (Pesticide Emission Assessment at Regional and Local scales) describes the fate of pesticides in the plant-soil system. The sorption process of a chemical to soil is currently defined by an input value for the sorption coefficient to soil organic matter, and can be further specified for additional soil components such as clay and metal-oxides. Charged ionizable organic chemicals (IOCs) will be attracted to oppositely charged soil substrates, and repulsed from equally charged soil substrates (Sigmund et al. 2022). These electrostatic interactions depend on pH and salinity, as well as ionic solutes that may be either competing for sorption sites or rendering charged substrate surfaces more favourable for electrostatic interactions. The pH dependency of the chemical can be included by providing input values for the sorption coefficient of the ionized species, the neutral species and the dissociation constant.

Although this framework is suitable to be used for PFAS and other ionizable species, it lacks detailed insight in the influence of the effects of ionic strength and pH on the speciation of the soil substrates, and the influence this has on the sorption coefficients. Several studies are available to characterize empirical correction factors to account for some of these ionic interactions, as for example described for series of organic cations binding to soil organic matter (Droge and Goss, 2012, 2013a) and different clay minerals (Droge and Goss, 2013b) in presence of different media compositions. Organic anions binding to soil organic matter also has been studied (Tulp et al, 2009), which indicated similar influence of inorganic counterions as has been more recently described for anionic groups of perfluorinated acids, e.g., carboxylate groups of perfluoroalkyl carboxylic acids (PFCAs) (Wang et al., 2022).

The PEARL model should best include the basic mechanistic relationships that influence the chemical-substrate interactions for ionizable chemicals. Just as the soil composition (e.g., organic carbon fraction) and pH profile is specified, or can be adjusted, for different depths in the soil column in PEARL, also ionic strength and co-solutes that influence speciation and binding of target solutes, may be specific for different layers in the soil column. This would allow for more specific local or regional scenarios to be evaluated with a higher level of realism regarding the prediction of mobility of various types of PFAS and other ionizable organic contaminants in soils. The deconstruction of soil sorption into different individual

substrates further poses the challenge of deriving meaningful soil descriptors that account for the contribution of each substrate type (e.g., cation-exchange capacity resulting from both clay minerals and organic matter) and the potential interactions between these substrates (e.g., clay minerals being partially covered by organic matter, reducing access of sorption sites). Example studies have demonstrated that the sorption coefficient of organic cations in different natural soils can be derived from sorption data on pure clay and pure soil organic matter, along with descriptors for the soil (Droge and Goss, 2013c).

Chemicals belonging to the PFAS group are of great environmental concern, and are well known to comprise strong acids such as PFOA and PFOS (both are banned from production sources but are still the most widely distributed persistent PFAS pollutants), which are fully anionic in the typical soil pH range. Analogue structures with similar charged moiety but different chain length exist, creating a substantial range of mobility due to hydrophobic interactions alongside the electrostatic interaction. Longer chains strongly decrease soil mobility but enhanced sorption to biotic components in the soil layers, while shorter chains strongly enhance soil mobility, favouring leaching from soils as well as enhanced uptake into above-ground plant tissues with root water uptake. Depending on the PFAS use or pollution scenario, e.g. fire-fighting foam or water-repellent paper lining, the PFAS chemicals may also contain cationic or zwitterionic functional groups, and a myriad of additional non-ionic features that influence the hydrophobicity driven influence on soil mobility. Not all of the estimated >6000 PFAS structures will have measured sorption coefficients available for modelling the mobility in soils. By evaluating the sorption behaviour of limited series of PFAS structures, the defined contribution of specific molecular fragments may be used to extrapolate of hydrophobic interactions from one analogue PFAS with measured sorption coefficients to other structures within a specific PFAS class. Comparable work on organic cations showed the consistency of this approach for certain molecular fragments, as well as the challenges in deriving sufficient calibration data to support this approach (Droge and Goss, 2013a).

Review of related models

A related model that could prove useful for modelling the sorption and mobility of PFAS on soil is ORCHESTRA (Meeussen, 2003). ORCHESTRA is a computer program used for simulating reactive transport processes and implementing chemical equilibrium models in the soil and water compartments, which specifically accounts for ionization of solutes and soil substrate surfaces, chemical speciation in solution and electrostatic interactions of solutes with soil substrate surfaces. The model includes sorption models such as NICA-Donnan, as suggested in the literature and data gap deliverable 1.2 to be of critical importance for PFAS sorption modelling. ORCHESTRA contains a graphical user interface, which facilitates quick setup of chemical equilibrium models. It consists of two main parts: the first is a chemical equation solver, a flexible tool that calculates chemical equilibria by solving mathematical equations. The solver does not have built-in

chemical rules; instead, it follows instructions given in text-based input files. The second part of the program consists of these same input files, where users define chemical reactions, variables (e.g., pH, metal concentrations) and equations. The solver proceeds with reading these files and using the provided equations to perform calculations (Meeussen, 2003).
ORCHESTRA contains a set of example systems, which are comprised of extensive adsorption models. One of these models is NICA-Donnan, a model used to describe the adsorption and distribution of metal ions onto soil organic matter, accounting for the effects of ionic strength and pH on the binding processes. Modifications on the code of the NICA-Donnan example can be performed such that the model can be applied to PFAS, as they share some similar chemical characteristics with metal ions, such as charge and the effect of ionic strength and pH. Additional code can also be written such that the model accounts for unique chemical properties of PFAS, such as effects of carbon chain length and the fact that some of them are zwitterionic. The advantage of the ORCHESTRA program lies in the fact that customized equations can be added to the model.

Plan for model improvement

The model components from ORCHESTRA will be evaluated for incorporation in the PEARL code. Core expertise with the ORCHESTRA model is available at the Soil Chemistry Group of Wageningen University. Transfer of these ORCHESTRA components into PEARL would require an additional layer of input values and soil system descriptors (e.g., pore water composition) at each layer depth. Examples of soil system descriptors from ORCHESTRA, relevant literature and the soil profile from case-study site will be evaluated for their relevance to construct example scenarios in PEARL. Step-by-step inclusion of additional parameters will be tested to evaluate and describe the influence on soil transport processes.

Model validation

To validate the improvement of PEARL for the specific use case, a set of priority PFAS contaminants will be selected, that represent the main contamination profile in the case study area(s) in connection to the local pollution source (apart from diffuse PFAS pollution). Feasibility to include these chemicals in the foreseen experimental sorption studies will be evaluated. The selected PFAS chemicals will be used in controlled sorption studies on various substrates under a range of different medium conditions. This will result in the delivery of a data matrix (chemical x substrate x medium), that will be used to test the functionality of ORCHESTRA alone, and PEARL model updates, to describe the medium-dependent and substrate-dependent sorption behaviour of ionized PFAS structures in soils.
A soil sample (or a set of samples) from the case study area could be used to perform sorption/desorption studies with a soil composition as close to the reality of the pollution site as possible. However, care should be taken that the

background contamination is not impeding the sorption experiments. This may be achieved in different ways, i.e., by initial desorption conditions (flow through with water only to leach the focal (most mobile) PFAS structures) prior to use in sorption studies, spiking with isotopically labelled PFAS structures, spiking at elevated concentrations, or spiking with alternative IOC chemicals (non-PFAS) used also in the sorption experiments on individual soil substrates.

The influence of organic matter as a key sorbent fraction may be further explored by sequential removal of organic matter from intact natural soil(s), e.g., by acid washing or loss on ignition, with further confirmation on the adequacy of such approaches from representative literature on this topic, as undesired side effects of the removal process may result in co-founding effect on sorption.

The representative soil profile from the VITO case-study site will be defined for a certain area of interest, that would allow for 2D spatial variation of the soil mobility modelling with the aid of the GeoPEARL model. Monitoring data of soil, groundwater and weather conditions of the area over at least 1 year period will allow for more realism in scenarios with variable pollution input schemes (in connection to process 7), including data on land use and plant coverage. The larger regional soil profile may be explored to extrapolate the contamination at the case study site to background contamination of the same PFAS pollution at a larger scale, for which OC content, clay content, metal-oxide content, soil pH, salinity and hardness would be required at a minimum. The presence of metal oxides in soil, as one of the few soil substrates bearing positive surface charge capacity, has been addressed in Deliverable 1.2 as a critical factor to understanding the soil mobility for anionic PFAS, while at the same time this appears to be a substantial knowledge gap. Metal oxide contents in soil profiles in the Netherlands were made available for the first time in a recent study, and this kind of data may not yet be available at the EU scale or at the regional scale of the study site. Therefore alternatives should be decided on. The GeoPEARL model can be used to assess potential scenarios to be elaborated in WP3.

REFERENCES

Droge STJ, Goss K-U. (2012) Effect of sodium and calcium cations on the ion-exchange affinity of organic cations for soil organic matter. Environ Sci Technol, 46(11):5894-901.

Droge STJ, Goss K-U. (2013a) Ion-exchange affinity of organic cations to natural organic matter: influence of amine type and nonionic interactions at two different pHs. Environ Sci Technol, 47:798-806.

Droge STJ, Goss K-U. Sorption of organic cations to phyllosilicate clay minerals: CEC-normalization, salt dependency, and the role of electrostatic and hydrophobic effects. Environ Sci Technol, 2013b;47(24):14224-32.

Droge STJ, Goss K-U. (2013c) Development and evaluation of a new sorption model for organic cations in soil: Contributions from organic matter and clay minerals. Environ Sci Technol, 47(24):14233-41.

Meeussen, J. C. (2003). ORCHESTRA: An object-oriented framework for implementing chemical equilibrium models. Environ Sci Technol, 37(6), 1175-1182.

Sigmund G, Arp HPH, Aumeier BM, Bucheli TD, Chefetz B, Chen W, Droge STJ, Endo S, Escher BI, Hale SE, Hofmann T, Pignatello J, Reemtsma T, Schmidt TC, Schönsee CD, Scheringer M. (2022) Sorption and Mobility of Charged Organic Compounds: How to Confront and Overcome Limitations in Their Assessment. Environ Sci Technol, 56(8):4702-10.

Tülp HC, Fenner K, Schwarzenbach RP, Goss K-U. (2009) pH-Dependent sorption of acidic organic chemicals to soil organic matter. Environ Sci Technol, 43(24):9189-95.

Wang W, Rhodes G, Zhang W, Yu X, Teppen BJ, Li H. Implication of cation-bridging interaction contribution to sorption of perfluoroalkyl carboxylic acids by soils. Chemosphere, 2022;290:133224.

ANNEX 5: PEARL–PLANT UPTAKE OF PFAS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	SWAP-PEARL
Process	Plant uptake
Pollutant	PFAS and other ionizable substances
Lead partner organisation	WR

UPGRADING–INTEGRATING NEEDS

Many contaminant groups can be taken up from the soil by plant roots and accumulate in food crops, which may pose a risk to human health (e.g., Blaine et al., 2014). Integrated soil-plant models are needed to predict accumulation of chemicals in plants. This requires an interdisciplinary approach, as knowledge on both soil physics must be combined with plant physiology. The integration between both fields can be improved. On the one hand, plant models have been developed that describe the transport processes in great detail, such as the models developed by Trapp (2000, 2007) and Trapp et al. (2023). The plant system is discretized into various compartments where accumulation and transport of chemicals is computed. These models take into account processes such as advective transport in the xylem and phloem system, as well as diffusion through and adsorption to cell membranes. However, the soil system is heavily simplified in these models. The chemical concentration in the soil pore water is often assumed to be constant in time and space, while in reality these concentrations can vary greatly. On the other hand, soil models such as SWAP-PEARL (van den Berg et al., 2016) can compute chemical transport in soils in great detail. This model solves the Richard's equation for water flow and the advection-dispersion-reaction equation for solute transport. However, plant uptake of chemicals is simply considered as a sink term and assumed to be linearly proportional to the root water uptake flux through the plant uptake factor. Translocation within the plant is not considered by these models. The description of the soil-plant system can therefore be improved by coupling a detailed plant model with a detailed soil model.

Additionally, plant uptake in soil models is generally focused on neutral compounds. The plant uptake factor is often estimated from the octanol-water partitioning coefficient based on an empirical relationship proposed by Briggs et al. (1983). While this relationship was derived for pesticide uptake in barley, it is widely applied to other crops and contaminant groups. However, this only takes into account lipophilic interactions and is thus only a reasonable approximation for neutral compounds. For ionisable compounds, which includes many types of PFAS, electrostatic interactions must also be considered in plant uptake models. Thousands of PFAS species have been identified, which may show different behaviour regarding plant uptake and translocation. Therefore, it is valuable to

identify the most relevant substance properties that influence plant uptake rate and translocation. Trapp et al. (2023) identified the ionic charge, pKa, and environmental pH-values as most relevant substance properties, but additional parameters such as chain length and head functional groups may be relevant for PFAS.

Review of related models

A version of the Trapp plant model has already been coupled to the HYDRUS model for both neutral compounds (Brunetti et al., 2019) and ionisable compounds (Brunetti et al., 2022). The latter study considered the uptake of five ionisable pharmaceuticals in green pea plants. Plant uptake is dependent on both the substance and crop properties. Therefore, it is valuable to extend the analyses of Brunetti et al. (2022) to other crops and compounds, which will be done within SOILPROM. The plant model considered by Brunetti et al. (2022) is based on the dynamic plant model for neutral compounds of Trapp (2007), but adapted for ionisable compounds based on the cell model of Trapp (2000).
Brunetti et al. (2019, 2022) used Bayesian analysis to estimate values for lumped plant transport parameters from experimental data. Afterwards, the estimated parameters were validated with the cell model of Trapp (2000) to determine whether the estimated parameters are physically realistic. As only the lumped parameters need to be estimated, this method greatly reduces the number of parameters to be estimated. The same approach will also be applied to the SWAP-PEARL model in SOILPROM.
A realistic description of growth of the various plant compartments is required to accurately predict processes such as growth dilution. The growth of the root compartment, including the root distribution and depth, is especially of interest as it directly impacts the uptake of chemicals from soils. The growth of the root system and associated water and solute uptake fluxes can be described in great detail with the CPlantBox model, which is also part of the SOILPROM project. Possibly, upscaled results from the CPlantBox model can be used to improve the description of root water and solute uptake fluxes in SWAP-PEARL.

Plan for model improvement

The plan for model improvement consists of two steps:

1. A dynamic version of the plant model of Trapp et al. (2023) will be developed. Compared to the version of Trapp et al. (2023), number of plant compartments will be flexible, such that various crop types can be simulated. A maximum of four compartments is envisioned (roots, stem, leaves, and fruit). A flexible setup is desired, as not all are appropriate for each crop. For example, leafy vegetables such as spinach do not require a fruit compartment. Additionally, the model will be general such that it is able to simulate uptake and translocation of both neutral and ionisable compounds.

2. The plant model requires a large number of input parameters, many of which are plant- or substance-specific and are difficult to measure experimentally. These parameters could be estimated from the cell model of Trapp (2000). However, including the cell model would further increase the already large number of input parameters that need to be estimated. Instead, the macroscopic plant parameters will be calibrated from experimental data, which are then validated against the cell mode of Trapp (2000). The replacement of octanol-water distribution coefficients as the key descriptor to model plant uptake factors with more suitable partition coefficients (from new experimental work and literature searches) will be evaluated. For example, phospholipid-water partition coefficients have been demonstrated to better predict fish bioconcentration factors (Droge et al. 2019), and cellulose-water partition coefficients may be derived from commercially available chromatographic columns.

The dynamic plant model developed in step 1 will be coupled to the SWAP-PEARL model. Chemical transport in the soil will be simulated in SWAP-PEARL. Root uptake of chemicals in SWAP-PEARL will be adapted for ionisable compounds. The root uptake computed by SWAP-PEARL feeds into the dynamic plant model, which simulates further translocation from roots to other plant compartments. The coupled soil-plant model will be calibrated and validated using experimental data. This will be discussed in more detail in the next section.

Model validation

As the plant model requires a large number of input parameter that are also difficult to measure, it is necessary to calibrate the model for specific crops and substances. This requires detailed measurements of chemical concentrations at various points in time. Plant uptake data in the literature is generally limited to single observation points, such as in terms of the bioconcentration factor (BCF). These factors attempt to relate the concentrations in soil to the concentration in the plant at the time of harvest, but does not take into account the concentration dynamics during the growing season. Information on the dynamics of PFAS concentrations in soil and plant are required for the integrated modelling of PFAS transport. For this reason, pot experiments will be carried out to obtain dynamic plant uptake information. The exact combinations of chemicals and crops that will be considered in the experiments are yet to be determined. Within SOILPROM, we will follow the approach of Brunetti et al. (2019, 2022) and use Bayesian analysis to estimate the input parameters for the plant model. A subset of the experimental data can be used to calibrate the plant model, while a different subset can be used to validate the model.

References

Blaine, A.C., Rich, C.D., Sedlacko, E.M., Hundal, L.S., Kumar, K., Lau, C., Mills, M.A., Harris, K.M., Higgins, C.P., (2014). Perfluoralkyl acid distribution in various plant

compartments of edible crops grown in biosolids-amended soils. Environmental Science & Technology,48(14), 7857-7865. https://doi.org/10.1021/es500016s.

Briggs, G.G., Bromilow, R.H., Evans, A.A., Williams, M., (1983). Relationships between lipophilicity and the distribution of non-ionised chemicals in barley shoots following uptake by the roots. Pesticide Science, 14, 492-500. https://doi.org/10.1002/ps.2780140506

Brunetti, G., Kodešová, R., Šimůnek J., (2019). Modeling the translocation and transformation of chemicals in the soil-plant continuum: a dynamic plant uptake module for the HYDRUS model. Water Resources Research, 55(11), 8967-8989. https://doi.org/10.1029/2019WR025432

Brunetti, G., Kodešová, R., Švecová, H., Fér, M., Nikodem, A., Klement, A., Grabic, R., Šimůnek J., (2022). A novel multiscale biophysical model to predict the fate of ionizable compounds in the soil-plant continuum. Journal of Hazardous Materials, 423, 127008. https://doi.org/10.1016/j.jhazmat.2021.127008

Trapp, S., (2000). Modelling uptake into roots and subsequent translocation of neutral and ionisable organic compounds. Pest Management Science, 56(9): 767-778. https://doi.org/10.1002/1526-4998(200009)56:9<767::AID-PS198>3.0.CO;2-Q

Trapp, S., (2007). Fruit tree model for uptake of organic compounds from soil and air. SAR and QSAR in Environmental Research, 18(3-4), 367-387. https://doi.org/10.1080/10629360701303693

Trapp, S., Shi, J., Zeng, L., (2023). Generic model for plant uptake of ionizable pharmaceuticals and personal care products. Environmental Toxicology and Chemistry, 42(4), 793-804. https://doi.org/10.1002/etc.5582

Van den Berg, F., Tiktak, A., Boesten, J.J.T.I., Van der Linden, A.M.A., (2016). PEARL model for pesticide behaviour and emissions in soil-plant systems: description of processes. Wot-technical report 61.

ANNEX 6: CPLANTBOX– PLANT UPTAKE OF PFAS AND PESTICIDES

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	Cplantbox, HYDRUS, SWAP-PEARL
Process	Water and solute (pesticides, PFAS) uptake by plant roots and transport in the plant
Pollutant	PFAS and other ionizable substances, pesticides
Lead partner organisation	FZJ, WR

UPGRADING–INTEGRATING NEEDS

Uptake via the root system of contaminants is an important process that determines how much pollutants are removed from the soil. This process might be used in phytoremediation. It is also important in exposure assessment since it controls how much of the soil pollution might end up in the food chain. We will upgrade the description of this process by coupling mechanistic approaches that describe the transport of the pollutants in the soil and in the plant, and that resolve the spatial distribution of the pollutant concentrations, of the soil water contents, and the roots in the root zone.

REVIEW OF RELATED MODELS

Models that describe transport in soil-plant systems can be grouped into three classes. The first class of models focuses on transport processes in the plant and considers processes like advection of solutes with the water flow in the apoplast (cell walls) and in the xylem vessels, diffusion across cell membranes, transport driven by electrochemical gradients, sorption in plant tissues (cell walls, lipids), and degradation in the plant tissues. These models have a detailed description of the electrochemical processes in different plant compartments (plant organs but also within cellular compartments like vacuoles) and describe the exchange of contaminants between these plant compartments. However, they do not describe in detail the variation of contaminants in the root zone or within the plant, which is typically represented by a limited number of compartments. These models have been parameterized using experiments in aquaponics or in soil pots with (presumed) uniform distribution of the contaminant. The second class of models focuses on the transport processes in the soil. These models simulate in detail the advective and dispersive transport processes in the soil and how they are affected by sorption and desorption processes to soil particles and degradation in the soil. In these models, the removal of contaminants from the soil by root uptake is described by sink terms. These sink terms are related to the water uptake by the root system and the uptake is equal to the advective flow of the contaminant to the roots which is multiplied by an empirical 'plant uptake factor' (PUF) to account for solute exclusion at the soil-root interface. These models describe the vertical distribution of the contaminants in the root zone and the

leaching out of the root zone but do not describe transport processes within the plant. Water uptake, which drives solute uptake in these models, is described as a function of the soil water content (or potential) and the root distribution but the functions used are empirical and lack a mechanistic basis so that the model accuracy to predict the water uptake distribution when the soil water contents vary with depth in the root zone is debated. In a third class of models, flow and transport in both the soil and plant are described based on mechanistic principles. The most detailed models represent the full 3D architecture of the root system, discretize the root system in a large number of connected root segments, and describe the flow and transport in this network. The root system is coupled with the soil system and flow and transport in the soil towards the root segments is described in 3D. Such models have been developed to simulate water uptake and water flow in the soil and the root system. They have been extended to include also solute transport (nutrient, salts, and pesticides) in the soil to the soil-root surfaces, the transfer of solutes/chemicals from the soil to the root xylem by advection and diffusion, and to include transport of solutes chemicals in the root xylem. With these models, the impact of plant tissue properties, soil properties, root distributions and root architecture, and chemical properties of the solute or contaminant on flow and transport in the soil and the plant are accounted for. The relation between root, water, and contaminant distributions in the root zone, and the water and contaminant uptake distributions emerge from these models and do not have to be parameterized with empirical functions. For instance, root water uptake compensation, hydraulic lift, and enhanced diffusive uptake by dilution are predicted by these models. Although these models are very detailed, they are in fact straightforward to parameterize since realistic root system architectures can be generated using models that are based on observations of root systems, root tissue hydraulic properties can be measured or estimated from anatomical cross sections of roots, and soil hydraulic properties can be measured or derived from other soil information using pedotransfer functions. Therefore, the argument that these models are, because of their high level of detail, over-parameterized is incorrect. However, the high level of detail makes these models extremely computationally expensive and not applicable for simulations of flow and transport processes in practical applications.

Plan for model improvement

The detailed 3D soil-root architecture models can be upscaled making assumptions about the water and contaminant distributions, which are generally assumed to be homogeneous in the horizontal direction. The flow and transport in the soil domain then be reduced to a 1D flow and transport process, which reduces the computational demand drastically when compared to 3D simulations. For water uptake, we have derived approaches to simplify the water flow in the root system and how it is coupled to the soil water flow model (Vanderborght et al., 2024a; Vanderborght et al., 2024b). For contaminant transport in the plant, we have developed 3D transport models that couple the transport processes in the

root system with advective and diffusive exchange with the soil, and transport in the soil (Jorda et al., 2021). However, we still need to investigate how we can simplify the 3D plant model to a simpler 1D model. One approach that was used by Jorda et al. (2021) is to represent the root system by a single 'big root'. Another option is to use a 'parallel root' model that connects all root segments at a certain depth directly to the root collar so that exchange with other soil layers only takes places by advective transport via the root collar. This model was shown to better for upscaling water flow (Couvreur et al., 2012; Vanderborght et al., 2021). But, for upscaling of contaminant transport, the parallel root model cannot account for the dilution that occurs due to uptake of contaminant free water at more distal root parts and that enhances the diffusive uptake in more proximal parts that are in contaminated soil layers. Therefore, it needs to be investigated which model configuration works best for simulating contaminant uptake and how different configurations for water and contaminant uptake can be combined.

Model validation

Since the model upgrades will have an impact, compared to the original models, on the uptake that is simulated when water and contaminants are non-uniformly distributed in the root zone, it is important to carry out experiments in which heterogeneous contaminant distributions can be created and setup in a controlled manner. Setups with soil layers with different contaminant concentrations would be needed. In addition, upscaling approaches can be validated by comparing 3D model simulations with 1D upscaled simulations. Comparisons between 3D and 1D simulations validate the model equations that are used in the upscaled model whereas comparisons between simulations and experimental data validate also the model parameters.

References

Couvreur, V., Vanderborght, J., & Javaux, M. (2012). A simple three-dimensional macroscopic root water uptake model based on the hydraulic architecture approach. Hydrology and Earth System Sciences, 16(8), 2957-2971. https://doi.org/10.5194/hess-16-2957-2012

Jorda, H., Huber, K., Kunkel, A., Vanderborght, J., Javaux, M., Oberdörster, C., Hammel, K., & Schnepf, A. (2021). Mechanistic modeling of pesticide uptake with a 3D plant architecture model. Environmental Science and Pollution Research, 28(39), 55678-55689. https://doi.org/10.1007/s11356-021-14878-3

Vanderborght, J., Couvreur, V., Javaux, M., Leitner, D., Schnepf, A., & Vereecken, H. (2024a). Mechanistically derived macroscopic root water uptake functions: The α and ω of root water uptake functions. Vadose Zone Journal, 23(4), e20333. https://doi.org/https://doi.org/10.1002/vzj2.20333

Vanderborght, J., Couvreur, V., Meunier, F., Schnepf, A., Vereecken, H., Bouda, M., & Javaux, M. (2021). From hydraulic root architecture models to macroscopic

representations of root hydraulics in soil water flow and land surface models. Hydrology and Earth System Sciences, 25(9), 4835-4860. https://doi.org/10.5194/hess-25-4835-2021

Vanderborght, J., Leitner, D., Schnepf, A., Couvreur, V., Vereecken, H., & Javaux, M. (2024b). Combining root and soil hydraulics in macroscopic representations of root water uptake. Vadose Zone Journal, 23(3), Article e20273. https://doi.org/10.1002/vzj2.20273

ANNEX 7: IFDM-(GEO-)PEARL- ATMOSPHERIC DEPOSITION OF PFAS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**IFDM- (Geo-)PEARL**
Process	Atmospheric deposition
Pollutant	PFAS
Lead partner organisation	VITO

UPGRADING-INTEGRATING NEEDS

PFAS are a large class of thousands of synthetic chemicals characterized by carbon-fluorine bonds. They are widely used and are increasingly detected as environmental pollutants contaminating groundwater, surface water and soil. Some are linked to negative effects on human health. Because of their properties, they resist degradation and cleaning up of polluted sites is technically difficult and costly (ECHA, 2025).

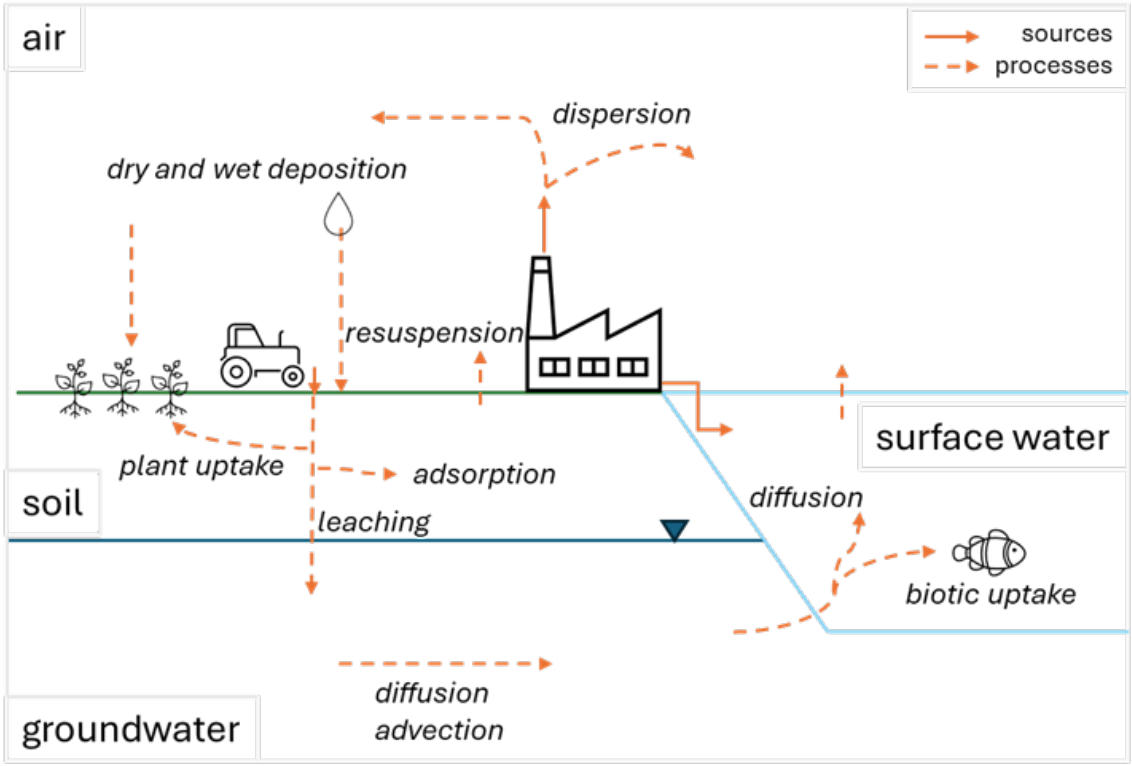

Figure 7. Schematic overview of known PFAS sources and processes controlling their transport and fate. Some known sources are industrial exhausts to the atmosphere, due to manufacturing processes, (industrial) wastewater containing PFAS that is discharged in surface water and fertilizers containing PFAS that are applied directly on plants and soil.

PFAS sources into the environment include industrial exhausts to the atmosphere (for example during industrial manufacturing processes), (industrial) wastewater containing PFAS that is discharged in surface water and fertilizers containing PFAS that are applied on plants and soil. The movement and behaviour of different PFAS components in the environment depend on their physico-chemical properties, as well as the characteristics of the site where they are present (ITRC, 2023).
Atmospheric PFAS inputs to soil and surface water are shown to be an important source and need to be accounted for when modelling soil processes (Young and Mabury, 2010; Schroeder et al., 2021). PFAS does not accumulate in the atmosphere but rather deposits unto the earth surface as dry or wet deposition within a time range of about 10 days (depending on the compound) (Hurley et al., 2004). Wet deposition is influenced by the physico-chemical properties of the specific PFAS compound such as solubility, volatility and Henry's Law constant as well as the molecular size (D'Ambro et al., 2021; Hurley et al., 2004). Dry deposition is mostly driven by the physical PFAS properties such as molecular size as well as the characteristics of the surface where it deposits (Zhang et al., 2001). Additionally, atmospheric factors such as temperature, wind speed, wind direction, concentration gradients and turbulence influence the atmospheric migration of PFAS (ITRC; 2023).
Although models calculating air concentrations and deposition onto soil from point emissions of contaminants are well established and models for transport of contaminants in soil and groundwater as well, a coupled approach is lacking. Nevertheless, a model chain from point source emission to concentration in soil and (ground)water has great value in developing regional soil and water remediation strategies. In a previous study in the Campine area (cross-border area Belgium-the Netherlands) a contaminant transport model for the unsaturated/saturated zone was developed using atmospheric deposition maps of the heavy metals Cd and Zn as input. The model was used to develop a region-wide assessment of the dispersion of Cd and Zn in the soil and water system and to evaluate different strategies to manage the risk of the regional heavy metal contamination (Joris et al., 2014).
The goal of integrating an atmospheric deposition model to a soil contaminant transport model for PFAS is twofold:

- In the case of historical emissions of PFAS into air, the model results can guide monitoring strategies resulting in a more efficient use of resources and can be used to evaluate risk management strategies.
- In the case of an active point source emission of PFAS into air, the model can simulate the full source-path-receptor route and can be used to evaluate the impact of reducing the emission on soil, groundwater and plant concentrations.

REVIEW OF RELATED MODELS

For the calculation of deposition values of PFAS onto soil the IFDM model will be used. The Immission Frequency Distribution Model (IFDM) is developed as a Gaussian model for applications in the framework of Environmental Impact Assessments of point sources. The model calculates concentrations in air and wet/dry deposition starting from meteorological data and emission data and has been shown to perform very well in the applications for point sources in Flanders (Lefebvre and Vranckx, 2013).
For the modelling of PFAS, there are some additional complexities in applying the IFDM-model. Knowledge is required on the PFAS type and plume characteristics of the emissions. Also the physical properties of the PFAS components in the emissions (gas vs particle and its size, conversion, hydrophobicity …) are not always well known. Parameterizing the model for PFAS transport and deposition is still quite novel and remains a challenge.
For the transport of PFAS-compounds in soil the PEARL model will be used. This model solves the Richards' equation for water flow and the convection dispersion equation for solute transport and is one of the regulatory models for pesticide registration risk assessment in the EU. Since the basic principles are the same as the Hydrus model, the model is expected to perform equally well in simulating solute transport. Moreover, in the SoilProm project improvements of the model will be made with regard to PFAS sorption in soil and PFAS plant uptake. Finally, the GeoPEARL model (spatially distributed version of the PEARL model) offers the possibility to apply the 1-D soil modelling to a wider region (in this case the vicinity of the point source of air emission).

PLAN FOR MODEL IMPROVEMENT

The plan for model improvement consists of two phases. The first phase focuses on better constraining PFAS deposition within the atmospheric deposition model to refine predictions of PFAS distribution in the topsoil. The second objective uses these refined deposition estimates to improve the upper boundary condition in a soil contaminant transport model, thereby enhancing the accuracy of model simulations and developing a model chain from emission to concentrations in soil-(ground)water-plant continuum.
In order to refine PFAS deposition estimates, first the parameterization of both dry and wet PFAS-deposition processes will be improved. Specifically, a literature study will be conducted on available PFAS deposition velocities and typical particle size distributions to identify existing knowledge gaps and establish a solid basis for parameterizing deposition mechanisms. Additionally, data from several PFAS measurement campaigns already performed in Flanders will be compiled and analyzed. This analysis will investigate whether component-specific deposition fluxes can be derived from measured airborne PFAS concentrations and whether these insights can inform parameter settings for the IFDM model.

These newly derived parameters will be applied in the IFDM model to simulate component-specific PFAS deposition patterns around a selected point source. The choice of the
site will be guided by both the availability of reliable PFAS emission data and existing measurements of PFAS in soil and groundwater. By incorporating empirically based deposition velocities into the model, this phase aims to produce more accurate, site-specific deposition estimates, enabling a deeper understanding of how PFAS migrates from emission sources into the surrounding environment. The outcomes from these simulations will provide a robust foundation for linking atmospheric deposition models to subsequent soil-leaching and plant uptake assessments.
In the second phase the refined atmospheric deposition outputs will be coupled to the leaching model (GeoPEARL) to more accurately simulate PFAS transport in the soil profile around the selected point source. By setting up a two-dimensional simulation grid, every pixel can use deposition estimates generated by the IFDM model, thereby capturing spatial variations in PFAS-deposition. This approach overcomes the limitations of earlier studies, which often relied on broad assumptions regarding the extent of PFAS deposition impacts around industrial facilities.
Specific aspects of the model integration that will be further investigated are the impact of the spatial and the temporal resolution on the model outcomes and an uncertainty analysis investigating how uncertainty in the atmospheric deposition model parameters affects the final model outcomes.

Model validation

For defining the boundary conditions of the model chain such as meteorological data, both locally available and EU-wide available datasets will be used. The soil parameters will be derived from Flemish databases and EU-wide databases and the sensitivity of the model output to the choice of these parameters will be investigated.
The model chain will be validated using measured values of PFAS deposition and measured concentrations of PFAS in soil and groundwater. A database will be compiled of available data in the use case and based on the analysis it is explored whether additional measurements of PFAS deposition will be carried out. An additional sampling campaign in soil and groundwater will be carried out at different locations from the source and at different depths in the soil profile.

References

D'Ambro, E. L., Pye, H. O. T., Bash, J. O., Bowyer, J., Allen, C., Efstathiou, C., Gilliam, R. C., Reynolds, L., Talgo, K., & Murphy, B. N. (2021). Characterizing the Air Emissions, Transport, and Deposition of Per- and Polyfluoroalkyl Substances from a Fluoropolymer Manufacturing Facility. Environ Sci Technol, 55(2), 862-870. https://doi.org/10.1021/acs.est.0c06580

ECHA (2025). Per- and polyfluoroalkyl substances (PFAS). Retrieved 04/2025 from https://echa.europa.eu/hot-topics/perfluoroalkyl-chemicals-pfas#.

Hurley, M. D., Andersen, M. P. S., Wallington, T. J., Ellis, D. A., Martin, J. W., & Mabury, S. A. (2004). Atmospheric chemistry of perfluorinated carboxylic acids: Reaction with OH radicals and atmospheric lifetimes. J Phys Chem A, 108(4), 615-620. https://doi.org/10.1021/jp036343b

ITRC (Interstate Technology & Regulatory Council). (2023). PFAS Technical and Regulatory Guidance Document and Fact Sheets PFAS-1. Retrieved 02/2025 from https://pfas-1.itrcweb.org/.

Joris, I, Bronders, J, van der Grift, B, Seuntjens, P. (2014) Model-based Scenario Analysis of the Impact of Remediation Measures on Metal Leaching from Soils Contaminated by Historic Smelter Emissions. Journal of Environmental Quality 43(3): 859-868. DOI: 10.2134/jeq2013.07.0287

Lefebvre, W., Vranckx, S. (Eds.) (2013). Validation of the IFDM-model for use in urban applications. Study accomplished in the framework of the ATMOSYS-project, 2013/RMA/R/56, 170 pp.

Schroeder, T., Bond, D., & Foley, J. (2021). PFAS soil and groundwater contamination via industrial airborne emission and land deposition in SW Vermont and Eastern New York State, USA. Environ Sci Process Impacts, 23(2), 291-301. https://doi.org/10.1039/d0em00427h

Young, C. J., & Mabury, S. A. (2010). Atmospheric Perfluorinated Acid Precursors: Chemistry, Occurrence, and Impacts. In P. De Voogt (Ed.), Reviews of Environmental Contamination and Toxicology Volume 208: Perfluorinated alkylated substances (pp. 1-109). Springer New York. https://doi.org/10.1007/978-1-4419-6880-7_1

Zhang, L. M., Gong, S. L., Padro, J., & Barrie, L. (2001). A size-segregated particle dry deposition scheme for an atmospheric aerosol module. Atmospheric Environ, 35(3), 549-560. https://doi.org/10.1016/S1352-2310(00)00326-5

ANNEX 8: ICECREAM–PHOSPHORUS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**ICECREAM (with extended application to SWAT)**
Process	Phosphorus sorption and desorption
Pollutant	Phosphorus
Lead partner organisation	NIBIO

UPGRADING–INTEGRATING NEEDS

The literature review in D1.2 for phosphorus transport from land to water re-confirmed the importance of phosphorus sorption and desorption processes in influencing dissolved phosphorus losses. These processes are the main focus of the work in the Norwegian case study in SOILPROM. We plan to use the ICECREAM model for the work for multiple reasons. First, with its origin in the CREAMS model (Knisel 1980), the ICECREAM model is a field-scale model with a strong capacity to represent actual farming practices. Second, the model has a strong ability to simulate hydrology. It has been modified to be able to simulate water flow under frozen soil and snowmelt conditions of northern Europe (Rekolainen and Posch 1993; Tattari et al. 2001) and simulate macropore flows in addition to matrix flow in clayey soils (Larsson et al. 2007). Third, the model has a wide "audience" of interest. The model has been used in Sweden for calculating national phosphorus losses from agriculture to waters and the sea for many years (Johnsson et al. 2008), and there has been an interest to test/adapt the model in Canada and the United States. In a previous review of water quality models, ICECREAM was identified as the most promising among all for simulating subsurface phosphorus losses (Qi and Qi 2016). Despite the strengths of the model, however, it should also be noted that the model tends to overestimate phosphorus losses due to limitations in the representation of phosphorus sorption and desorption processes in the soil (Liu et al. 2012). This forms another main objective of the Norwegian use case, i.e., improving the representing of soil phosphorus sorption and desorption processes in ICECREAM and other relevant models such as SWAT.

In the ICECREAM model, soil phosphorus is divided into six pools (Fig. 8), i.e., stable inorganic P (PS), active inorganic P (PA), labile inorganic P (PL), manure P (PMAN), fresh organic P (PFO) and slowly mineralizable humus P (PSO). Phosphorus is added to the soil through fertilization with inorganic phosphorus or manure. Phosphorus is lost from the soil through harvest of plant biomass or through losses of particulate-bound phosphorus (PP) and dissolved reactive phosphorus (DRP) with surface runoff and percolating water. Losses of PP through subsurface pathways occur only through macropores, while DRP is transported through both micropores and macropores. All soil phosphorus pools (i.e. PS, PA, PL, Pman, PSO

and PFO) contribute to PP losses. PL is the source of DRP, which can be taken up by plants (P-PLANT) or immobilized into PFO and lost through surface runoff and subsurface drainage. Additions of animal manure are placed into PMAN and mineral phosphorus fertilizer into PL. Phosphorus is transferred to PL from the organic pools through mineralization and from the mineral pools through sorption/desorption processes.

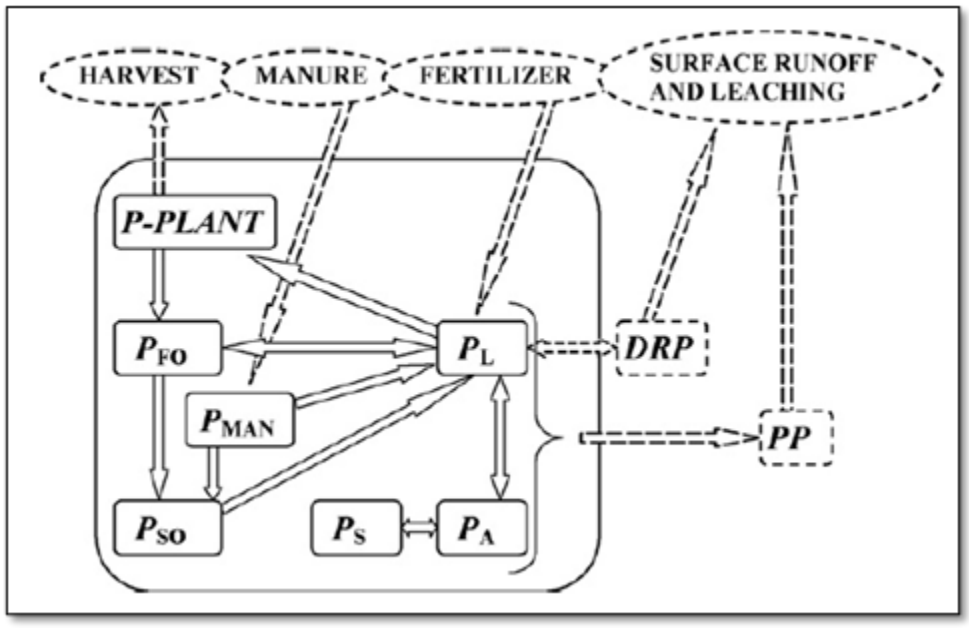

Fig. 8. Phosphorus pools and flows in the ICECREAM model (Larsson et al. 2007).

In ICECREAM and other water quality models for simulating phosphorus loss, a sorption distribution coefficient (kdl) determines the phosphorus sorption/desorption between PL and PA and between PA and PS. There are different ways of calculating kdl (Sharpley and Williams 1990). In ICECREAM, kdl is calculated based on Siimes et al. (1998), derived from 18 long-term field experiments on Finnish soils. kdl is a function of pH (pH), degree of base saturation (bsat), and clay content (solcly).

$$\text{kdl} = 0.0025\,\text{solcly} \cdot (0.46 - 0.0916 \cdot \log(100\,\text{solcly}) + (0.35 - 0.0025\,\text{solcly})(0.0054\,\text{bsat} + 0.116\,\text{pH} - 0.73) \quad (1)$$

The distribution of phosphorus between PL and DRP is described by a linear sorption isotherm assuming instantaneous equilibrium and, with the sorption distribution coefficient (kdw). The coefficient kdw is a function of the clay content (Knisel 1993):

$$\text{kdw} = 100 + 250\,\text{solcly} \quad (2)$$

With the current representation of phosphorus sorption and desorption processes derived from kdl and kdw, the ICECREAM model was found to overestimate total phosphorus leaching by a factor of 5 to 9 (Liu et al. 2012). This

is because it does not have proper routines to represent the high sorption capacity of the subsoil that is caused by iron (Fe), aluminium (Al), and Calcium (Ca) (Liu et al. 2012). Unfortunately, improvement has not been made since the publication of Liu et al. (2012) over a decade ago.

Review of related models

There have already been a couple of good reports that have reviewed progress of phosphorus modelling (Qi and Qi 2016, Wang et al. 2020). For instance, after reviewing a collection of models, Wang et al. (2020) concluded that "soil P routines in many of today's watershed-scale hydrological models are derived from a simple soil and plant P model (Jones et al. 1984) designed for use in the Environmental Policy Impact Climate (EPIC) model (Williams, 1990)." These include some of the world widely used models like SWAT. Although many improvements have been made to these models over the last two decades, the phosphorus sorption-desorption processes have not been sufficiently addressed by considering recent research data. The review work indicates that many water quality models will need further improvement on the representation of phosphorus sorption/desorption processes.

Plan for model improvement

The modelling of phosphorus sorption and desorption processes will be improved by synthesizing relevant recent knowledge. There has been an increasing amount of evidence that the degree of phosphorus saturation in the soil plays a crucial role in influencing dissolved phosphorus loss (Blombäck et al. 2021; Liu et al. 2024). The degree of phosphorus saturation is calculated as the ratio between plant-available phosphorus and phosphorus sorption capacity, on a molar basis. The use of degree of phosphorus saturation has the potential to replace the current routines in the ICECREAM (see section above) for representing phosphorus sorption and desorption capacity in the soil. However, it is not clear yet what is the best way to estimate phosphorus sorption capacity and what is most feasible to be included in farmers routine soil testing. For example, while the sorption capacity was well represented by extractable Fe and Al contents in a Norwegian study (Liu et al. 2024), a maximum phosphorus sorption derived from a single phosphorus addition approach in Sweden was found to best explain CaCl2-extractable phosphorus in the soil (Blombäck et al. 2021) (Fig. 2). Therefore, efforts will be made to synthesize findings from different studies to achieve one or more best solutions for representing sorption and desorption processes in the ICECREAM model. NIBIO will conduct the work in collaboration with SOILPROM's subcontractor the Swedish University of Agricultural Sciences (SLU) who owns the ICECREAM model.

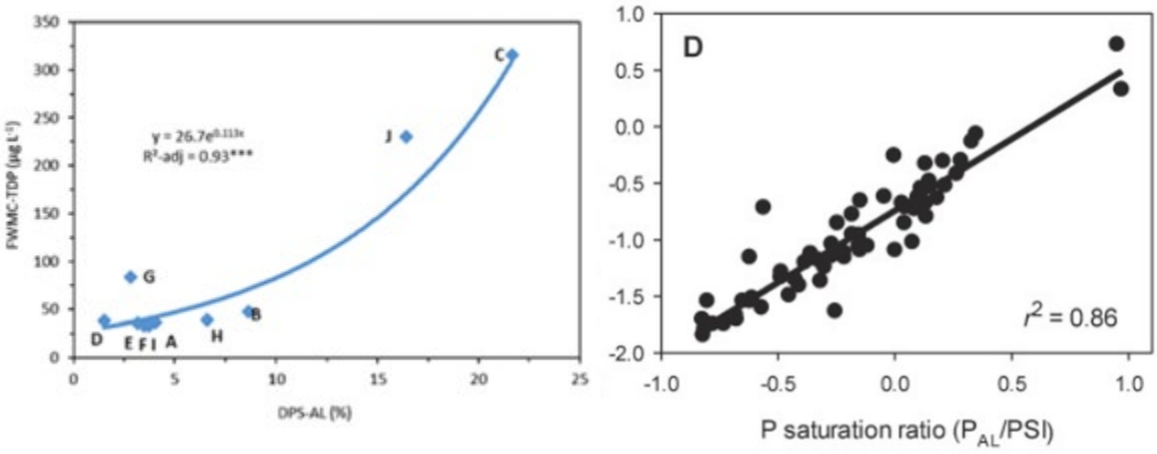

Fig. 9. Relationship between flow-weighted mean concentration of total dissolved phosphorus and degree of phosphorus saturation based on ammonium lactate extraction found in Norway (left), and relationship between CaCl2-extractable phosphorus and degree of phosphorus saturation found in Sweden (right).

Model validation

The new phosphorus routines developed in SOILPROM will be tested in the Norwegian use case using ICECREAM. In the Norwegian use case (Fig. 10), detailed climate and field management data have been collected since 1992 until present. The climate data include air temperature, precipitation, wind and solar radiation. The field management data include dates of sowing and harvesting, type of crop, crop yield, nutrient application date, form, rate and method, and tillage date and method, etc. Various soil physical and hydrological measurements were also made in previous studies. These data serve as valuable inputs for the setup and parameterization of ICECREAM. Water runoff in both overland flow and subsurface drainage were monitored at a 5- or 10-min time-step and water samples were collected for analyses of different forms of phosphorus using a flow-proportional sampling approach during 1992-2005. In addition to other historical data, the farmer has done detailed field sampling in 2011 and 2019 using a grid sampling approach for analyses of plant-available phosphorus and pH, etc. Such data will be available to SOILPROM. In April 2025, NIBIO has also conducted a campaign for collecting soil samples at depths of 0-20 cm, 20-40 cm and 40-60 cm, from the same locations where the farmers have done the sampling before. The samples will be analysed on a series of phosphorus related parameters, including plant available phosphorus, extractable Fe, Al and Ca, texture, pH, maximum sorption capacity, and water extractable phosphorus, etc.

Fig. 10. The Norwegian use case: location in Norway (left), field map (middle), and field photos (right).

The hydrological and water quality data collected from 1992-2005 in the Norwegian use case will be partially used for calibrating the ICECREAM model and partially for validating the model. The performance of the new phosphorus routines to be developed in SOILPROM in simulating phosphorus loss will be compared to the measured results, as well as to the performance of the old (existing) routines. The modelling performance will be assessed using multiple quantitative methods.

After the validation of the new phosphorus routines in the Norwegian use case, they will further be validated in the Polish use case using SWAT. For the description of the Polish use case and SWAT, please refer to other sections of this report.

REFERENCES

Blombäck, K., Bolster, C. H., Lindsjö, A., Hesse, K., Linefur, H., & Parvage, M. M. (2021). Comparing measures for determination of phosphorus saturation as a method to estimate dissolved P in soil solution. Geoderma, 383, 114708. https://doi.org/10.1016/j.geoderma.2020.114708.

Johnsson, H., Larsson, M., Lindsjö, A., Mårtensson, K., Persson, K., & Torstensson, G. (2008). Nutrient leaching from Swedish arable land. Swedish Environmental Protection Board report 5823. http://www.naturvardsverket.se/Documents/publikationer/978-91-620-5823-4.pdf.

Jones, C. A., Cole, C. V., Sharpley, A. N., & Williams, J. R. (1984). A simplified soil and plant phosphorus model: I. Documentation. Soil Science Society of America Journal, 48, 800–805. https://doi.org/10.2136/sssaj1984.03615995004800

Knisel, W. G. (1980). CREAMS: A Field-scale Model for Chemicals, Runoff and Erosion from Agricultural Management Systems. Conservation Research Report No. 26. Washington, DC: USDA.

Larsson, M. H., Persson, K., Ulén, B., Lindsjö, A., & Jarvis, N. J. (2007). A dual porosity model to quantify phosphorus losses from macroporous soils. Ecological Modeling 205, 123–134. https://doi.org/10.1016/j.ecolmodel.2007.02.014.

Liu, J., Aronsson, H., Blombäck, K., Persson, K., & Bergström, L. (2012). Long-term measurements and model simulations of phosphorus leaching from a manured sandy soil. Journal of Soil and Water Conservation, 67(2), 101–110. https://doi.org/10.2489/jswc.67.2.101.

Liu, J., Sævarsson, H. T., Bechmann, M., Krogstad, T., & Øgaard, A. F. (2024). Chemical processes and prediction of dissolved phosphorus leaching in mineral and organic soils. Geoderma, 445, 116890. https://doi.org/10.1016/j.geoderma.2024.116890.

Rekolainen, S., & Posch, M. (1993). Adapting the CREAMS model for Finnish conditions. Nordic Hydrology, 21, 309–322. https://doi.org/10.2166/nh.1993.10.

Sharpley, A. N., & Williams, J. R. (1990). EPIC- Erosion/Productivity Impact Calculator: 1. Model Documentation. USDA Technical Bulletin No. 1768. Washington, DC: USDA.

Siimes, K., Yli-Halla, M., & Tuhkanen, H.R. (1998). Simulation of P cycle in soil by ICECREAM. In Practical and Innovative Measures for the Control of Agricultural Phosphorus Losses to Water, ed. R.H. Foy and R. Dils, 38–39. Poster presented at the OECD Sponsored Workshop, Greenmount College of Agriculture and Horticulture, Northern Ireland, June 16–19, 1998.

Tattari, S., Bärlund, I., Rekoalainen, S., Posch, M., Siimes, K., Tuhkanen, H.R., & Yli-Halla, M. (2001). Modeling sediment yield and phosphorus transport in Finnish clayey soils. Transactions of American Society of Agricultural Engineers, 44(2), 297–307. http://doi.org/10.13031/2013.4691.

Qi, H., & Qi, Z. (2017). Simulating phosphorus loss to subsurface tile drainage flow: a review. Environmental Reviews, 25(2), 150–162. https://doi.org/10.1139/er-2016-0024.

Wang, Z., Zhang, T., Tan, C. S., & Qi, Z. (2020). Modeling of phosphorus loss from field to watershed: A review. Journal of Environmental Quality, 49(5), 1203–1224. https://doi.org/10.1002/jeq2.20109.

Williams, J. R. (1990). The erosion-productivity impact calculator (EPIC) model: A case history. Philosophical Transactions of the Royal Society B, 329, 421–428. https://doi.org/10.1098/rstb.1990.0184.

ANNEX 9: MICROHH-WIND EROSION AND ATMOSPHERIC TRANSPORT OF PESTICIDES, MICROPLASTICS, AND METALS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**MicroHH (microhh.org)**
Process	Wind erosion and atmospheric transport
Pollutant	Pesticides, microplastics (WU) and metals (UPCT)
Lead partner organisation	WU, UPCT

UPGRADING-INTEGRATING NEEDS

MicroHH will be used to model wind erosion and the atmospheric transport of dust-bounded pesticides, metals and microplastics from agricultural areas. This open-source computational fluid dynamics (CFD) model uses Direct Numerical Simulations (DNS) and Large-Eddy Simulations (LES) to model turbulent atmospheric flows (van Heerwaarden, et al., 2017). Given that our domains will range from the field to large-scale applications, LES is preferred over DNS for its balance of computational efficiency and accuracy in representing the physical processes of interest. In LES, larger energy-containing eddies are directly simulated, while the small-scale turbulent motions are parametrised. The plan is to initially run high-resolution field scale simulations to compare the model results to fine-scale field observations. Once validated, WU and UPCT will use larger domains to look at transport of dust-bounded pesticides, metals and microplastics to offsite non-target areas.

Although, MicroHH has been extensively used to simulate turbulent flows in the atmosphere, it requires the integration of physically-based dust emission and deposition modules to simulate the atmospheric transport of dust-bounded pesticides, metals and microplastics. These modules allow for the numerical estimation of dust emission, transport and deposition.

Over the years, various dust emission schemes have been developed and tested in different models at field and regional scales (Shao, 2008). These schemes can generally be categorised into three main types according to their approach to modelling dust emissions: empirical schemes, energy-based schemes and volume-removal schemes. To select the most suitable scheme, it is important to evaluate the underlying assumptions on parameters such as the threshold friction velocity (u^*_t), the horizontal sand flux (Q) and the vertical dust flux (F). Given the dominant focus of SOILPROM on agricultural areas, a physically-based scheme that is adapted and has been applied to cultivated fields could be a good choice. On the other hand, dry dust deposition is governed by gravitational settling and turbulent diffusion, and this is considered by using the vertical settling velocity of soil particles (w) (Shao, 2008).

By integrating dust emission and deposition modules into MicroHH, the model can be extended to simulate the emission and transport of pollutants such as pesticides, microplastics and metals.

After their application, pesticides can bind to soil particles, which through wind erosion, can be released into the atmosphere and subsequently transported by wind (Bento, et al., 2017). Agricultural areas are among the most affected landscapes by wind erosion, as farming practices such as tillage frequently disturb the soil, breaking the structure into smaller, less stable and more easily erodible particles. Accurately modelling pesticide behaviour requires additional key parameters such as adsorption coefficients, which reflect how strongly different types of pesticides bind to soil particles. Another crucial factor is the relationship between the pesticide content and the particle size distribution of a soil sample. Research indicates that certain pesticides (e.g. glyphosate and AMPA) tend to bind to finer soil particles, and that their content diminishes with the increasing particle size (Bento, et al., 2017).

Unlike mineral particles, microplastics vary widely in density, size, and shape, which significantly influence their behaviour in the environment (Koelmans, et al., 2022). Existing dust emission and deposition modules cannot be directly applied to microplastics, as they were developed for mineral particles with different physical properties. For example, the relationships between wind friction velocity and emission fluxes used for dust does not account for the lower density and irregular shapes of microplastics. For these reasons, the dust module developed for pesticides cannot be applied directly for microplastics. As this parametrisation is essential to simulate the emission, transport and deposition of microplastics from the soils into the atmosphere, additional data on key physical parameters such as threshold friction velocity (u^*_t) and the settling velocity (w) of different microplastics is needed. For metals, the same procedure of pesticides transport seems to be applicable as they also get adsorbed to soil particles.

Review of related models

Models that simulate the emission and transport of pesticides, microplastics, and metals in the atmosphere on high spatial resolution are limited. For pesticides transport in the atmosphere, most of the modelling work has focused on spray drift and volatilization processes. When it comes to spray drift, dispersion models such as AGDISP or AgDRIFT are commonly used to model the transport of pesticide droplets though the air (Teske, et al., 2009) (Bilanin, Teske, Barry, & Ekblad, 1989). However, these models are primarily designed for liquid sprays rather than dust-bounded phase and use simplified physics to estimate spray drift.

To model the transport of dust-bounded pesticides and metals, we can learn from existing wind erosion models. These models can enable the simulation and prediction of wind erosion, from particle entrainment, transport and deposition (Shao, 2008). Dust models can be typically divided into empirical and physical

models. Empirical models are based on functions derived from field measurements or wind tunnel experiments for different soil types and surface roughness. In contrast, physical dust models rely on the physical processes that drive the emission, transport and deposition of soil particles, which are driven by not only soil properties but also weather conditions, land use and land management (Chen, Meng, Song, & Zheng, 2022). Physical dust models provide a good overview on dust dynamics at regional and global scale, but they lack the ability to represent the surface processes that drive the emission of dust.

Models for atmospheric microplastics have primarily focused on source apportionment and transport modelling within the urban and marine environments, with limited emission from agricultural soils. For example, Lagrangian particle dispersion models such as FLEXPART, HYSPLIT, MILORD, and LAGRANTO are widely used to trace sources and simulate the long-range dispersion of microplastics (Brahney et al., 2021; Evangeliou, et al., 2020; Martina & Castelli, 2023). However, these models rely on turbulence parametrizations developed for coarse resolution grids and are therefore less accurate in predicting the transport of microplastics at the field scale. Besides, these Lagrangian particle dispersion models lack the emission schemes that account for the uplift of microplastics from soil surfaces.
Computational fluid dynamics models at field scale could provide a more detailed representation of fine-scaled turbulence and near-surface transport, including how pesticide/metal-laden particles and microplastics interact with atmospheric turbulence, saltation, and suspension, which are not considered by currently used larger-scale models. These microscale interactions are essential for predicting how far these pollutants can travel, their concentration at different heights, and potential inhalation risks.

PLAN FOR MODEL IMPROVEMENT

MicroHH requires the integration of physically-based dust emission and deposition modules to simulate the atmospheric transport of dust-bounded pesticides, metals and microplastics from the soil into the atmosphere. The focus will first be on pesticides, as they bind to soil particles which have been extensively studied in terms of emission, transport and deposition. Later, metals will be simulated based on the developed model for pesticides. In contrast, modelling the emission and deposition of microplastics is more complex due to their different physical processes compared to mineral particles. Therefore, WU will focus on simulating the transport of microplastics when the scheme adapted for pesticides and assessed against UPCT data for metals (see also below). A dust deposition module will be further developed by using the settling velocity as a function of the diameter and density of the particles.
Developing an emission scheme that best fits the case, considering its underlying assumptions, parametrizations, and required inputs, such as soil parameters and

climate data is an important step in SOILPROM. Once a suitable scheme is selected, the next step is to implement and test it offline to identify potential errors and evaluate its effectiveness. At this stage, WU and UPCT will also introduce an adsorption coefficient, which describes how certain types of pesticides and metals bind to soil particles. If the scheme performs well, it can be tested semi-online using model statistics. This step is essential to verify whether the scheme behaves realistically before full integration in the model.

Once the semi online tests reveal good results, the scheme can be fully integrated in MicroHH, allowing for dynamic two-way interactions. To test the performance of the scheme in MicroHH, WU will start with uniform idealised cases. These are simplified cases where atmospheric and land properties such as wind are spatially and temporally uniform. The advantages of starting with uniform idealised cases are that it is easier to understand how the scheme behaves since the conditions are controlled and the physical assumptions are correct, and it is easier to identify potential errors and bugs that can be solved before introducing real world conditions.

Once the scheme behaves correctly in idealised conditions, WU will introduce real-world complexities such as heterogeneous surface properties and realistic meteorological forcing based on ERA5 data. Coupling MicroHH to ERA5 ensures that the model accounts for the day-to-day weather variability (van Stratum et al., 2023).

Once these preliminary steps are completed, the next step will be to simulate realistic events at the field scale. This will be done considering the use-cases for the wind erosion and atmospheric transport of microplastics and pesticides in the WU use-case in Valthermond and for metals in the UPCT use-case in Spain. That location and weather conditions will be simulated in the model. The model will be extended then beyond the field scale domain by using open boundary conditions, which allow the dust particles to move outside the edges of the domain. This enables interaction with larger scales atmospheric conditions. This is important to understand how large-scale weather systems affect the transport on more local scales.

Unlike pesticides and metals, microplastics travel as individual particles and have distinct properties compared to mineral particles, including irregular shapes and lower densities. By using data from wind tunnel experiments or field measurements, parametrisations can be introduced to the existing emission scheme for dust-bounded pesticides to account for microplastics. Similarly, the dust deposition module will be adapted by incorporating drag coefficients for both spherical and non-spherical particle to determine the settling velocities.

MODEL VALIDATION

To simulate real-life weather, MicroHH uses several public datasets as input. For meteorological data, we use the ERA5 reanalysis. Despite its coarse resolution, it provides sufficient detail to introduce the large-scale weather patterns into LES. At the surface, we prescribe vegetation properties derived from the Top10NL (10

m resolution) and CORINE (100 m resolution) datasets, as well as soil hydraulic properties obtained from BOFEK2020 (10 m resolution) and SoilGrids (250 m resolution).

To validate the model simulations from the model for WU's specific use-cases, a comprehensive field campaign using multiple instruments will be conducted to gather relevant data. The validation of the model will involve comparing the outputs of the model with observed data to assess how accurate MicroHH was in predicting the spatial distribution of pesticides and microplastics in the atmosphere, as well as their airborne concentrations and their transport fluxes.

BSNE (Big Spring Number Eight) sediment catchers will be installed downwind of the designated field for the WU campaign. These catchers can collect sediments at five different heights over a two-week period during the field campaign. The collected data will provide insights into vertical distribution patterns, which can be compared against the predictions of the model. By gradually increasing the vertical resolution of MicroHH, the ability of the model to accurately simulate spatial distributions, airborne concentrations, and transport fluxes near the emission source can be evaluated.

A high-volume sampler will be deployed to measure airborne particle concentrations at a fixed height of 2.5 meters. This instrument has an omnidirectional inlet and provides direct measurements of airborne concentrations. This can be incorporated in MicroHH. Additionally, data from the RIVM station, which monitors PM10 concentrations, will be incorporated to cross-check airborne PM10 level at different locations.

To obtain information on local meteorological conditions, a sonic anemometer and a meteorological station equipped with a rain gauge, four cup anemometers, and a wind vane will be installed. The instruments will provide data on wind profiles and allow for calculations of friction velocity and aerodynamic surface roughness. These parameters will be compared with the input of the model, particularly the aerodynamic surface roughness, which in MicroHH are related through the Monin-Obukhov similarity theory (MOST). Additionally, measurements of the sensible heat flux, momentum flux, and turbulent kinetic energy will also be analysed to assess whether the effects of turbulence simulated by the model is consistent with observed values.

To validate the large-scale meteorological conditions, which are crucial for accurately capturing the spatial dispersion patterns of pesticides and microplastics, we will use routine weather observations from nearby stations operated by the Royal Netherlands Meteorological Institute (KNMI) and Deutsche Wetterdienst (DWD). These stations provide data like wind speed and direction, as well as fundamental meteorological parameters such as temperature, humidity, and air pressure.

To ensure that MicroHH accurately simulates the emission and transport of metals, a comprehensive validation strategy will be implemented through an extensive field campaign at a heavy metal-polluted site located at the former mining district of Cartagena-La Unión (UPCT use-case). This validation process will involve a

rigorous comparison of model outputs with real-world data to assess the accuracy and reliability of MicroHH in predicting the spatial distribution, airborne concentrations, and transport fluxes of heavy metals in the environment.

The field campaign will be conducted at a designated flat site known for its heavy metal contamination. To monitor the distribution and movement of airborne heavy metals, a combination of multiple instruments and methodologies will be deployed. One key element of the field campaign will be the installation of BSNE sediment catchers downwind from the emission source, specifically in the direction of the dominant winds. These catchers are capable of collecting sediment samples at five different heights. Catchers will be monitored every two weeks during two campaigns in spring-summer 2025 and 2026.

The sediment catchers will provide crucial data on the vertical distribution of dust-bounded metals in the atmosphere, which can then be compared with the model's predictions. This vertical resolution of data will allow for an in-depth evaluation of how well MicroHH simulates the vertical profile of metal concentrations and transport fluxes. These measurements will help assess the model's capability to simulate real-time airborne concentrations under varying meteorological conditions, such as fluctuating wind speeds and directions. Samples collected in the catchers will be analysed by UPCT to characterise properties including soil particle size distribution, pH, salinity and total metal concentrations.

To further enhance the validation process, local meteorological conditions will be monitored by UPCT using data from nearby meteorological stations. These stations will provide essential information on wind speed, direction, and other weather-related parameters that affect the dispersion and transport of pollutants. Wind profiles, in particular, are a critical input for the model's simulation of airborne particle movement. The meteorological data will be used to validate and refine the wind parameters in MicroHH, ensuring that the model's predictions of metal transport are aligned with actual atmospheric conditions.

Discrepancies between the model predictions and field measurements will be used to identify potential model adjustments or calibrations that may improve accuracy. As the field data provides detailed insights into the vertical and horizontal distribution patterns of metals, these will be incorporated into the model through iterative testing and refinements.

References

Bento, C. P., Goossens, D., Rezaei, M., Riksen, M., Mol, H. G., Ritsema, C. J., & Geissen, V. (2017). Glyphosate and AMPA distribution in wind-eroded sediment derived from loess soil. *Environmental pollution*, *220*, 1079-1089.

Bilanin, A. J., Teske, M. E., Barry, J. W., & Ekblad, R. B. (1989). AGDISP: The aircraft spray dispersion model, code development and experimental validation. *Transactions of the ASAE*, *32*(1), 327-0334.

Brahney, J., Mahowald, N., Prank, M., Cornwell, G., Klimont, Z., Matsui, H., & Prather, K. A. (2021). Constraining the atmospheric limb of the plastic cycle. *Proceedings of the National Academy of Sciences*, *118*(16), e2020719118.

Evangeliou, N., Grythe, H., Klimont, Z., Heyes, C., Eckhardt, S., Lopez-Aparicio, S., & Stohl, A. (2020). Atmospheric transport is a major pathway of microplastics to remote regions. *Nature communications*, *11*(1), 3381.

Martina, M., & Castelli, S. T. (2023). Modelling the potential long-range dispersion of atmospheric microplastics reaching a remote site. *Atmospheric Environment*, *312*, 120044.

Koelmans, A. A., Redondo-Hasselerharm, P. E., Nor, N. H. M., de Ruijter, V. N., Mintenig, S. M., & Kooi, M. (2022). Risk assessment of microplastic particles. *Nature Reviews Materials*, *7*(2), 138-152.

Shao, Y. (2008, September). Progress and Challenges in Dust Emission Theory and Schemes. In *3rd International Workshop on Mineral Dust. Leipzig, Germany, University of Cologne*.

Teske, M. E., Thistle, H. W., & Londergan, R. J. (2009). Considerations of time step and evolving droplet size in the simulation of fine droplet motion using AGDISP. In *2009 Reno, Nevada, June 21-June 24, 2009* (p. 1). American Society of Agricultural and Biological Engineers.

van Heerwaarden, C. C., van Stratum, B. J. H., Heus, T., Gibbs, J. A., Fedorovich, E., & Mellado, J. P. (2017). MicroHH 1.0: A computational fluid dynamics code for direct numerical simulation and large-eddy simulation of atmospheric boundary layer flows. *Geoscientific Model Development*, *10*(8), 3145-3165.

van Stratum, B. J. H., van Heerwaarden, C. C., & Vilà-Guerau de Arellano, J. (2023). The Benefits and Challenges of Downscaling a Global Reanalysis With Doubly-Periodic Large-Eddy Simulations. *Journal of Advances in Modeling Earth Systems*, *15*(10), e2023MS003750.

ANNEX 10: OPENLISEM- WATER EROSION AND RUNOFF-PESTICIDES AND METALS

Plan for model upgrade and validation

MODEL AND PROCESS INFORMATION

Model	**OpenLISEM**
Process	Water erosion and runoff
Pollutant	Pesticides and metals
Lead partner organisation	WU and UPCT

UPGRADING-INTEGRATING NEEDS

The following upgrades and coupling to the OpenLISEM-Pesticide model (OLP, Commelin et al., 2024) have been identified in D1.2:

- The current version (Commelin et al., 2024) of OLP works with 1D kinematic wave flow descriptions. However, the OpenLISEM base model (i.e. the runoff and erosion model) has a 2D dynamic wave option that is better, especially for simulating floods. The 2D dynamic wave option for pesticides transport will be tested.
- The testing and prediction of Best Management Practices (BMPs) that reduce the erosion, transport and deposition of pesticides with overland flow and sediment transport has not yet been conducted.
- As OpenLISEM is an event-based model, it needs initial conditions at the start of the rainfall events for soil moisture and the concentration of pesticides in the soil. To enable good estimation of these initial conditions OpenLISEM-pesticide will be coupled with a soil water transport model such as SWAP and/or PEARL.

REVIEW OF RELATED MODELS

As far as we are aware of, there is no other model that simulates the transport of pesticides with both runoff and water erosion (dissolved and sediment-bounded) on this small temporal scale of individual rainfall events, and that is spatially explicit, i.e. uses a grid-based approach. Several well-known models have been thoroughly tested and evaluated on pesticide leaching, degradation in the soil and transport with runoff, including PRZM, FOCUSPEARL, RZWQM and SWAT (DeMars et al., 2018; Purnell et al., 2020; Van den Berg et al., 2016; Young and Fry, 2019). In Figure 11 some commonly used pesticide fate models for overland flow are compared on the inclusion of concepts relevant for the simulation of pesticide transport during rainfall-runoff events, such as spatial representation (i.e. homogeneous representation of the area, semi-distributed or as raster cells), temporal scale and, with a focus on transport of pesticides with detached sediment, if erosion is included in the model and whether or not pesticide

transport in sediment-bounded form is taken into account. However, the spatial representation (homogenous or hydrologic response units) and temporal scale (minimal timesteps of hours or days) of these models is often not targeted at event scale dynamics. Besides these, other models have been developed, aiming at specific topics of pesticide fate. For example, ZinAgriTra analyses the transport of transformation products of pesticides (Gassmann, 2013) and VFSMOD was specifically designed to predict the effect of vegetative filter strips to trap pesticides (Muñoz-Carpena et al., 2018). The GSSHA model does have a promising combination of model concepts (Pradhan et al., 2014), however very little prior research or information on applications for pesticides transport was found. In a recent review, 17 different models were identified that were used in the past decade to simulate the fate and transport of pesticides (Centanni et al., 2023). In most of these modelling studies sediment-bounded transport of pesticides is not taken into account due to lack of data or model limitations (e.g. Gassmann, 2013; Purnell et al., 2020; Young and Fry, 2019). When sediment-bounded transport was included, the model performance in terms of sediment transport is reported as not adequate (e.g. Chen et al., 2017; DeMars et al., 2018). Dynamics within the runoff event and different contributing areas could not be simulated with these lumped edge-of-field models.

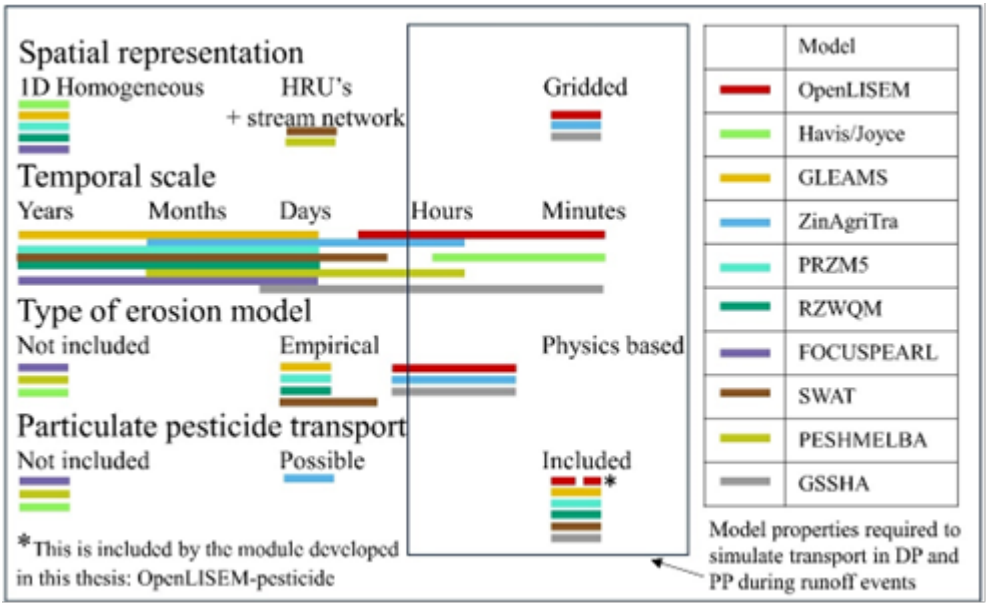

Figure 11. Overview of model concepts of some commonly used pesticide fate models Source: Commelin, 2024. Model concepts compared include spatial representation (i.e. homogeneous representation of the area or semi-distributed or based on gridded raster cells), the temporal scale considered, if erosion is included and if so, which type of model is considered and if particulate phase transport of pesticides is taken into account. HRU = Hydrological Response Unit; DP – Dissolved Phase; PP = Particulate Phase.

Plan for model improvement

To improve the model with regards to the items mentioned in the first section, the following will be done:

- Using the already available data (Commelin et al., 2023) and the acquired new data in the WU use-case for pesticides, the existing model will be adapted for additional calibration of both the runoff, sediment/erosion and pesticides. The model will be further calibrated and validated by UPCT for metals (OpenLISEM-Metals model).
- Developing and testing 2D Dynamic flow can be done using either the existing data and/or the newly acquired data. The erosion and sediment has not yet been tested with this flow routing algorithm. Then, the simulation of the pesticides with both overland flow (dissolved phase) and erosion (sediment-bounded) need to be tested using the 2D Dynamic wave approximation and OpenLISEM-Pesticide needs to be re-calibrated using this flow routing approach.
- Some options to reduce runoff and erosion are available in the base OpenLISEM model. The effect of such measures to reduce pesticide transport has not been evaluated. The existing measures can be adapted to try and simulate reduction of pesticides, but, as far as we know, hardly any specific data exists to validate such simulation results. However, for especially vegetative filter strips (VFS), literature with some data is available (Reichenberger et al., 2019), so this could be used to verify OpenLISEM-Pesticide simulations of VFS effects. Another option is the simulation of scenarios of land use/cropping pattern change as BMP, or the reduction of pesticide use in a certain cropping pattern, to evaluate the effects of such BMPs on the transport of pesticides.
- Coupling of OpenLISEM with SWAP will be further developed, using the first set-up of WU to transfer SWAP output to OpenLISEM-Pesticide input.

Model validation

Pesticides. The model improvement (see previous sections) can make use of the already existing dataset, as measured by Commelin et al. (2023) and which is publicly available. This mainly consists of measured discharge (l/s), sediment (g/l) and pesticide concentration at the small catchment outlet.
In addition, in the WU use-case, similar measurement campaign will be continued in WU use-case in 2025. Therefore, a more extensive dataset will become available for model validation. This also includes information from the farmers on pesticides application in the use-case.
OpenLISEM uses some basic input data such as a DEM, soil type and land use maps. However, the specific OpenLISEM input data still has to be either derived from this data or measured in the field. It is up to the user which methods is used, with more specific methods (e.g. field measurements, or availability of local data)

giving better datasets than using globally available data. In WU use-case, field and local data will be collected for a better validation.

Metals. The validation of the OpenLISEM-Metals model requires comprehensive field data on soil erosion, sediment-bound metal concentrations, and relevant climatic variables such as rainfall amount and intensity. For this purpose, data will be collected from a drainage catchment located within the former mining area of Cartagena-La Unión (UPCT use-case). This area has been extensively impacted by past mining activities, and several ephemeral watercourses drain from the mining sites toward lower-lying zones.

For the evaluation of sediment retention, selected sedimentation dams constructed along the main watercourse of a representative catchment will be studied (Castillo et al., 2007; Romero-Díaz et al., 2007). Erosion nails will be installed in the selected dams to measure sediment elevation changes (i.e., the difference in height of accumulated sediments) after each erosive event over an 18-month monitoring period.

A known volume of accumulated sediment will be sampled and weighed in order to estimate the total amount of sediment eroded from the source catchment as well as the expected concentration of metals. The collected sediment samples will be analysed for metal content, texture, pH, and salinity. Using the measured metal concentrations and the estimated sediment quantities, the total amount of metals transported via water erosion will be calculated and expressed relative to the area of the source catchment.

The necessary input data from the source catchment will be gathered from existing databases and through direct field measurements. This dataset includes a land use/vegetation map, slope map, leaf area index, vegetation cover and height, root cohesion, soil cohesion, soil porosity, aggregate stability, soil particle size distribution, and stone cover, among other parameters.

Climatic data from local meteorological stations will be used to determine the amount and intensity of each rainfall event. Meteorological data will be obtained from nearby weather stations and will include total rainfall, peak rainfall intensity, rainfall duration and storm distribution. These climatic variables are essential for accurately simulating hydrological responses and erosion dynamics within the catchment.

The collected data will serve as the primary input for model calibration and validation. Erosion rates derived from erosion nails, along with soil and sediment characterization and rainfall data, will be used to assess the model's ability to simulate soil erosion.

Specifically, model validation will involve comparing predicted soil erosion rates with those measured in the field after rainfall events. This will allow an evaluation of the model's accuracy in representing soil loss under varying meteorological and land cover conditions. Additionally, measured metal concentrations in sediments collected after erosive events will be compared with model outputs to assess its performance in simulating metal transport associated with erosion.

REFERENCES

Castillo Sánchez, V. M., Mosch, W., Conesa García, C., González Barberá, G., Navarro-Cano, J. A., & López-Bermúdez, F. (2007). Effectiveness and geomorphological impacts of check dams for soil erosion control in a semiarid Mediterranean catchment: El Cárcavo (Murcia, Spain). CATENA 70: 416-427.

Commelin, M. C., Baartman, J. E. M., Wesseling, J. G., & Jetten, V. (2024). Simulating event-based pesticide transport with runoff and erosion; OpenLISEM-pesticide v.1. Environmental Modelling and Software, 174. https://doi.org/10.1016/j.envsoft.2024.105960

Commelin, M., Baartman, J. E. M. (J., Peters, P., Zomer, P., Riksen, M. J. P. M. (M., & Geissen, V. (2023). Runoff, erosion and pesticide transport in a small catchment in Limburg - The Netherlands (Version 3) [Data set]. 4TU.ResearchData. https://doi.org/10.4121/19690684.V3

Commelin, M. C. (2024). Pesticide transport during erosive rainfall-runoff events [Dissertation]. Wageningen University.

Reichenberger, S., Sur, R., Kley, C., Sittig, S., & Multsch, S. (2019). Recalibration and cross-validation of pesticide trapping equations for vegetative filter strips (VFS) using additional experimental data. Science of the Total Environment, 647, 534–550. https://doi.org/10.1016/j.scitotenv.2018.07.429

References in the text that was taken from the PhD thesis of Commelin:

Centanni, M., Ricci, G.F., De Girolamo, A.M., Romano, G., Gentile, F., 2023. A review of modeling pesticides in freshwaters: Current status, progress achieved and desirable improvements. Environmental Pollution 316, 120553. https://doi.org/10.1016/j.envpol.2022.120553

Chen, H., Zhang, X., Demars, C., Zhang, M., 2017. Numerical simulation of agricultural sediment and pesticide runoff: RZWQM and PRZM comparison. Hydrol. Process. 31, 2464–2476. https://doi.org/10.1002/hyp.11207

DeMars, C., Zhan, Y., Chen, H., Heilman, P., Zhang, X., Zhang, M., 2018. Integrating GLEAMS sedimentation into RZWQM for pesticide sorbed sediment runoff modeling. Environmental Modelling and Software 109, 390–401. https://doi.org/10.1016/j.envsoft.2018.08.016

Gassmann, M., 2013. Environmental fate modelling of agrochemicals and their transformation products at catchment scale.

Muñoz-Carpena, R., Fox, G.A., Ritter, A., Perez-Ovilla, O., Rodea-Palomares, I., 2018. Effect of vegetative filter strip pesticide residue degradation assumptions for environmental exposure assessments. Science of the Total Environment 619–620, 977–987. https://doi.org/10.1016/j.scitotenv.2017.11.093

Pradhan, N.R., Downer, C.W., Johnson, B.E., 2014. A Physics Based Hydrologic Modeling Approach to Simulate Non-point Source Pollution for the Purposes of Calculating TMDLs and Designing Abatement Measures, in: Leszczynski, J., Shukla, M.K. (Eds.), Practical Aspects of Computational Chemistry III. Springer US, Boston, MA, pp. 249–282. https://doi.org/10.1007/978-1-4899-7445-7_9

Purnell, S., Kennedy, R., Williamson, E., Remesan, R., 2020. Metaldehyde prediction by integrating existing water industry datasets with the soil and water

assessment tool. Water Research 183, 116053. https://doi.org/10.1016/j.watres.2020.116053

Romero-Díaz, A., Alonso-Sarriá, F., & Martínez-Lloris, M. (2007). Erosion rates obtained from check-dam sedimentation (SE Spain). A multi-method comparison. Catena, 71(1), 172-178.

Van den Berg, F., Tiktak, A., Boesten, J., Van der Linden, A., 2016. PEARL model for pesticide behaviour and emissions in soil-plant systems. Statutory Research Tasks Unit for Nature & the Environment.

Young, D.F., Fry, M.M., 2019. Field-scale evaluation of pesticide uptake into runoff using a mixing cell and a non-uniform uptake model. Environmental Modelling & Software 122, 104055. https://doi.org/10.1016/J.ENVSOFT.2017.09.007