SOILveR

Soil and land research funding platform for Europe

PREMISS : Executive Summary

Priorisation of emerging chemical compounds in soils

Acknowledgments

To Soilver funders

How to refer to this report

C. Merly, A; Togola, E. Bouhoulle, R. Van der Meiracker, J. Lijzen, L. Oste, P. Van Breemen, H. Passier, B. Lopez, R. Cartuyvels, K. Van Geert and S. Willems. PREMISS Executive summary: Priorisation of emerging pollutants in soils, March 2022 . BRGM/RP-71530-FR.14p.

This report is available on https://www.soilver.eu/

Project PREMISS is supported by:

Project leader:

Bureau de Recherches Géologiques et Minières

Project members:

Funded by:

SOILveR in brief : The SOILveR platform strongly believes in the need for integrated soil and land research and knowledge exchange in Europe. We acknowledge the added value of coordinating, co-funding and disseminating cross- border soil and land management research. SOILveR is a self-financed platform. The platform members have a common interest in sharing and implementing integrated multidisciplinary research. SOILveR builds on the experiences from other funding networks such as SNOWMAN and address knowledge needs identified by e.g. the Horizon 2020 project INSPIRATION and other initiatives as well as those proposed by the members of SOILveR.

Context and Objectives of PREMISS

Contaminants of Emerging Concerns (CECs) are raising increasing attention for the last few decades in the water media. CECs were officially considered for the monitoring of surface water in 2015 (Directive 2008/105/EC of the European Parliament) and the CECs watch list is currently in elaboration for groundwater under voluntary action of Member States. As for soil, besides some European initiatives (as for example LUCAS soil quality monitoring survey, International network on Emerging soil contaminants and in soil (EmConsoil), etc.) hardly any attention has been paid on CECs for soil. However, some CECs, such as the PerFluorinated Alkylated Substances (PFAS) in soil have recently raised serious concerns for the industries and the contaminated land communities (CONCAWE, NICOLE). CECs in soil is becoming a subject of high interest, which is proposed to be tackled in the newly launched Healthy Soil strategy (November 2021), which aims to develop, by 2024, an EU priority list for contaminants of major and/or emerging concern that pose significant risks for European soil quality, and for which vigilance and priority actions at European and national level are needed.

Responding to CECs increasing concerns in soil, the R&D PREMISS (PRioritisation of EMerging chemical compounds In SoilS) project aimed to explore approaches to prioritise CECs in groundwater and subsoil and to propose associated recommendations for CECs management, regulation and R&D actions. The PREMISS project was a one year project (2021) funded by the SOILVER platform (including ADEME, Ministry of IenW, SPW-ARNE and OVAM) and was performed by a French (BRGM – coordinator), Belgium (ISSeP, Arcadis and Witteveen&Bos) and Dutch (Deltares and RIVM) R&D team.

The PREMISS project was built on several experiences from prioritisation of CECs in water compartments: such as the need to compile and share existing data, the necessity to deal with CECs data lacking, the capitalisation of existing CECs prioritisation tool developed for water.

PREMISS developed CECs prioritisation approach through the following actions:

- Make an inventory of CECs occurrence data in soil, sludge from wastewater treatment plants (WWTP), sediments and groundwater available for the Netherlands, Belgium and France. This enabled to assess which groups of substances were monitored and to select pilot families and compounds to be tested in the prioritisation prototype.
- Estimate CECs concentrations in soil based on substance physico-chemical properties, substance emission and substance migration pathways with a fate and transport module focussing on the soil/solid compartment.
- Estimate the soil concentration threshold at which the substance may pose a risk to ecosystems or human health by developing a toxicological module based on derived toxicological data and exposure equation.
- Design a prioritisation prototype using a Source-Pathway-Receptor approach and rating the substances based on the ratio of substance soil concentration (measured or estimated) and the derived substance soil concentration threshold.
- Co-design the prioritisation prototype based on stakeholders' demands on CECs prioritisation and draw recommendations from PREMISS outcomes from stakeholders' engagement.

PREMISS methodology and outcomes are summarised below.

Inventory of existing data on CECs occurrence in soil, groundwater and sources to soil (sludge, manure), led by ISSeP

First of all, CECs were defined as substances that are not regulated in the environmental matrices of interest at the national or EU level. Therefore, CECs *did not* include BTEX, PAHs, PCBs, dioxins, furans, solvents and inorganic substances (for which fate is rather different to estimate than organics). As the CECs include a high number and diversity of substances, it was decided in the timeframe of the PREMISS project to undertake the inventory on a selection of four CECs categories. These categories were chosen from the 11 categories defined by Bunting et al. (2021)[1] and correspond to substances that were relevant for the Source-Pathway-Receptor conceptual schemes which were tested in the prioritisation prototype: a) WWTP sludge/pesticides application on agricultural soil, b) Sediment application on soil, c) Industrial emissions. The four selected categories of CECs included 1) PFAS, 2) phenols & alkyphenols, 3) pesticides and 4) pharmaceuticals & veterinary products. Occurrence data of CECs in soil, sediments, WWTP sludge, and groundwater were collected from national reports, scientific literature (only for soil) and national databases (DB). The inventory was in principle limited to occurrence data measured from 2010 until the end of 2020, in France, the Netherlands, the Flemish or the Walloon regions in Belgium. However, some exceptions were made when occurrence data were already aggregated for a larger geographic zone (e.g. JRC, 2012) or a larger time scale (e.g. TNO published Dutch soil background concentrations in 2004).

The inventory was undertaken with various levels of detail depending on the targeted substances/groups of substances:

- **Overall listing of databases, reports and scientific literature were undertaken for the CECs of the 4 selected CECs categories**. A distinction was made between data reflecting background concentrations, referred to as "Global monitoring", and concentrations measured near risky activities or potentially contaminated sites referred to as "Point source". Twenty databases and reports were found with global monitoring data and 12 with point source data, for a total of 32 including CECs occurrence data. Data in soil are scarce. Only 6 of the 20 global monitoring sources were related to the soil matrix. On the contrary, most of the point source databases and reports (9 out of 12) referred to the soil matrix. Soil, sediment, groundwater and sludge data were scarce for phenols & alkyphenols, pesticides and pharmaceuticals for France, the Netherlands and Belgium. Altogether, it encompasses 46 substances. As for the PFAS, we observed two contrasted situations: there was hardly any PFAS data in French and Walloon databases, while numerous PFAS data were recently acquired in Flanders and the Netherlands. For PFAS, 31 substances were inventoried. The number of samples and the quantification frequencies[2] were listed in soil, groundwater, sediments, and/or sludge for the 77 substances covered by the inventory of the four selected CECs categories. This allowed a first overview of the available data for CECs in Belgium, France and the Netherlands. However, the quantification limits (QL) for some CECs were heterogeneous and sometimes very high, making the exploitation of the data difficult (eg data comparison or compilation). Therefore, the measured concentrations (absolute values) were deemed to be more valuable information that quantification frequencies.
- **Complete inventory of existing data for a selection of 18 "pilot" substances.** The previous global inventory identified 77 substances for the four categories, which still represented a very large number of substances to be working on in the one-year PREMISS project. Consequently, it has been necessary to further limit the complete inventory to a panel of substances among the 77 identified. A set of 18 "pilot" substances (7 PFAS, 2 phenols & alkylphenols, 5 pesticides and 4 pharmaceuticals & veterinary products) was selected based on physico-chemical properties and data availability and served as showcase for a demonstration of the prioritisation tool to be developed in PREMISS. Complete inventory enabled to assess substance occurrence in each media and to compare dataset

[1] Chemical intermediates; Flame retardants; Lifestyle; Personal care products (PCPs); Pesticides (including biocides); PFAS; Pharmaceuticals (for humans and animals); Phenols & Alkylphenols; Plasticisers; Solvents and trihalomethanes (THMs); Other CECs

[2] The quantification frequency in a matrix is defined as the percentage of samples where the substance was quantified divided by the total number of measures in that matrix.

among countries. For the PFAS, PFOA and PFOS were the substances for which there were more data and the highest concentrations observed. 4-Nonylphenol (branched mixture, #CAS 84852-15-3) was found in most sewage sludge samples listed in Belgium, France and the Netherlands with quantification frequencies between 57% (FR) and 98% (BE-WAL). As for pesticides, glyphosate was rarely found in groundwater. As for pharmaceuticals, diclofenac was found in most sewage sludge with quantification frequencies ranging from 71% to 100% in global monitoring data.

From this inventory, several observations can be taken up as recommendations at national levels and EU level, which include:

- **Harmonisation** of the associated **metadata** for national database (CAS numbers, unit, QL, context) online with achieving FAIR[3] data treatment.
- **Acquisition of CECs data** in soil, sediment and sludge supported by a coherent EU regulatory framework. Besides PFAS for Flanders and the Netherlands, selected CECs in PREMISS were hardly covered by existing French, Belgium and Dutch databases.
- Need to **centralize data storage** for **WWTP solid effluents,** manure, compost and digestate.
- **Development of a harmonised** EU (or in the meanwhile national) **CECs database for soil**.

Overarching concept of the prioritisation approach

PREMISS aimed to design of **a robust and flexible** prototype to prioritise CECs in soil and sub-surface, to which updates and improvements could be made in the future, such as substance and data addition. To do so, the prioritisation prototype used practices from risk assessment (RA) to prioritise a large number of substances. In RA, the risk is determined by combining the substance concentration actually present in the environment (estimated or measured) with the effect concentration of the substance on a receptor (human, ecosystem), according to a set Source-Pathway-Receptor scheme. The prototype tool consisted of the following steps:

- ***Selection of relevant Source-Pathway-Receptor (S-P-R) scheme***. Prototype development was based on three main S-P-R schemes encompassing 1) Industrial emissions to the environment (for PFAS and phenols & alkyl phenols); 2) WWTP sludge (pharmaceuticals) or other materials (manure, sediment) application on rural soil; 3) Direct pesticides application on agricultural land.
- ***Assessing which CECs are likely to be present in soil and subsoil,*** from measured data (see the inventory section above) or from calculated concentration (see details in the section below).
- ***Evaluation of CECs toxicity*** – see the development of toxicological module below.
- ***Evaluating which of these CECs may pose higher risks for human health and/or the environment***. This risk being a combination of the fate of the substance in the environment and the toxicity of the substance (expressed as Risk Characterisation Ratio (RCR): the quotient of the fate and toxicity) for each relevant receptor (human beings, ecological receptors, etc.).

The prioritisation approach can be illustrated as follows :

[3] In 2016, the 'FAIR Guiding Principles for scientific data management and stewardship' were published. They intend to provide guidelines to improve the **F**indability, **A**ccessibility, **I**nteroperability, and **R**euse of digital assets by both humans and machines.

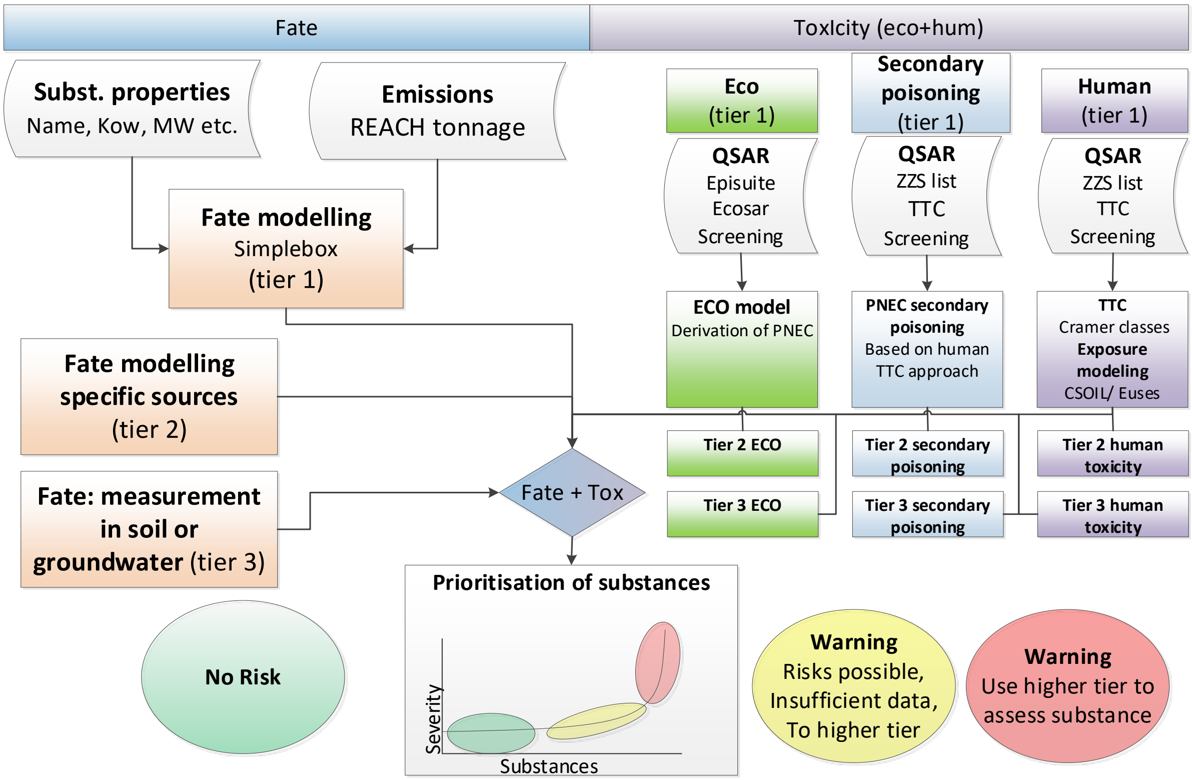


Illustration of the prioritisation approach including second and third tier prioritisation steps.

Moreover, as the demands on data quality in RA are high and require extensive quality evaluation, the RA process can be laborious and time-consuming. Therefore, it is important to prioritise those substances for which elaborating extensive RA is of added value. This can be done by splitting the prioritisation into three tiers. This implies that first a large number of substances can be assessed and that further analysis of compounds can be performed for substances with higher priority. The following tiers are proposed.

- Tier 1: Basic prioritisation with generic emission, fate modelling, and toxicity data;
- Tier 2: Prioritisation with additional national or regional data on emissions, more detailed fate modelling (Predicted Environmental Concentration - PEC) and/or experimental toxicity;
- Tier 3: Risk assessment based on measurements in exposure media and/or appraised toxicity data.

The prioritisation approach which was developed in PREMISS aimed to be useful and meaningful for foreseen users. This is why PREMISS promoted stakeholders' involvement over the course of the project by engaging with them twice. A couple of meetings was held with actors who may have to deal with CECs in soil and sub-surface (including problem owners (site managers, natural resources managers, industries), regulators, service providers (consultants, contractors and laboratories), funders and researchers) in order to 1) at the beginning of the project draw what were the expectations and needs from stakeholders on prioritisation of CECs in soil and 2) at the end of the project have stakeholders' feedback from PREMISS project output and exploitation.

Stakeholders' demands on prioritisation of CECs in soil are summarised as follows:

- CECs, not being part of a normative system and gathering many substances, prioritisation was deemed useful and necessary by the stakeholders, as it enabled to develop actions (regulatory, monitoring and remediation) from the most problematic substances (to the less), to save time, and to be more cost-effective.
- Main demands on prioritisation include, 1) accelerating prevention and regulation, 2) gaining insight on risk assessment, 3) developing a robust management approach and 4) identifying knowledge gaps.
- Stakeholders also raised specific demands on topics such as scenario types (S-P-R conceptual scheme), substances (sources and emissions), toxicity/risk assessment. For each of the specific demand, it was indicated whether or not the demand could be included in the PREMISS prioritisation approach or not.

Assessment of which CECs are likely to be present in soil and sub-soil based on fate and transport module (led by Deltares)

The first step of the prioritisation approach is to estimate the concentration of CECs in soil and sub-soil in order to assess which substances are likely to be present in these media. Two tiers were proposed to evaluate expected concentration in soil and sub-soil.

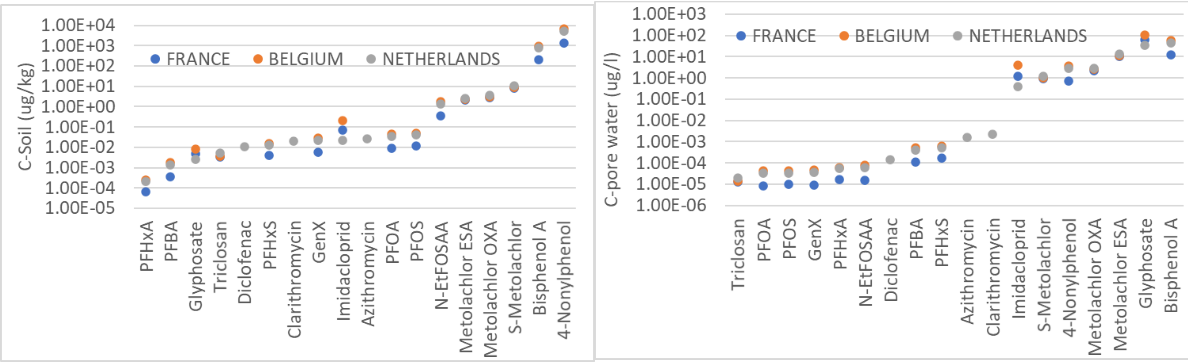
- Tier 1: Estimation using emission data. The fate of CECs was determined using the SimpleBox 4.0[4] tool, which is a steady-state model enabling to predict environmental concentration for one substance at a time. In Tier 1, general chemical and physical substance properties and national landscape data were collected. Assumptions on emissions, expressed as use/production volume were made. Fixed emission pathways regarding the distribution over the compartments air, soil and water were set.
- Tier 2: Estimation using specific source and input to soil. A CECs specific source and input to soil replaced the direct load to soil calculated from substances properties and emissions in Tier 1.

The Tier 1 – Fate Module - was run for the three following S-P-R schemes:

- Emissions of industrial (REACH) chemicals (PFAS and phenols & alkylphenols) to the environment.
- Release of CECs in soil according to pharmaceuticals consumption (total use or total sale) and its percentage of loss along the chain (excretion, WWTP sludge concentration, soil application).
- Direct application of pesticides on agricultural soil (based on de the total use or total sale per surface unit).

The figure below shows that the resulting soil concentrations for all compounds in all countries range from 0.07 ng/kg to 7 mg/kg. Most of the compounds have a concentration of 0.003 to 10 µg/kg (a factor of 3000 difference). Soil concentrations seem to be the result of combined model parameters, with no direct correlation with emission nor Kow. The differences between countries are limited compared to the total range.

[4] SimpleBox4.0, see rivm.nl/en/soil-and-water/simplebox



Calculated soil concentrations (left) and pore water concentrations (right) for the 18 pilot substances.

The calculated pore water concentrations show another picture. Most of the compounds have a concentration of 10^{-5} to 10^{-3} µg/l, whereas the five plant protection products and the two alkylphenols have a concentration ranging from 1 to 60 µg/l. The main reason for these differences is the amount of emissions.

The emission data were very heterogeneous depending on the substances and on the countries:

- Emissions of pesticides to the soil, and therefore pesticides emission load were the most reliable as pesticides sales are well registered for all countries.
- The sale / use of pharmaceuticals is also well known in many countries, however information about the percentage of sewage sludge applied to soil need to be better known in countries practicing sludge application (France and Belgium) to gain confidence in calculated soil load.
- REACH-regulated industrial chemicals are the most difficult compounds to estimate the emissions. The production volumes within the EU are registered in categories (1—10 tons/year, 10-100, etc.). However, losses to the environment can occur in the whole chain, from the production of the substance, via use in (end) products, consumer use, to recycling and waste disposal. These various potential emissions steps were not distinguished in PREMISS and a fixed emission to surface water, soil and air was used. Moreover, REACH data are not available for small volumes (<1 ton/year) and for former REACH substances (which are now prohibited).

The Tier 2 – Fate Module - was applied to the application of material on rural soil, including direct sources (such as pesticides, illustrated for glyphosate) and indirect sources (such as manure, organic fertilizer/compost, digestate, sewage sludge, and dredged material application, illustrated for PFOS).

Tier 2 on direct source enabled, for the Netherlands, to target specific crops (taking into account crops rotation) on which glyphosate was applied, as opposed to Tier 1 in which uniform spreading of the glyphosate for a long time application over the overall Dutch agricultural surface area was assumed. Unsurprisingly, direct source Tier 2 yield to soil load ranging from 0.7 to 1.7 kg/ha/y, which is higher than Tier 1 calculation (0.07 kg/ha/y).

Tier 2 on indirect source required two types of information: the amounts of materials applied to the soil and the contaminant concentrations in the applied material. The first type of information appeared difficult to obtain (derived from the total amount produced). Results for PFOS concentration in sewage sludge in Wallonia and France, and in dredged material in the Netherlands are presented in the table below. There are large differences both in the amounts applied, and the contaminant concentrations.

Country	Applied material (kg/m2/y)	Amount applied (kg/m2/y)	Concentration (µg/kg)	Added concentration in top 30 cm/y
Be-Wal	Sewage sludge	0.004-0.04	112	0.001-0.01
Fr	Sewage sludge	0.5-1.0	243	0.27-0.54
NL	dredged material	5.5	2.8	0.031

Estimated yearly added concentration of PFOS as caused by the application of sewage sludge (Wallonia, France) or dredged material (The Netherlands)

Contaminant concentrations in dredged sediments, sewage sludge, compost, digestate and manure had been collected giving a first insight in contaminant concentrations in soil amendments. However, the analysis showed that:

- A need for **an advanced search for more data** (particularly with respect to manure) and a more comprehensive analysis of available data.
- Filling data gaps by undertaking **monitoring programs of materials** added to the agricultural land. The Netherlands is currently working on a program to monitor CEC in soil and sediments, but other soil amendments (such as manure widely used in the Netherlands) are out the scope of this monitoring program.
- **Identification of additional CECs** to the existing substances included in the **European fertilizer regulation**, through a monitoring program. Wallonia is currently undertaking this action.

Estimation of CECs toxicity (lead by RIVM)

In PREMISS, CECs toxicity was assessed according to one generic method (Tier 1) which makes use of existing assessment tools allowing for a large number of substances to be assessed. The PREMISS Tier 1 toxicological module was limited to toxicity assessment of organic chemical substances. Toxicity assessment consisted in deriving a threshold value (i.e PNEC of HBGV) for the four receptors targeted by the prioritisation: a) human toxicity (drinking water); b) human toxicity (ingestion of soil and crops); c) direct ecotoxicity and d) secondary poisoning (indirect ecotoxicity).

It was performed using five steps:

- Determination of the **chemical structure representation** for each substance (Simplified Molecular Input Line Entry Specification, SMILES).
- **Hazard labelling** using the Substances of Very High Concern similarity tool (ZZS-similarity tool). Flagging the substances as Carcinogenic, Mutagenic, Reprotoxic (CMR), Persistent, Bioaccumulative, Toxic (PBT), Very persistent, very Bioaccumulative (VpVB) and Endocrine Disruptive (ED), was considered relevant additional information for the prioritisation itself.
- **Assessment of human toxicity of a substance**: The human toxicity of the pilot substances was assessed using two separate aggregated pathways, namely toxicity through ingestion of soil and crops, and toxicity through direct consumption of groundwater as drinking water (untreated). For both pathways the daily exposure was estimated and the tolerable daily intake was determined. The critical concentration for human toxicity was derived according to:
    - The tolerable daily intake for humans was based on the Threshold of Toxicological Concern (TTC), estimated by the substance properties, through the SMILES.
    - The human exposure estimation, calculated from substance properties and environmental CECs concentrations (estimated or measured).
- **Assessment of direct ecotoxicity**: The direct ecotoxicity consists in deriving PNEC value using ECOSAR 2.0 using CAS or SMILES input.

- **Assessment of secondary poisoning:** The risks associated with secondary poisoning were assessed using the more critical of two RCRs, namely the RCR for consumption of worms, or for the consumption of plants. Therefore, the PNEC was calculated for both plants and worms (based on the TTC approach) and the PEC (environmental CECs concentrations) calculated in the fate module used was transformed into a PEC in worms or plants.

Toxicity assessment was performed for the 18 “pilot” substances. The main outputs of each steps are described below:

- Chemical structure representation and substance labelling: a SMILES code was available for all the substances. As for the labelling, the similarity tool only assigns labels based on the substances included in its database, and not based on substance properties. Therefore, some **substances could not be labelled** (eg N-EtFOSAA, a precursor to PFOS), even though their properties may suggest potential hazard. Discussion are underway to fill this gap such as the development of **a methodology for PMT assessment** and the on-going classification of **identification of ED-label** (under review, https://echa.europa.eu/fr/ed-assessment).
- Human toxicity: human exposure was calculated for ingestion of soil and crops and consumption of groundwater as drinking water (untreated). Exposure was not calculated for diclofenac, triclosan, clarithromycin, azithromycin for France and Belgium as no emission data were available. Exposure determination depends highly on CECs estimated environmental concentration: directly for groundwater and partly for ingestion of soil and crops, other variables influencing exposure due to the uptake by plants.
- Direct ecotoxicity: the estimated ecotoxicity of the “pilot” substances ranges from 5.6^{-3} up to 4.96^{+5} µg/l (8 orders of magnitude). Derivation using SMILES could lead to unexpected results and using CAS number when possible was recommended.
- Secondary poisoning: When testing the “pilot” substances, the PNEC worm and PNEC leaf both ranged almost 3 orders of magnitude. For the PEC, substance properties and the soil, pore water, and air concentrations are important for the prioritisation.

Prioritisation prototype and results

After performing the fate and toxicity assessment, the prioritisation of the substances was carried out by calculating and sorting the Risk Characterisation Scores (RCR), which was the quotient of fate and toxicity (see equation below), for multiple substances from high to low. It follows that when comparing the RCR of multiple substances, the substance with the highest RCR poses the highest risk for the assessed receptor. This is in essence a prioritisation. A prioritisation list was generated for each of the four receptors: human toxicity (drinking water); human toxicity (soil and crops ingestion); direct ecotoxicity; secondary poisoning (indirect ecotoxicity).

$$RCR = \frac{[Predicted\ environment\ concentration\ (PEC)]}{[Predicted\ no\ effect\ concentration\ (PNEC)]}$$

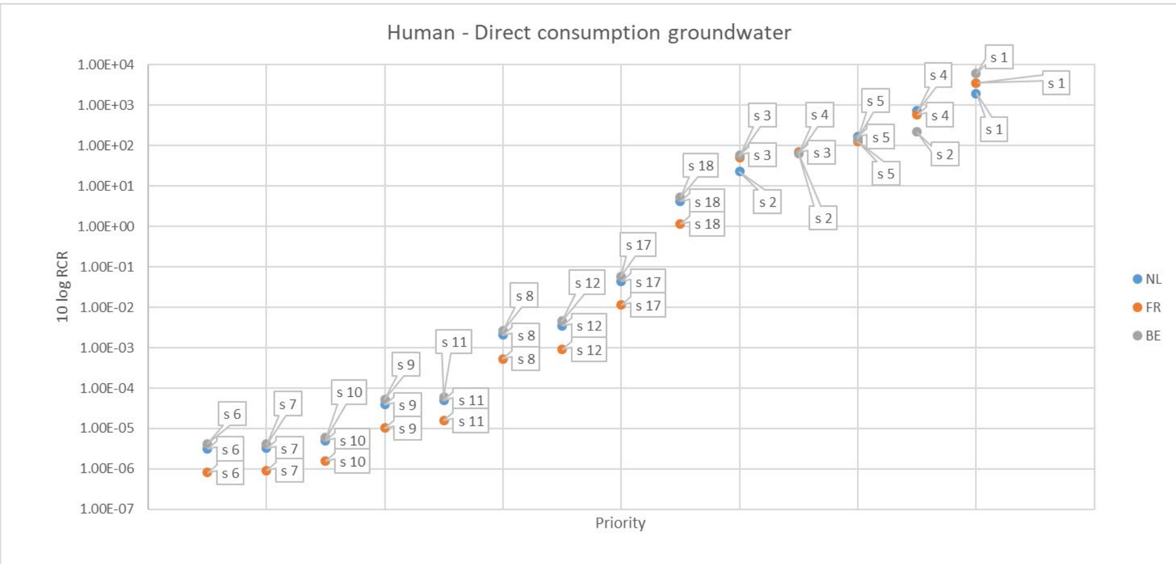
Calculation of the Risk Characterisation Ratio (RCR). For human health the PNEC is substituted by the Health Based Guidance Value (HBGV)

The quality of a prioritisation depends on the quality of the RCR, which depends in turn on the quality of the data and prioritisation scale. A lower quality RCR based on more uncertain data will result in prioritisation with high uncertainty. Three prioritisation tiers calculation were proposed based on the data input used in the prototype:

- Tier 1: basic prioritisation with generic emission, calculated CEC concentration in environmental media from fate modelling, and generic toxicity data (QSAR & SMILES). The main purpose of tier 1 is to **prioritise a wide set of substances** based on information that is available for many substances, either collected in large databases or calculated. The absolute value of the RCR is uncertain and not relevant at this tier.
- Tier 2: prioritisation with additional national or regional data on emissions for a specific source and customised fate modelling (PEC) and/or toxicity value from specific literature and/or experimental. One of the exploitation of Tier 2 output could be **to decide on what specific compounds actions may be targeted, for a specific SPR scheme** (such as monitoring in soil or groundwater).
- Tier 3: risk assessment based on CECs measurements in exposure media and/or appraised toxicity data. In Tier 3, the **potential risk of a substance can be assessed**.

Prioritisation outcomes (Tier 1)

Prioritisation prototype was run for the "pilot" substances for the four receptors and for France, Belgium and the Netherlands. Results are exemplified and illustrated for human toxicity ingestion of soil and consumption of vegetables in the following figure.



RCRs for Human toxicity ingestion of soil and consumption of vegetables for all three countries.

Results showed that:

- There was **no significant difference between countries**. Because the estimated toxicity was the same it could be concluded that differences in the fate calculations for soil and groundwater in different countries were small.
- The **order of magnitude in the prioritisation was much higher for the human endpoint of 'ingestion of soil and crops' than for 'consumption of groundwater'**. The reason is that the uptake of contaminants from the soil into crops gives an additional differentiation of compounds, implying higher uncertainties in the prioritisation.
- For all four receptors the **prioritisation returned RCR values differing at least 9 orders of magnitude** (direct ecotoxicity and human groundwater consumption, more for human ingestion and secondary poisoning). **This difference is large enough for substances with a relatively high RCR to be prioritised and with a low RCR to be discarded**. Taking into account the effect of uncertainties in

parameters, it is not expected that the priority will change from high to low or vice versa. This is also due to the use of worst-case assumptions limiting the impact uncertainties may have on the model outcome. Additionally, some grouping occurred in the RCR scores, notably the pesticides all received similar RCR scores, whereas for other substance types the RCR scores could differ multiple orders of magnitude. Within the group of pesticides with a high prioritisation it could be recommended to go to a tier 2 assessment.

- **A clear cut-off point for which substances ought to be prioritised was however not present**. Such a cut-off point may depend on other factors than the RCR itself, such as the number of substances that can be assessed in tier 2 due to time constraints. For example, a top 20 or top 100 of the substances on the prioritisation could be prioritised for tier 2. If different groups of compounds can be identified, the group with the highest RCR could be prioritised in tier 2. It is important advice to end-users (for ex. policy makers) and further discussion on this topic is required.

The ranking of substances for the four receptors is presented in the table below.

Priority	Eco-direct	Eco-secpois	Human-drink	human-ingestion
1	4-Nonylphenol	Metolachlor ESA	Glyphosate	Metolachlor ESA
2	S-Metolachlor	Glyphosate	Metolachlor ESA	Glyphosate
3	Bisphenol A	Metolachlor OXA	Metolachlor OXA	Metolachlor OXA
4	Imidacloprid	Clarithromycin	S-Metolachlor	Clarithromycin
5	Metolachlor ESA	Imidacloprid	Imidacloprid	Imidacloprid
6	Metolachlor OXA	Azithromycin	Bisphenol A	Azithromycin
7	Clarithromycin	S-Metolachlor	Clarithromycin	S-Metolachlor
8	Azithromycin	N-EtFOSAA	Azithromycin	Bisphenol A
9	Triclosan	4-Nonylphenol	4-Nonylphenol	PFHxS
10	N-EtFOSAA	Bisphenol A	N-EtFOSAA	PFHxA
11	Glyphosate	GenX	GenX	Diclofenac
12	PFOA	Triclosan	Triclosan	4-Nonylphenol
13	Diclofenac	PFHxS	PFHxS	Triclosan
14	PFOS	PFHxA	PFBA	PFOS
15	PFHxS	PFOS	Diclofenac	N-EtFOSAA
16	PFHxA	PFOA	PFHxA	GenX
17	PFBA	Diclofenac	PFOS	PFBA
18	GenX	PFBA	PFOA	PFOA

Table showing the ranking of substances in the four prioritisations side by side. Industrial chemicals are in blue, pesticides in green, pharmaceuticals in red and PFAS in purple

Results showed that:

- **Pesticides** have a relatively **high priority as a group**. This is strongly related to the high estimated concentrations in pore water and the high human toxicity and ecotoxicity of this group.
- **Industrial chemicals have a high score for direct eco-toxicity**. This difference can be explained by the expected low human toxicity.
- **PFAS have a relatively low ranking** of PFAS-compounds for all four endpoints. This can be explained by low calculated concentrations in soil (due to low Kow or low emission tonnage, chosen in absence of REACH data). PFAS toxicity data derived from Tier 1 may be underestimated.
- **Pharmaceuticals prioritisation is more scattered**.

Uncertainties and key parameters associated with the prioritisation prototype

Emission data, log Kow and degradation rate (which has been set to zero for soil, in absence of available data) have a big influence on CECs calculated environmental concentrations. CECs environmental concentration being a pivotal element for RCR determination, these parameters play also a key role in the prioritisation approach. Toxicity can also be an important factor, but for the “pilot” substances, does not lead to a complete change in ranking when using pore water concentration.

Besides the calculated CEC environmental concentration, other parameters can lead to the difference in priorisation. For direct ecotoxicity and human toxicity from consumption of drinking water, the estimated toxicity is relevant for differences. Due to the methodological approach, human toxicity will be more uncertain than direct ecotoxicity. For secondary poisoning and human toxicity consumption and ingestion (of soil and crops) the exposure parameters will also contribute to the uncertainty (Kow, solubility and vapour pressure), additional to uncertainties in the estimated toxicity.

Modalities of use and limitations of the prototype

Prototype testing and associated uncertainties highlighted the importance of clearly stating the current limitations of the prioritisation prototype and its modalities of use in order to ensure fair exploitable results.

The main modalities of use are summarised below:

- **Importance to choose scenario according to substance type and use**: scenario refers here to the Source – Pathway – Receptor scheme which describes how the substances may be emitted, enter the environment, migrate and reach the targeted receptor(s). In order to have a proper risk-based approach, it is crucial to select the scenario(s) that are relevant to the substance type and use. Note that a substance may come from multiple sources, follow multiple pathways and reach the same receptor.
- **Scale of application**: PREMISS prioritisation prototype was mainly tested at a national scale, either using REACH data from which national emissions were derived or using national emission data and considering that the distribution was homogeneous over the country (according to landscape setting and agricultural surface area). In principle, the PREMISS prioritisation prototype can be used from European to local scale, as long as the input data are available at the chosen working scale and the SPR scheme is set in the prototype.

- **Data input**: the prioritisation approach developed in PREMISS aims to undertake a relative prioritisation approach among substances and not an absolute prioritisation approach. Data input quality (emission, rate and toxicity) especially plays a key role in the quality of a prioritisation. Moreover, it was deemed very important not a) not to mix different types of emission data (source, scale, etc.) for the same scenario/pathway; b)not to mix different toxicological data (measured PNECs & QSAR based PNECs, TTC & evaluated HBGV and PNEC for secondary poisoning).

Current limitations of the prioritisation prototype are listed below:

- **Data collection and input**: data collection and input in the prototype were undertaken manually. Automatisation of data collection and input may be enabled to deal swiftly with large sets of substances. Moreover, some data were missing (eg emission data) and were approximated. Proper methodology to deal with data gaps may be further developed.

- **Fate and transport model**: degradation rate could not be included in the prototype, as no data were available. REACH emission data are not available for historical substances and/or prohibited substances and for substance emission below 1 ton/year. In this case, predicting CEC environmental concentrations at Tier 1 level was not easy, and REACH emission was chosen as 0.1 to 1 ton/year. The emission duration of the substance over the years in the media was not taken into account in the model. Set assumptions on the distribution of substance between the water, soil and air media may induce a bias in the prioritisation, as it does not take into account the solubility or the volatility of the substance (which are important parameters for partitioning in various environmental media).

Stakeholders' feedback on PREMISS outcomes

At the end of the project, PREMISS results were concisely presented to the Dutch, Belgium and French stakeholders' group. The key points of stakeholders' feedback included:

- A **wide range of potential uses** for the prioritisation approach was spotted by the stakeholders, such as regulation, prevention, monitoring and management purposes.
- There was a need to clearly indicate the uncertainties, assumptions, limitations associated with the prioritisation approach and to get more insight on **uncertainties** (associated with the results) **management**.
- The **scale of the application** was discussed thoroughly by the stakeholders. The prioritisation approach was deemed useful for diffuse contamination (which was the scale at which the tool was demonstrated). Stakeholders also showed interest in point-source applications, which need to be further tested.

Moreover, stakeholders emphasized that the **future development of the tool depended strongly on the objectives of the prioritisation and its scale**. Improvements towards a full-scale prioritisation prototype were suggested on tool development, tool validation and tool "instructions for use". In addition, stakeholders recommended developing the **exploitation of the results towards a policy instrument** and sharing results with **European institutions** or **EU networks**.

Comparison of calculated and measured CECs environmental concentrations

Despite some limitations identified in occurrence data inventory (e.g. lack of occurrence data, high quantification limits, etc), measured environmental concentrations and calculated environmental concentrations from the fate module (Tier 1) were compared. For this comparison, it was decided to focus on the substances having the largest set of measured data in a given country:

- PFAS environmental concentrations in the Netherlands,
- Pesticides environmental concentrations in Belgium.

With regard to PFAS, estimated groundwater and soil concentrations (with the exception of N-EtFOSAA and GenX for soil) were underestimated by the model compared to the measured concentrations. The most probable reason is that PFAS emissions were underestimated because the prototype use current emissions that does not exist for banned substances. The default value used in the prototype to get around the problem is not adapted for PFAS emission.

With regard to pesticides, estimated glyphosate concentration in groundwater were higher than measured ones by a factor ~1000 whereas estimated soil concentrations were underestimated by a similar factor. This could be related with its low log Kow (-3.12). Regarding imidacloprid, estimated concentrations in soil were in the same range as measured concentrations, whereas they were overestimated in groundwater. Estimated concentrations for metolachlor in groundwater were in the same range as maximum measured concentrations but higher than average concentrations with a factor 100. Comparison for metolachlor in soil was impossible due to the absence of reported data in soil for Belgium.

Recommendations and Perspectives

PREMISS prioritisation output and stakeholders' feedback enabled to draw recommendations on CECs prioritisation in the soil at a general level and more specifically related to prioritisation prototype improvement.

General recommendations

- CECs occurrence data:
    - Facilitating data inventory and compilation by the **harmonisation** of substance international signalling, data treatment and associated metadata.
    - **Central storage** of existing occurrence data in WWTP solid effluents, manure, compost and digestate shall be organised.
    - **(Prospective) soil monitoring** in order to acquire CECs data in soil.
    - Development of a **more coherent EU policy framework on soil** (including CECs).
    - **Promote CEC data collection in soil and storage** in a common database as initiated by the LUCAS survey and beyond.
- Application and exploitation of prioritisation prototype
    - The prioritisation prototype needs to be used according to **its modalities of use** and **its limitations** and associated **uncertainties**.
    - The way to exploit and transfer results **towards a policy instrument and monitoring or management guidelines** needs to be developed.
- The following links with EU initiatives are foreseen:
    - R&D projects in support of the **European Green Deal**: e.g. the PROMISCES HORIZON 2020 project.
    - A **Soil Deal for Europe** launched by the European Commission.
    - **The EU Soil Strategy and the EU Soil Observatory (EUSO)**, which are themselves integrated into a diversified regulatory framework which includes the "Zero emission action plan", the "Circular Economy action plan", the "Chemical strategy", the "Biodiversity Strategy for 2030", the "Farm to Fork strategy" and the "Forest strategy".
    - Share knowledge and methodology and collaborate **with European institutions** (such as JRC, EEA) or **EU networks** (such as NORMAN, NICOLE).

Specific recommendations to improve the prioritisation prototype.

It is crucial to note that the recommendations proposed are very dependent (and of various relevance) of the different prioritisation ambitions. The recommendations are divided into 3 categories: technical developments, technical validation and a user-friendly interface development:

- Technical tool development:

- Design and study additional scenarios – “Source-Pathway-Receptor” schemes: scenarios having a focus on **urban and industrial soil** and encompassing several scales (especially **local and site** scale) may be further developed and tested in the prioritisation tool. Scenarios on **fire-fighting foam use** (targeting PFAS) or **sediment deposition due to flooding** on agricultural soil were also among stakeholders’ demands.
- Data gaps management: development of a **methodology to deal and manage data gaps** (emission, occurrence, fate and toxicity).
- Fate module: several improvements on **emission** (include past emission, emission duration, point source emission), **persistence** (include CEC degradation in soil), and others (e.g. development of Tier 2, a wider range of soil types) were suggested.
- Toxicity module: collect / acquire **more accurate toxicity data**, **including PMT classification**, develop Tier 2.

- Technical tool testing and validation:
    - Tool verification may include: undertaking a more **extensive sensitivity and uncertainties analysis**, additional **comparison** of calculated and measured CECs environmental data, **comparison** of PREMISS results with other prioritisation exercises.
    - Tool testing may include: testing the tool **with known substances** (for which there are abundant occurrence data); testing the tool with a new **emission dataset on a national level**; testing the tool for the **additional scenarios**; testing the tool for **additional countries**; testing the tool for a **larger set of substances.**

- Development of an end-user friendly interface: this could be achieved by 1) **automatizing** the data collection and data input in the prototype, 2) developing a **user-friendly interface** according to stakeholders’ needs and uses and 3) providing an **extensive “user guide”**.