OPUR : Observatoire d'hydrologie urbaine en Île de France

Thème de recherche R4: Scénarisation de la gestion des eaux pluviales urbaines dans un contexte de changements globaux

Action de recherche R4.4 : Modélisation de l'émission des polluants à l'échelle urbaine

URBAN SCALE MODELING OF BIOCIDES EMITTED IN RUNOFF FROM BUILDINGS ENVELOPE

Rapport d'avancement de la thèse de

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Janvier 2024

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Abstract

Microbes, fungi, algae, and other microorganisms have the ability to colonize the building envelope, resulting in the deterioration of the aesthetic look of building structures. Biocides are widely used to prevent this invasion and extend the lifespan of building external elements. These potent agents have played a crucial role in maintaining the aesthetic appeal of cities; however, their growing and excessive usage has raised concerns about their adverse effects on the environment, particularly on stormwater runoff and on aquatic ecosystems. This paper aims to provide an in-depth review of biocides, focusing on key aspects related to their usage and effects. It will explore the properties of biocides, clarifying their chemical characteristics, as well as the legal frameworks governing their use and applications. Also, it will present two models that will be employed throughout the thesis: COMLEAM, which will simulate rainfall interception by building facades and roofs and incorporate various research-derived leaching law models, and the TEB model, which will be modified to represent the interception of precipitation by vertical surfaces and the influence of wind, as well as the available stocks and emitted fluxes at the grid scale. Finally, it will give a concise overview of the next steps that will follow.

1 Introduction

Commenté [AB1]: Le plan est peut-être à revoir légèrement, avec des parties plus organisée et des titres plus parlant :
Mélanger le 2 et 3 en « généralités sur les biocides »
Mélanger le 4 et 5 en « usages de biocides »
Changer le titre du 6 en transfert des biocides depuis l'usage vers le milieu récepteur
Préciser le titre du 7 en « modéliser le transfert des biocides dans la ville » ou quelque chose d'approchant

Commenté [RS2R1]:

Diffuse pollution carried by urban stormwater runoff has been considered for several decades to be a major cause of the degradation of aquatic ecosystems. The large number of regulations and research on micropollutants shows the growing interest in this type of contamination (Briand, 2018; Paijens, 2019; Wicke et al., 2022). Biocides are among the micropollutants that are toxic to aquatic organisms at low concentrations (Mohr et al., 2008). Their use in urban environments is increasing; they are often added to building materials (coatings, paints, waterproofing membranes) or used in preventive (impregnation of wood) or curative (cleaning of tiles and masonry) treatments to fight against the growth of microorganisms in humid conditions (Shirakawa et al., 2002). During rainy weather, biocides are released into the building runoff (Bester and Lamani, 2010; Bollmann et al., 2014b; Burkhardt et al., 2012, 2011; Schoknecht et al., 2003), and are then discharged into the ground or the stormwater management system, negatively impacting aquatic and terrestrial ecosystems (Bollmann et al., 2017a; Giacomazzi and Cochet, 2004; Hernández-Moreno et al., 2019) The thesis work of Claudia Paijens (2019), conducted at the scale of the Parisian agglomeration, showed that biocides are ubiquitous in urban waters with a risk for the aquatic environment, and for several substances (diuron, mecoprop, terbutryn, carbendazim, etc.), a stormwater origin via the leaching of building materials was highlighted.

Researchers have studied biocide emissions from building materials in the laboratory or *in situ* on a small spatial scale (test bench or roof) (Bollmann et al., 2016; Burkhardt et al., 2011; Schoknecht et al., 2016; Van de Voorde et al., 2012) and less frequently on the scale of small urban districts (Hensen et al., 2018; Wicke et al., 2022). The results obtained have improved the knowledge of emissions and leaching processes of biocides used in the building construction. These studies also allowed the development of modeling tools to assess biocide emissions in runoff from the building scale to the small urban watershed scale using deterministic and stochastic approaches (Burkhardt et al., 2020; Van de Voorde et al., 2012). Modeling biocide emissions at the city scale is a scientific challenge due to the complexity of the phenomena studied and the variability in space and time of the factors controlling emissions. The use of stochastic approaches and the exploitation of available urban data are necessary steps to meet this challenge. The thesis, begun in mid-February 2023, mainly aims to develop and implement a modeling framework to evaluate, at the urban scale, the fluxes of biocides emitted in runoff water from buildings envelope. To achieve the main objective, the thesis will be structured with several specific objectives:

- Develop a methodological framework for assessing the spatial distribution and characterization of the stock of biocides present in the building's envelope;
- Adapt existing biocide leaching models to urban-scale use;
- Implement the identified modeling approaches in a distributed urban hydrology model and test it at the scale of an urban area.

2 General information about biocides

Approximately five decades ago, the term "biocide" appeared to gain popularity; however, its use until now is not universally accepted, and there is no agreement on its precise definition. Many definitions are used that cause confusion with other terms, such as sterilizers and pesticides, leading them to be interchanged in different sectors and for different purposes. Literally, the term bio-cide is derived from the Greek "bios," which means life, and the Latin "cida," which means to kill, thus giving it the meaning of a substance that kills living organisms. Block (2001) defined it as "a chemical or physical agent that kills all living organisms, pathogenic and nonpathogenic." Warne and Reinchelt-Brushett (2023) define it as "a chemical that is designed to have the same properties as a pesticide but is not used to protect plants or plant products".

The meaning of the term "biocide" varies between countries, especially among decision-makers, emphasizing the many viewpoints and contextual subtleties that influence how it's interpreted. The American Chemistry Council uses the terms "biocides," "antimicrobials," and "antimicrobial pesticides" synonymously. It defines them as "substances that prevent the growth and spread of microbes like bacteria, viruses, and fungi such as mold. Antimicrobial products are used in hospitals, homes, schools, and countless other spaces to help kill germs, disinfect drinking water, ensure everyday products last longer, and keep manufacturing processes running safely" (American Chemistry Council, 2023). Instead of having a specific standalone definition for "biocide" in its regulations, the United States Environmental Protection Agency (US EPA) regulates biocides found in household and industrial products as pesticides under the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA). Accordingly, a pesticide is (1) "any substance or mixture of substances intended for preventing, destroying, repelling, or mitigating any pest"; (2) "any substance or mixture of substances intended for use as a plant regulator, defoliant, or desiccant"; and (3) “any nitrogen stabilizer with some exceptions” (US EPA, 2014). It covers a wide range of products, including insecticides, herbicides, fungicides, rodenticides, and other substances used to control pests. Thus, the term "biocide" is not a separate or unique regulatory category under the US EPA. Instead, products typically referred to as biocides, such as disinfectants and antimicrobial substances, are classified as pesticides and are controlled by the EPA as such.

In the European Union and Switzerland, although biocidal products are one of the two main product families of pesticides, along with phytopharmaceutical products, according to Pesticides Framework Directive 2009/128/EC, each of them is subject to a different regulatory framework. Distinction between both families relies on the intended application of the product. Phytopharmaceutical products are the ones used in agriculture to protect plants and vegetated areas and follow EU regulation 1107/2009 (repealing Directives 79/117/EEC and 91/414/EEC). Biocidal products follow EU Regulation No. 528/2012 (repealing Directive 98/8/EC). Referring to this regulation, Article 3, May 22, 2012, biocidal products include:

“- any substance or mixture, in the form in which it is delivered to the user, consisting of, containing or generating one or more active substances, which is intended to destroy, repel, render harmless, prevent the action of or otherwise control harmful organisms by any means other than physical or mechanical action,

- any substance or mixture generated by substances or mixtures which do not themselves come under the first indent, intended for use in destroying, repelling, rendering harmless or preventing the action of harmful organisms, or for controlling them in any other way by means other than mere physical or mechanical action.”

Since this definition, like the ones mentioned before, may lead to confusion with other terms, the BPR went further into specifying the fields of use in which a product is called a biocidal product, thus following Regulation No. 528/2012. In order to identify a product as a biocidal, this regulation categorizes a total of 22 types of biocidal products into 4 major groups, as shown in Table 1.

Commenté [AB3]: Tu peux éventuellement ajouter une phrase comme quoi une même substance chimique peut avoir plusieurs usages, pesticides et biocides (au sens de la réglementation européenne). Voir même d’autres usages PPCP

Table 1. Biocidal Products’ types as per BPR

Group	Product Type
1- Disinfectants	type 1: Human Hygiene type 2: Disinfectants and algaecides type 3: Veterinary Hygiene type 4: Food and Feed Contact Surfaces type 5: Drinking water
2- Preservatives	type 6: Preservatives for products during storage type 7: Film preservatives

	type 8: Wood preservatives type 9: Fibre, rubber and polymerised materials preservatives type 10: Construction material preservatives type 11: Preservatives for liquid-cooling and processing systems type 12: Slimicides type 13: Working or cutting fluid preservatives
3-Pest control	type 14: Rodenticides type 15: Avicides type 16: Molluscicides, vermicides and products used to control other invertebrates type 17: Piscicides type 18: Insecticides, acaricides and products used to control other arthropods type 19: Repellents and Baits type 20: Control of other vertebrates
4- Other biocidal products	type 21: Antifouling products type 22: Embalming and Taxidermy Fluid

The biocide authorization in France is governed by the biocidal products regulation BPR under the European Chemicals Agency (ECHA), which applies to all EU members. As previously mentioned, EU Regulation No. 528/2012 is applied “to biocidal products which, in the form in which they are supplied to the user, contain, generate, or consist of one or more active substances." So, following this statement, any product that contains, generates, or consists of active substances must follow this regulation and must be authorized before it can be placed on the market.

Before explaining the authorization process, it’s important to distinguish an active substance from a product type. The key chemical component included in a biocidal product is an active substance, and a single product can contain multiple active substances. So, each active substance must obtain approval at the European level for each biocidal use. For example, if an active substance receives authorization for film preservation (type 7) but is not authorized as a disinfectant for drinking water (type 5), it can be added to paints but not to drinking water. A product must include only authorized active substances for its type to proceed in the authorization process.

To sell a product, a producer must apply for a marketing authorization, which assesses the entire formulation and determines if the product can be launched in the market. In practical terms, the paint producer applies for a marketing authorization for its paint containing an authorized preservative, and the formulation is assessed as a whole. Sometimes there may be restrictions on the quantity of preservatives in a finished product. The entity that is in charge of getting the marketing authorization is the first one to sell the product, specifically the producer (formulator). To get authorization for launching in a European Union member state market, companies must apply for product marketing authorization, which includes the following stages:

1. **Active substance approval:** The active substances used in the biocidal product must get EU approval first; otherwise, they can’t be used. Thorough assessments are conducted to evaluate the effectiveness and safety of these substances. The active substances (biocides) mentioned in Regulation No. 528/2012 are not definitely authorized. A biocide can be authorized, non-authorized, or under assessment (transitional regime). The transitional regime is defined by Article 89 as “the period during which the placing on the market and use of biocidal products is governed by the national provisions in force in each member state, pending approval of all the active substances they contain at the community level” (ANSES, 2020). In 2023, 756 active substance/product-type are mentioned in the regulation, of which 282 are authorized, 49 aren’t, and 425 are still under assessment (ECHA, 2023a).
2. **Biocidal product authorization application:** Once the active substances present in the product have been approved, the company can apply for product authorization in specific EU member states, including France. The application requires thorough data on the product's composition, intended usage, effectiveness, and risk assessment. Certain products might be authorized at the Union level, giving companies the possibility to place their biocidal products on the market throughout the Union without having to obtain a specific national authorization. This Union authorization will grant the same rights and obligations in all Member States as national authorizations (ECHA, 2023b).
3. **Evaluation and decision:** Within 365 days, the Member State's competent evaluation authority must review the application and determine whether to grant the permission.

3 Uses of biocides

Biocides may be found almost everywhere on the building's envelope. They may be present on roofs, facades, foundations, terraces, and even around the building. Biocides commonly prevent the growth of mold, algae, and other microorganisms that can damage building materials. Applying them during the construction phase or as part of regular maintenance ensures the longevity of the structure. The following section will elaborate on the presence of biocides on roofs, facades, and foundations.

3.1 Roof

Different types of roofs (mineral, polymeric, and wooden) contain biocides, both during construction and during service. The next sections go into further detail on the types of biocides that may be found on these roofs.

3.1.1 Mineral roofing

Concrete, natural slate, and ceramics are minerals used as roofing materials, especially for sloped roofs (e.g., tiles) (De Buyck et al., 2021). Mineral roofs may be colonized by different types of microorganisms, which may cause many functional problems in addition to their unpleasant and dirty appearance. Speaking of tiles, the growth of moss and other microorganisms may result in leaks. Also, the microorganisms raise the water content of the tiles by preventing evaporation, which causes the tiles to burst during cold weather (Van de Voorde et al., 2012). The solution for biological attacks on such roofs might be preventative or curative. Biocidal coatings (titanium, iron, zinc, or tin oxides) are used as a preventive treatment to hinder biological development through photocatalytic activity. A biocidal material can also be directly applied to the roof as strips (e.g., copper strips on the roof ridge). As for curative treatments, moss removal is the dominant one. Anti-moss products treat roofs, and they mostly consist of quaternary ammonium salt aqueous solutions (alkyldimethylbenzylammonium or benzalkonium) in the French market (Van de Voorde et al., 2012).

3.1.2 Polymer roofing

Several types of polymer materials are used on roofs for waterproofing applications, some of which include biocides. A high concentration of arsenic was found in polyvinylchloride (PVC) roofing material runoff, which may be linked to the presence of arsenic as a biocide in the PVC composition. Other than PVC, bituminous is also used in roofs as shingles, membranes, and built-up roofs (De Buyck et al., 2021). The formation of roots within the bitumen sealing membrane on vegetated flat roofs may cause structural waterproofing issues. Preventol B2, a roof protection chemical containing mecoprop as a polyglycol diester, is so used. Hydrolysis of this chemical produces (R,S)-mecoprop (Figure 1) (Bucheli et al., 1998). Herbitect, including mecoprop as an ethyl hexyl ester, can also be used. Mecoprop is a widely used herbicide in agricultural and urban areas. Despite being classified and approved as a pesticide (E-Phy, 2024) and not as a biocide, it is used three times more on roofs than in agriculture (Paijens, 2019). People sometimes use these membranes even when root protection is not required (Wicke et al., 2015). The problem with mecoprop is that it can leak from its matrix to join contacting water. (Bucheli et al., 1998) detected concentrations between 1 to 500 µg/l in roof runoff in preliminary tests.

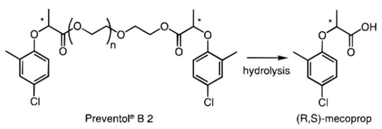


Figure 1. Preventol B2 and (R-S) Mecoprop structures (Bucheli et al., 1998)

Commenté [AB4]: Il est classifié comme un pesticide ET comme un biocide. Est-ce qu'il est interdit comme pesticide ? C'est ce que tu voulais dire ?

Commenté [AB5]: Poruquoi citer Paijens ? Mieux vaut revenir à la source originale

3.1.3 Wood roofing

Wood roofing components are treated with biocides (Wood preservatives-PT08) to prevent their degradation and provide resistance against weathering, insects, fungi, bacteria, etc. Referring to Table 3, wood preservatives PT08 can be used as a preventive treatment to prevent insects and other organisms from destroying or deforming wood, especially during the sawmill's processing stage (INERIS, 2012). Also, they can be applied as a curative treatment to eradicate microorganisms that have already infected the wood and to stop harmful microorganisms from spreading to other parts of the wood (European Chemicals Agency., 2018a).

Table 2 categorizes the biocidal active substance families currently employed as wood preservatives based on their function. Here, the term "family" refers to biocidal active substances having the same chemical structures.

Table 2. Classification of biocidal active substances families used as wood preservatives based on their function (INERIS, 2023)

Fungicidal action	Insecticidal action	Fungicidal and/or insecticidal action
Azoles	Synthetic pyrethroids	Boron compounds
Sulphonamides	Neonicotinoids	Copper compounds
Isothiazolinones	Cyanides	Quaternary ammoniums
Morpholine derivatives	Potassium salts	Carbamates
Tetrahydrothiadiazines	Benzoylureas	Coal distillation products
	Diphenyl ethers	Pyrazole carboxamides

3.2 Façade

Facade coatings are recognized in contemporary times for their potential to enhance the quality of life. Many people consider these coatings essential elements that contribute to aesthetics, allure, design, and emotional appeal. However, the main purposes of facades are to offer protection, allow the exchange of air with the external environment, permit the transmission of light into the interior, and create a division between public and private spaces. It is necessary to consider many environmental factors, such as temperature, sun, rainfall, humidity, wind, air pollution, and noise. Facade-coated surfaces are susceptible to the growth of microorganisms; thus, biocides are added to the paints and render formulations. During its service life, the walls should be periodically repainted in order to improve the performance of film preservatives during the service stage (Paulus, 2004).

Film-preservatives in facades' coatings have become more important with the introduction of thermal insulation systems. In the late seventies, algaecides were added as a new segment of film preservatives in Europe since they found that thermal insulation systems provided a favorable environment for their proliferation (Paulus, 2004). In fact, this system requires installing an insulating layer on top of the original exterior coat of the structure. A fresh coat of polymeric-based render is applied to protect this layer from the environment (Bester et al., 2014). These coatings are susceptible to microbial degradation and thus require film-preserving biocides to inhibit the growth of fungus and, more importantly, algae on building surfaces (Bollmann et al., 2017a).

3.3 Foundations

Naturally, termites are helpful insects that degrade cellulose-containing materials (e.g., dead trees). However, this particularity is dangerous and harmful when talking about buildings, since they can invade wood used in construction materials and damage the building's components. They are present in different types and species, one of which is the "subterranean," which is considered the main threat to buildings (European Chemicals Agency., 2018b). This type of termite is responsible for the majority of damages caused by termites to structures in metropolitan France as well as the French overseas departments (MEDD and METL, 2013).

On a building scale, termites generally colonize the soil due to the high moisture needed, so they build their nest at the foundation level and then find ways to percolate into the building (European Chemicals Agency, 2018). The route of termites is related to any cavity that has a millimeter-sized continuous open space from the ground to the inside of the structure (MEDD and METL, 2013).

In order to protect the buildings, the French legislative and regulatory system (articles L. 126-4 to L. 126.6, L. 126-24 and L.126-25, L. 131-2 and L. 131-3, L. 183-18 for sanctions, L. 271-4 as well as articles R. 126-2 to R. 126-4, R. 131-1 to R. 131-4, R. 126-42 and D. 126-43, R. 184-7 and R. 184-8 for sanctions and R. 271-1 to R.271-5 of the building and housing code) specify the conditions under which the prevention and control of termites and other wood-eating insects as well as merula are organized by the authorities (MEET, 2023). When termite outbreaks are identified in one or more municipalities, the authorities issue a prefectorial decree in accordance with the building code. Once issued, the decree applies the measures for protecting structures against termite activity to the entire department (MEET, 2023). The map shown below in Figure 2 specifies the status of each department towards prefectorial decrees in 2023 (MEET, 2023).

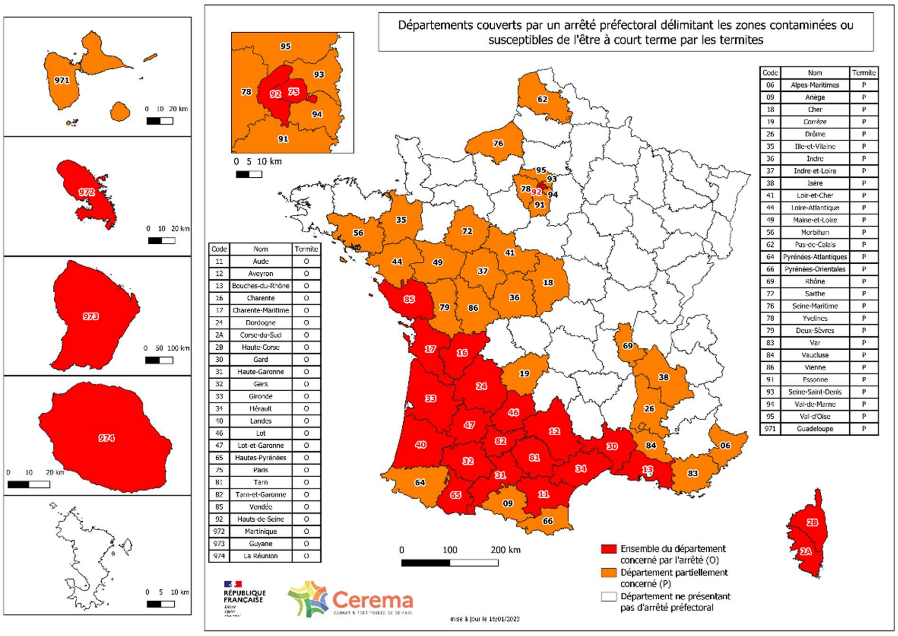


Figure 2. Departments covered by a prefectorial decree delimiting the areas contaminated or likely to be contaminated in the short term by termites: red for departments concerned with a prefectorial decree, orange for departments with partial concern and white for departments with no prefectorial decree (MEET, 2023, Accessed in 15/03/2023)

Several types of treatments against termites (physical, chemical, biological, etc.) are used at the foundation level. As for biocide usage, solutions can be split into two main groups. The first one includes the solutions applied directly to wood where PT08 is employed. The second group includes solutions used away from wood where PT18 is employed.

3.3.1 Wood treatment from termite attacks

Termite control methods can rely on applying wood preservatives (PT08) to infected or exposed wood (European Chemicals Agency, 2018). Verma et al. (2009) cited several studies that showed the effectiveness of some chemical substances on fighting termites: butylene oxide and triethylamine from (Akio et al., 1990); disodium octaborate tetrahydrate from (Maistrello et al., 2001); copper borate, water-borne copper naphthanate, and N’ N-naphthaloylhydroxylamine from (Arango et al., 2006). Multi-component biocide solutions based on borate with either 0.1% azole or 0.5% thujaplicin are also used against subterranean termites and several fungi (Verma et al., 2009).

3.3.2 Termite control products not applied to wood

Termite control products can be used as preventive or curative treatments when applied away from wood.

3.3.2.1 Termites' Preventive treatments

Instead of applying wood preservatives (PT08), one can use a barrier to block the pathways subterranean termites use to enter the structure from underground. Most barriers are made of insecticides (PT18) combined with polymers or other materials (European Chemicals Agency., 2018b). The treatment may be applied by injection all around the building before or after the construction. Prior to the beginning of building construction, the insecticide is uniformly applied to the entirety of the designated area intended for the establishment of the structure. After completing the construction, the insecticide is injected into the soil surrounding the structure. The pest control operator applies pesticide in a manner that aligns with the wall in a parallel line (OECD, 2008).

3.3.2.2 Termites' curative treatments

Apart from PT08, termites' curative treatments consisting of chemical barriers or bait systems are currently used in Europe. Chemical barriers get rid of subterranean termites from the construction and preserve it from further damage for a couple of years. They have no effect on the nest that is in the soil, though (European Chemicals Agency., 2018b). Different brands of termicides are present around the world, which mainly contain bifenthrin, chlorfenapyr, cypermethrin, fipronil, imidacloprid and permethrin as active substances (Verma et al., 2009). Bait systems can help in completely eliminating the colony. Generally, it is formed of a wood or cellulose matrix coated with a slow-acting insecticide that can reach the whole colony via trophallaxis (European Chemicals Agency., 2018b).

The building envelope is affected by biocides contained in product types (PT) 02 from group 1, PT 06, 07, 08, 09, 10 from group 2, and PT18 from group 3. The various types are outlined in Table 3, along with their primary functions, and the subsequent sections will provide further elaboration on each type of product.

Table 3. Types of biocides used in buildings and their main function

Type de biocide	Naming of uses (MEDD, 2011)	Function	Reference
PT02: Disinfectants and algaecides	Algicide, fungicide, disinfectant	Disinfect the surfaces materials equipment and furniture without direct contact with food or feed	(INERIS, 2012)
PT06: In-can preservatives	Preservatives of inks, paints and adhesives	preserve manufactured products by controlling microbial changes to ensure their long-term preservation. Food, animal feed, cosmetics, pharmaceuticals, and medical devices preservatives are not included in this type.	(INERIS, 2012)
	Preservatives of polymers and plastics		
	Preservatives of biocidal products (e.g. rodenticide baits, insecticide gels, wood protection products)		

PT07: Film preservatives	Preservatives of adhesives, glues, coatings, sealants and papers Preservatives of paints and varnishes	protect films or coatings by preventing microbial alterations or algal development, so preserving the original surface quality of materials such as paints, plastics, waterproof coatings, etc.	(INERIS, 2012)
PT08: Wood preservatives	Preservatives against the large beetle Preservatives against the small beetle Preservatives against the cubic rot (basidiomycete, fungus) Preservatives against fibrous rot (basidiomycete fungus) Preservatives against soft rot (fungus) Preservatives against blue stain fungus of fresh wood Preservatives against blue stain fungus in service Preservatives against house beetles (insects of the order Coleoptera) Preservatives against hesperophanes (insects of the order Coleoptera) Preservatives against fresh wood insects (insects of the order Coleoptera) Preservatives against wood boring insects (Coleoptera). Does not cover termites. Preservatives against lyctus (insects of the order Coleoptera) Preservatives protection against moulds (fungi) Preservatives against termites (insects)	- preventive treatment prevents insects and other organisms from destroying or deforming wood, especially during the sawmill's processing stage. - curative treatment is also utilized to eradicate microorganisms that have already infected the wood and to stop harmful microorganisms from spreading to other parts of the wood.	(INERIS, 2012) (European Chemicals Agency., 2018a)
PT09: Fibre, rubber and polymerised materials preservatives	Preservatives of polymerized materials and fibers	prevent microbial degradation in fibrous or polymerized materials like leather, rubber, etc.	(INERIS, 2012)
PT10: Construction material preservatives	Curative products for the preservation of roofs, walls and facades Products for the preservation of metals including iron and aluminum Products for concretes, mortars or plasters Preventive products for the protection of roofs, walls and facades Products for the protection of other building materials	protect masonry, composite materials or construction materials other than wood against microbiological attack and algae.	(INERIS, 2012)
PT18: Insecticide, Acaricides & other Biocidal Products against Arthropods	Termite control products not applied to wood	control arthropods such as insects, and arachnids by means other than repelling or attracting them.	(INERIS, 2012)

3.4 PT02: Disinfectants and algaecides

Disinfectants and algaecides (PT02) are biocidal products used to disinfect air, water, surfaces, materials, equipment, and furniture that are not used in direct contact with food or feed. On the building envelope, they can be found on walls and balconies floors in private, public, industrial, and other workplaces, as well as being incorporated in paints to produce treated components with disinfectant properties (ECHA, 2023a). As of today, the BPR includes 125 active substances categorized as PT02, as summarized in Appendix a (ECHA, 2023c). These active substances have the following statuses:

- 2 Commission decision (participant withdrawal)
- 64 Initial application for approval in progress
- 29 Approved
- 1 Approved Renewal in progress
- 1 Cancelled application
- 21 No longer supported
- 7 Not approved

3.5 PT07: Film preservatives

The proliferation of fungi and algae on painted surfaces leads to their discoloration as well as the formation of cracks. So, to prevent microbiological deterioration, film preservatives (PT07) are added to the paints during the production process or applied to the final coating material. The various fungal and algae species call for combinations of 3 to 5 biocides, resulting in a total concentration of almost 0.5% in renders and exterior paints (Burkhardt et al., 2011). The most common film-preserving classes of biocides are triazines, urea derivatives, isothiazoline-3-one derivatives, sulfenic acids, dithiocarbamates, benzimidazole derivatives, benzothiazole derivatives, sulfones, carbamates, thiophthalimide derivatives, triazoles, and pyridine-N-oxide derivatives (Jungnickel et al., 2008). Carbamates are well known as fungicides, triazines and phenylureas as algicides, and isothiazolinones as bactericides (Bester et al., 2014). Terbutryn, diuron, and octylisothiazolinone are usually mixed together and/or with other substances in coatings (Linke et al., 2021). As of today, 37 active substances, summarized in Appendix c, are included in the BPR as PT07 (ECHA, 2023c). These active substances have the following statuses:

- 6 Approved
- 1 Commission decision (participant withdrawal)
- 20 Initial application for approval in progress
- 1 Approved Other updates in progress
- 6 Not approved
- 3 No longer supported

3.6 PT06: In-can preservatives

In-can preservatives protect products from microbial growth, increasing their shelf life during storage before utilization. The most prevalent sources of in-can microbial growth include contaminants, bacteria, and yeast in the raw materials (Paulus, 2004). On a building envelope, they may be found in products used as surface coating, which refers to any substance that may be applied as a thin continuous layer on a surface for decorative and/or protection reasons. It may be paints, lacquers, varnishes, etc. (Van der Aa et al., 2004). In-can preservatives are isothiazolinones with a low molecular weight that are more hydrophilic than film preservatives (Kiefer et al., 2023). Early in the 1980s, coatings frequently contained a mixture of 5-chloro-2-methylisothiazolin-3-one (CIT) and 2-methylisothiazolin-3-one (MIT). At the beginning of the 21st century, CIT/MIT was still present in most water-based paints, with an interest in substituting it with MIT or 1,2-benzisothiazolin-3-one (BIT)/MIT mixture in Western Europe. The formaldehyde releasers are another group of active substances that have been widely employed, either alone or in combination with CIT/MIT. They may be combined with 3-Iodopropinylbutylcarbamate (IPBC), BIT or MIT to broaden their activity spectrum (Paulus, 2004). Currently, the BPR for biocides PT06 includes 61 active substances, which are summarized in Appendix b, are included in the BPR for biocides PT06 (ECHA, 2023c). These active substances have the following statuses:

- 11 Approved
- 5 Commission decision (participant withdrawal)
- 3 Cancelled application
- 1 Expired (no application for renewal of approval)
- 31 Initial application for approval in progress
- 4 Not approved
- 6 No longer supported

The question with in-can preservatives is whether they remain present in the coating or whether they leach shortly after the coating application on a surface. Under natural weather conditions on render equipped walls, (Bester et al., 2014) found that in-can preservatives were completely washed off during a 6-month period. (Schoknecht et al., 2009) observed an initial leaching peak for the BIT used as an in-can preservative, after which it was not detectable in the leaching material. (Styszko et al., 2015) got the same results as (Schoknecht et al., 2009) for methyl- and benzisothiazolinone.

Even if in-can preservatives degrade faster than film preservatives, they are of great importance for the receiving environment. (Kiefer et al., 2023) found that in-can preservatives can leach at very high concentrations for the first few weeks and are toxic for aquatic and sediment organisms even when highly diluted. (Bollmann et al., 2014a) identified large amounts of biocides used as in-can preservatives at irregular intervals in municipal wastewater treatment facilities, showing their importance in the leachate of buildings as well as other sources.

3.7 PT08: Wood preservatives

Biocidal products used for protecting wood from biological degradation are grouped under PT08 (wood preservatives). The types of products used as well as their formulations have changed over time with the evolution of legislation. The first and second generations of wood preservatives were usually based on copper, zinc, and arsenic. Copper and zinc can increase the resistance of wood to fungi, whereas arsenic provides wood with insecticidal characteristics. Examples of historically common wood preservatives include chromated copper arsenate (CCA), creosote, pentachlorophenol (PCP), acid copper chromate (ACC), alkaline copper quat (ACQ), copper azole (CA), copper 8-quinolinolate, copper naphthenate, ammoniacal copper zinc arsenate (ACZA), zinc naphthenate and zinc borate(s) (De Buyck et al., 2021). Until the 1990s, most EU members restricted or completely banned the use of creosote, PCP, and CCA in Europe (Jones and Brischke, 2017). CCA was banned for usage in residential buildings in the early 2000s under Directive 2003/2/EC. Later, the arsenic element in general was banned for different uses, including wood preservation, in 2006 under Directive 76/769/CEE. For creosote, it is currently allowed for railway sleepers and electricity or telecommunications poles, but it is prohibited for buildings (ECHA, 2023d). Concerning the PCP, it was still authorized for exterior wood treatment based on Directive 1999/51/EC until 2008. In fact, this authorization benefited France and some EU members, and it expired at the end of 2008. After that, a maximum concentration of 0.1% for the PCP, its salts, and esters is allowed (INERIS, 2005). As an alternative to these substances, a "new generation" of biocides based on combinations of inorganic and organic chemicals is used. Boron and copper are examples of inorganic elements present in new biocides (Tiruta-Barna and Schiopu, 2011). As for organic substances, azoles, particularly tebuconazole and propiconozole, now occupy a leading position. The combination of various biocides, such as quats and copper, tebuconazole, and propiconozole, can increase their efficiency and spectrum activity (Paijens, 2019).

As of today, 46 active substances, summarized in Appendix d, are included in the BPR as wood preservatives (PT08) (ECHA, 2023c). These active substances have the following statuses:

- 13 Approved
- 1 Approved Other updates in progress
- 14 Approved Renewal in progress (approval end dates have passed, application for renewal of approval is in progress)
- 1 Cancelled application
- 15 Expired (no application for renewal of approval)
- 1 Initial application for approval in progress
- 1 Not approved

3.7.1 Main types of wood preservatives

Wood treatment products may be categorized into three groups based on their composition:

- inorganic or salt-based preservatives
- organic wood preservatives
- distillates from coal tar

3.7.1.1 Inorganic wood preservatives

Inorganic or salt-based wood preservatives are products that have water as an active substance carrier. There are two types: products based on mineral salts and those based on organic metal salts. Products based on organic metal salts, which are generally organic combinations of copper, have the advantage of not being water-soluble and are therefore not very sensitive to leaching. Mineral salt-based products are formulations based on copper, chromium, arsenic, or boron (Xhonneux, 2008). Chromated copper arsenate, alkaline copper quaternary, and copper azole are three of the most often used ones. Other widely used copper compounds include copper HDO, copper citrate, and acid copper chromate. These salts are tending to be replaced by more environmentally friendly products, despite improvements in their effectiveness and the acceleration of fixing processes (Xhonneux, 2008). Rather than copper, sodium silicate- and potassium silicate-based preservatives have a latter mostly derived from natural resources like plants (Construction Chemicals, 2010).

Inorganic chemical compositions can be categorized based on their interaction with the wood:

- **Fixating** biocides that are bonded chemically with the wood.
- **Non-fixating** biocides are highly diffusible, thus requiring paint or lacquer to avoid their excessive leaching from wood surfaces.

3.7.1.2 Organic wood preservatives

Organic wood preservatives contain a combination of two or three active ingredients in an organic solvent, most often derived from petroleum, as well as elements that fix and stabilize the active ingredients in the wood (Xhonneux, 2008).

3.7.1.3 Products obtained by distilling coal

These products provide protection for wood that is intended to remain in contact with the ground. These products still contain many impurities (cresols, pyrrols, polycyclic aromatic hydrocarbons, and phenols) and are reserved for outdoor applications (Xhonneux, 2008).

3.7.2 Wood treatment

Through its lifecycle, wood can be protected from biological degradation either before the infestation (preventively) or after (curatively):

3.7.2.1 Preventive treatment

Preventive treatment inhibits or delays the growth of fungus, bacteria, and wood-boring insects on wood elements. It can be applied by professionals or the general public in situ; however, it is mostly treated by industrials prior to putting it into service. Applying this type of treatment to wood can be done in several ways that can be divided into two major groups: fast superficial treatment and deep slow treatment

3.7.2.1.1 Fast and superficial treatments

Fast and superficial treatments include:

- The **brush-on treatment** involves the application of the substance in two layers using a brush, covering all the planed and/or sanded surfaces of the wood.
- **Treatment through immersion** involves submerging the wood in a solution for a specified duration, which depends on factors such as the wood's properties, the concentration of the solution, and the solution's ability to penetrate the outermost layer of the wood. The immersion period can range from a minimum of 3 minutes to 60 minutes.
- The **spray treatment** involves the application of a solution onto the wood as it traverses a tunnel or into a cabin (Xhonneux, 2008).

3.7.2.1.2 Deep and slow treatments

The treatment fluid is introduced into the wood under conditions of elevated pressure. Wood treatment enhances the durability of wood intended for long-term use. This treatment method theoretically enables thorough penetration of the wood tissues.

- **Treatment in an autoclave under vacuum and pressure** involves the initial step of creating a vacuum to eliminate air from the wood cells. Afterwards, the treatment fluid is aspirated into the autoclave. The wood is soaked by the use of hydraulic or pneumatic overpressure. Following this injection, the solution is subsequently given, culminating in the completion of the procedure by the utilization of suction to facilitate an extensive cleansing of the wood.
- The **vacuum double autoclave treatment** is a modified version of the autoclave treatment method, whereby vacuum and pressure are applied alternately. In this process, soaking is conducted at atmospheric pressure instead of during the pressure phase (Xhonneux, 2008).

3.7.2.2 Curative treatment

Curative treatment is used by professionals or the general public in situ to treat infestations that have already developed on wood structures.

3.8 PT09: Fibre, rubber and polymerised materials preservatives

Preservatives in Product Type 09 (PT09) are used to preserve fiber, rubber, and polymerized materials. These preservatives specifically formulated in this product category impede or inhibit the proliferation of microorganisms, including bacteria, fungus, algae, and molds, on these materials. Consequently, the inclusion of these preservatives serves to prolong the durability of the materials and safeguard against deterioration (ECHA, 2023c). Relevant applications are for:

- fibre (textile and fabrics (wood, cotton)
- leather and hides
- rubber, plastics and polymerised material
- pulp, paper and cardboard.

On a building envelope, PT09 can be found in several exterior building materials, including rubber roofing materials, polymer-based exterior claddings, and other polymerized materials. As of today, 51 active substances, summarized in Appendix e, are included in the BPR as wood preservatives (PT09) (ECHA, 2023c). These active substances have the following statuses:

- 2 Commission decision (participant withdrawal)
- 30 Initial application for approval in progress
- 4 Approved
- 1 Approved Other updates in progress
- 1 Cancelled application
- 9 No longer supported
- 4 Not approved

3.9 PT10: Construction material preservatives

PT10 masonry preservatives preserve and treat masonry or building materials, excluding wood (covered by PT08), by controlling microbiological and algal attack. The products covered by this PT include those used for preserving mortar, concrete, concrete additives, baked clay, slate, etc. (Lassen et al., 2001). In the BPR for biocides PT10 (ECHA, 2023c), there are currently 33 active substances summarized in Appendix f. These active substances have the following statuses:

- 1 Commission decision (participant withdrawal)
- 20 Initial application for approval in progress
- Approved
- 1 Approved Other updates in progress
- No longer supported
- 3 Not approved

3.10 PT18: Insecticide, Acaricides & other Biocidal Products against Arthropods

Product type 18 (PT18) is used to control arthropods such as insects and arachnids by means other than repelling or attracting them. This type of product is further discussed in Section 3.3. As of today, 67 active substances, summarized in Appendix g, are included in the BPR as insecticides, acaricides, and other biocidal products against arthropods (PT18) (ECHA, 2023c). These active substances have the following statuses:

- 30 Approved
- 1 Approved Other updates in progress
- 12 Approved Renewal in progress (approval end dates have passed, application for renewal of approval is in progress)
- 1 Cancelled application
- 2 Expired (no application for renewal of approval)
- 12 Initial application for approval in progress
- 3 Not approved
- 6 No longer supported

4 Transfer of biocides from use to the receiving environment

According to earlier studies, leaching from construction materials is a significant source of biocide contamination in urban waters (Bucheli et al., 1998; Burkhardt et al., 2007). Rain transports the biocides into soil, surface rivers, and streams, where they degrade. The pollution of storm water is of particular concern since precipitation runoff is frequently collected in sewage systems, immediately released into surface water bodies, or infiltrated into groundwater. Many studies detected biocides in different environmental compartments, including urban sewage systems (Bester et al., 2014; Bollmann et al., 2014b; Burkhardt et al., 2011; Masiá et al., 2013), stormwater runoff (Blanchoud et al., 2004; Bucheli et al., 1998), soil (Bollmann et al., 2017a), surface water (Burkhardt et al., 2012; Götz et al., 2010; Paijens, 2019; Quednow and Püttmann, 2009, 2007; Wittmer et al., 2010), urban wastewater treatment plant effluents (Bester et al., 2014; Paijens, 2019), etc.

4.1 Leaching process

During rainy events, biocides present on the surfaces may enter the environment through leaching. According to (Uhlig et al., 2019), the leaching of biocides from coatings, is a multi-step process (Figure 3):

1. When it rains, rainwater diffuses into the coating and travels further within.
2. As water fills the matrix, certain biocidal molecules desorb from their carrier particles and dissolve in water.
3. The molecules in the coat move according to concentration gradients within the layers of the coat, from high to low concentration layers.
4. When exposed to sunlight, the biocides on the top layer can undergo hydrolysis or photolysis.
5. Surface water transports the biocides and transformation products, which are then washed away by runoff.

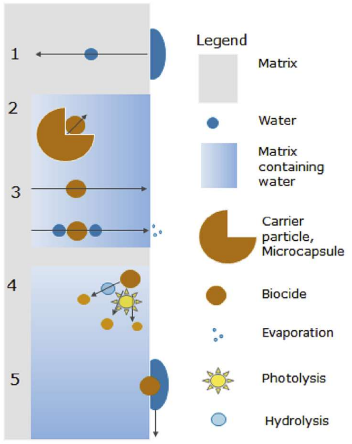


Figure 3. Leaching mechanisms (Uhlig et al., 2019)

4.2 Release of biocides from construction materials

Multiple studies show that emissions are significant immediately following construction (concentrations of the order of several mg/L), but then gradually decline (concentrations of tens of µg/L) (Bollmann et al., 2017b; Burkhardt et al., 2011; Schoknecht et al., 2009; Wangler et al., 2012; Wittmer et al., 2011). Generally, in urban areas, the dynamics of pollutants in stormwater runoff are linked to the first-flush phenomenon, which is defined by (Bertrand-Krajewski et al., 1998) when "at least 80% of the pollutant mass is transported in the first 30% of the volume". This happens when the release of biocides accumulated on the surface or in dust is particularly rapid at first and then slows down afterwards. On the other hand, from their analysis of storm water runoff from a small urban catchment in Switzerland, (Burkhardt et al., 2011) found that the biocides were emitted continuously from the facades, meaning that there is a highly diffuse transport mechanism. This behavior correlates well with laboratory experiment-based models done by (Wangler et al., 2012) and (Schoknecht et al., 2009), which explain a diffusion-based process as the governing mechanism for the release of biocides from render. Bester et al. (2014) found that there is no general trend for the transport of all the tested biocides into stormwater runoff. Out of 12 events, 5 showed linear emission, similarly to the results obtained by (Burkhardt et al., 2011). Terbutryn, methylisothiazolinone, cybutryn, and diuron exhibited first-flush behavior during three events. Iodocarb and cybutryn emitted more at the end than during two separate events (post-flush behavior) (Bester et al., 2014).

4.3 Transport mechanisms

The process of leaching is based on how compounds interact with the matrix and water. The transfer of substances from the matrix into water occurs through various transport mechanisms that can be subdivided into two main groups: chemical and physical.

4.3.1 Chemical processes

Biocides' release is controlled by two distinct chemical mechanisms: mineral dissolution and adsorption processes.

4.3.1.1 Dissolution (solubility control)

Biocides have the potential to gradually dissolve into the adjacent water. The pace at which dissolution occurs is dependent upon many factors, including the solubility of the biocide and the pH level of the surrounding environment. Biocides with high solubility exhibit a greater tendency to dissolve rapidly, hence augmenting their potential for leaching.

4.3.1.2 Adsorption process (sorption control)

Biocides have the ability to adsorb, or adhere, to various components, such as soil particles, surfaces, or organic materials within the surrounding environment. This phenomenon has the potential to decrease their mobility and leaching capacity. The degree of adsorption is dependent upon the chemical characteristics of the biocide as well as those of the materials with which it interacts. For instance, heavy metal cations, which are not regulated by the dissolution of a mineral, exhibit a tendency to adsorb onto reactive surfaces. These surfaces can include organic material or oxide surfaces, which possess a negative charge (van der Sloot and Dijkstra, 2004). Following adsorption, biocides have the potential to undergo desorption, a process in which they separate from surfaces and regain mobility. It is subject to the effects of several parameters, including temperature, pH, and the existence of competing ions within the surrounding environment.

4.3.2 Physical transport processes

Physical transport mechanisms, in addition to chemical ones, determine the transfer of components from the substance to the aqueous phase. Three main types of transport mechanisms will be briefly discussed here

4.3.2.1 Advection

Advection refers to the transportation of elements alongside rainwater as it percolates through or travels along a medium. Currents, tides, and flow facilitate the transport of biocides across significant distances in water bodies through the process of advection. The phenomenon of percolation may only occur in materials that possess porosity, such as granular substances (van der Sloot and Dijkstra, 2004).

4.3.2.2 Surface wash-off

Surface wash-off and advection are similar mechanisms. The term "surface wash-off" pertains to the first removal of soluble substances from the outer surface of the products (van der Sloot and Dijkstra, 2004).

4.3.2.3 Diffusion

Diffusion is the spontaneous and random movement of particles under a concentration gradient from a high concentration area to a low concentration area. The phenomenon occurs over time and is influenced by the difference in concentration of components between the matrix and the aqueous phase with which it is in contact. Prior to the transfer of components due to increased advection, diffusion may serve as the constraining factor (Schoknecht and Töpfer, 2013). In the absence of flow, diffusion is the only mechanism present for the transfer of components. Frequently, dense materials with exceptionally low porosity and permeability often exhibit this phenomenon, as the presence of stagnant water within the pores becomes particularly significant. The process of release will persist, although it occurs via diffusion-based transport.

The dimensions and shape of a product largely influence the rate at which a material diffuses and leaches from it. The extent of the product's exposed surface area is a crucial determinant in the process of diffusion, closely correlating with the aforementioned characteristics. Substances with a greater surface area per unit of weight accelerate the rate of diffusion (van der Sloot and Dijkstra, 2004).

4.4 Parameters influencing the leaching of biocides from buildings

A variety of factors affect the leaching of biocides from buildings. In the following sections, we will concentrate on the most important ones.

4.4.1 Initial conditions

The initial amount of biocidal material in the coating influences the leaching processes. The conditions of spreading the treatment affect the formation of the stock. Mass, concentration, and volume define the size of the mobilizable stock. Furthermore, distributing higher volumes of product can alter the position of the stock, affecting both the stock that penetrates deep into the surface and the stock that is ready to be mobilized (Van de Voorde et al., 2012). Encapsulation of biocides is also an essential component. Encapsulation compositions began to be used in construction materials in 2006. This method delays the release of biocides and keeps them on the surface for a longer period of time. The capsules frequently comprise a variety of organic or silica-based polymers, within which biocides can be effectively absorbed. Alternatively, the biocides may be present as nanoparticles that are disseminated throughout the paint matrix (Junginger, 2022). (Vermeirssen et al., 2018) produced eluates from a render system that was both encapsulated and non-encapsulated and subsequently examined the toxicity of these eluates. They demonstrated that encapsulated biocides exhibited reduced leaching concentrations in comparison to free biocides, resulting in eluates having less toxicity.

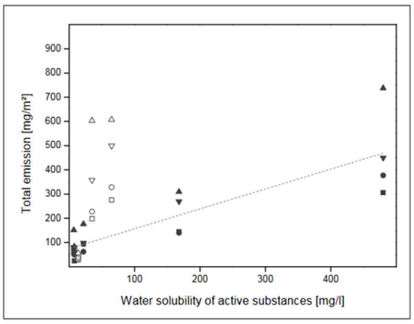
4.4.2 Active substance properties

The release of biocides in water is affected by the chemical properties of the active substances as well as their interactions with the matrix. It is affected by the solubility of the biocide in water and the partitioning coefficients, which can differ considerably from one to another. Table 4 presents the water solubility and octanol-water partition coefficient (log k_{ow}) of commonly used active substances.

Table 4. Water solubility and octanol-water partition coefficient (log Kow) of commonly used active substances

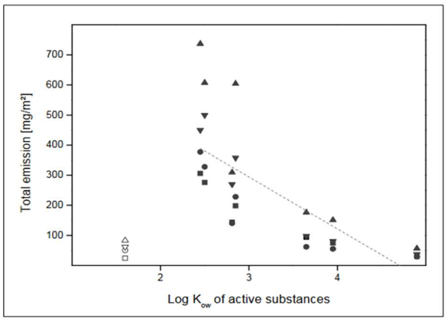
Active substance	Water solubility (mg/l)	Log K_{OW}	Reference
Carbamates			
Carbendazim	8	1.51	(Tomlin, 2009)
IPBC	168	2.81	(Paijens, 2019)
Phenylureas			
Diuron	37	2.85	(Tomlin, 2009)
Isoproturon	65	2.5	(Paijens, 2019)
Isothiazolinones			
OIT	480	2.45	(Paulus, 2004)
MIT	96.1	-0,486	(Paijens, 2019)
BIT	1000	0.64	(Paijens, 2019)
DCOIT	14	4.9	(Paijens, 2019)
Triazoles			
Tebuconazole	97	3.7	(Paijens, 2019)
Propiconazole	11	3,72	(Paijens, 2019)
Triazines			
Terbutryn	22	3.65	(Tomlin, 2009)
Cybutryn, Irgarol 1051	7	2.8	(Paijens, 2019)
Miscellaneous			
Mecoprop	471	0.1	(Paijens, 2019)

The solubility of the biocide in water and the partitioning coefficients differ considerably from one biocide to another. (Schoknecht et al., 2009) conducted laboratory tests to study the influence of water solubility and the octanol-water partition coefficient (log K_{ow}) of commonly used active substances on their leachability. The compounds were leached from textured façade coatings applied to polystyrene test specimens using intermittent wet/dry cycles. They found that emission rates and total emissions increased with the water solubility (in an ascending order of water solubility: Cybutryn, Carbendazim, DCOIT, Terbutryn, Diuron, Isoproturon, IPBC, OIT) and decreased with the log Kow (in an ascending order of log Kow : Carbendazim, IPBC, OIT, Isoproturon, Diuron, Terbutryn, Cybutryn, DCOIT) (Figure 4 and Figure 5). However, these two parameters don’t explain the leachability of biocides on their own even under controlled laboratory conditions; they are influenced by other factors.



Active substance	*water solubility (mg/L)*
BIT	1000
OIT	480
DCOIT	14
IPBC	168
Carbendazim	8
Isoproturon	65
Diuron	35
Terbutryn	22
Cybutryn	7

Figure 4. Effect of different active compounds' water solubility on total emission of (Schoknecht et al., 2009) laboratory tests ((Schoknecht et al., 2009) cited in (Schoknecht and Töpfer, 2013))



Active substance	*log Kow*
BIT	0.64
Carbendazim	1.6
OIT	2.45
Isoproturon	2.5
Cybutryn	2.8
IPBC	2.81
Diuron	2.85
Terbutryn	3.65
DCOIT	4.9

Figure 5. Effect of different active compounds' log Kow on total emission of (Schoknecht et al., 2009) laboratory tests ((Schoknecht et al., 2009) cited in (Schoknecht and Töpfer, 2013))

On the other hand, (Styszko et al., 2015) deduced that the concentrations of biocides in the water are not controlled by the solubility of these chemicals in water. High biocides concentrations in the leachate were found to be significantly lower than the solubility of the compounds in water (i.e., terbutryn concentrations reached in water are 0.38 mg/L for silicone render and 5.9 mg/L for acrylic render, while its water solubility is 25 mg/L).

Furthermore, the interactions between active substances and materials are influenced by structural similarities, namely the presence of functional groups. These functional groups play a crucial role in processes such as adsorption onto material components and the rate of chemical reactions with these components (Schoknecht and Töpfer, 2013). Biocides of the same class may exhibit comparable leachability even when having different water solubilities as for phenylureas and triazines (Burkhardt et al., 2012, 2009; Schoknecht et al., 2009). For instance, the water solubility of the triazines Terbutryn and Irgarol 1051 is 22 mg/l and 7 mg/l, respectively. Despite this difference in solubility, both compounds were found to be leached to similar extents from façade coatings in laboratory leaching experiments (Schoknecht et al., 2009) as well as in a weather chamber (Burkhardt et al., 2009).

4.4.3 Organic matter content of coating

The organic content of the coating influences the emission rate. Coatings with a greater organic content (acrylic renders) emit more biocide than those with a lower content (silicon renders) (Junginger, 2022). (Bollmann et al., 2017a) found significantly higher OIT concentrations in the leachates from the acrylate renders than those from the silicone renders (Bollmann et al., 2017b). Same results were gotten by (Styszko et al., 2015) for different types of biocides (i.e., terbutryn concentrations reached in water are 0.38 mg/L for silicone render and 5.9 mg/L for acrylic render; isoproturon concentrations reached in water are 1.61 mg/L for silicone render and 2.03 mg/L for acrylic render). Concerning the transport fluxes of biocides, (Styszko et al., 2015) found that a more consistent delivery of biocides to the material's surface may be anticipated from the silicone render than from the acrylate render.

4.4.4 pH

The leaching of biocides can be considerably influenced by the pH of both the substance itself as well as its surrounding environment. Some pH ranges can increase the solubility and mobility of some biocides, resulting in a greater potential for leaching under specific conditions.

4.4.5 Structure properties

When considering release behavior, it is necessary to differentiate between two distinct categories of products. The materials under consideration can be classified into two categories: monolithic and granular. Monolithic materials often release components primarily through diffusion, indicating diffusion-controlled release mechanisms. On the other hand, percolation dominates the release mechanisms in granular materials, where constituents are released due to the percolation of water through the product. Monolithic products encompass a range of cementitious materials, such as concrete, bricks, and coated materials (van der Sloot and Dijkstra, 2004).

Substances with high permeability allow water to enter easily and pollutants to be released quickly over time. Permeability may be a problem for monolithic materials (van der Sloot and Dijkstra, 2004). Water generally circulates around low-permeability materials instead of entering them, which is why they exhibit diffusion-controlled release rather than advection (Schoknecht and Töpfer, 2013). Porosity significantly influences the rate at which components are transported towards the aqueous phase. A higher porosity often results in a larger release because it is simpler for water to transport through high-porosity material than through low-porosity media (van der Sloot and Dijkstra, 2004).

Other than all the previously mentioned surface properties, a crucial factor in determining the actual exposure to weathering of inclined and vertical surfaces is their exposure orientation. Orientation has a great influence on the amount of biocides leached into the runoff. (Vega-Garcia et al., 2020) proved that the biocide loads found in the runoff were the highest for the façades facing the predominant weather orientation.

4.4.6 Environmental factors

The leaching process is influenced by environmental and climatic conditions (rain intensity, total precipitation, wind direction and speed, temperature, UV irradiation, etc.). (Bester et al., 2014; Schoknecht et al., 2016) consider that the critical factor influencing the leaching of chemicals is the amount of water in contact with exposed surfaces. The emission of biocides is mainly related to the amount of water reaching the surface, which depends on the wind-driven rain as well as the rain's intensity. The temperature also has an influence on the leaching of biocides. The rise in temperature promotes diffusion and so increases the rate of biocide emission (van der Sloot and Dijkstra, 2004; Wangler et al., 2012). Increased solubility is typically the result of rising temperatures.

UV irradiation can cause the photodegradation of active substances present in the biocide (Bollmann et al., 2017b, 2016; Jirkovský et al., 1997; Schoknecht et al., 2009). (Jirkovský et al., 1997) described the photodegradation products of diuron and (Bollmann et al., 2016) of terbutryn. In Schoknecht et al. (2009) laboratory tests, lower concentrations of OIT, DCOIT, IPBC, isoproturon, diuron, terbutryn, and cybutryn were detected in UV-exposed specimens, implying photolysis of these substances. Bollmann et al. (2017b) performed a laboratory experiment in dissolved water using UV light that confirmed the photodegradation of OIT into mainly seven transformation products. Then, they studied its photodegradation on artificial facades and artificial walls under natural conditions. On a 19-month period, the majority of the OIT was still in the coating, with a large variation of concentrations from event to event generally following a decreasing trend; however, no trend was found for the degradation products. Also, the photodegradation products made up less than 30% of the OIT mass balance on acrylate and 40% of the OIT mass balance on silicone render. This is because DCOIT is present and breaks down in the same way as OIT (Bollmann et al., 2017b).

4.5 Persistence in the environment

Biocide persistence in an environment refers to its ability to remain stable and active over time. The degradation of the substance depends on several factors related to the substance itself as well as the characteristics of the environment. The half-life of a compound reflects its persistence in a given environment. Table 5 presents the half-lives of commonly used active substances in different environments. Some substances are highly persistent, such as diuron, while others aren't (e.g., dichloro-octylisothiazolinone). Also, the same compound may persist more in one environment than in another.

Table 5. Half-life of commonly used active substances in different environments

Biocide	Environment	Half-life (days)	Reference
Terbutryn	Soil	231	(Bollmann et al., 2017a)
	Water under aerobic conditions	193–644	(Talja et al., 2008)
	Water under anaerobic conditions	266–400	(Talja et al., 2008)
Diuron	Soil	> 2500	(Bollmann et al., 2017a)
	Sea water	Persistent	(Ekblad, 2014)
OIT	Soil	9.3	(Bollmann et al., 2017a)

		immobile but microbial degradation after 120 days	(Ekblad, 2014)
	Water	>30	
Isoproturon	Soil	40	(Ekblad, 2014)
	Water	30	
DCOIT	Water	<1	(Ekblad, 2014)
Cybutryn	Sea Water	100-250	(Ekblad, 2014)
	Soil (anaerobic sediments)	persistent	
Carbendazim	Soil	3-12 months	(Ekblad, 2014)
	Aerobic water	2 months	
	Anaerobic water	25 months	
IPBC	Water	139	(Ekblad, 2014)
Tebuconazole	Water	28	(Paijens, 2019)
Propiconazole	Water	30	(Paijens, 2019)
Mecoprop	Water	31	(Paijens, 2019)

5 Modelling the transfer of biocides in cities

After having a deep overview of the characterization and modeling of the emission of biocides in the water runoff of roofs and facades and developing a methodological framework for the evaluation of the spatial distribution and characterization of the stock of biocides present in the building's envelope, the third and fourth parts of the thesis will deal with the modeling of runoff and leaching processes and implement the modeling framework in a distributed hydrological model. The modeling of runoff and leaching processes will be based on the principles of the COMLEAM (COnstruction Materials LEAching Model) (www.comleam.ch, (Burkhardt et al., 2020)), which simulates rainfall interception by building facades and roofs and integrates various research-based leaching law models. The proposed modeling framework will be implemented in TEB (Masson, 2000; Stavropulos-Laffaille et al., 2021, 2018), a distributed hydrological model adapted for the simulation of the urban environment. The following section will provide an overview of the two models to be used: COMLEAM and TEB.

Commenté [AB6]: Préciser d'où vient le modèle. Mettre une référence comme tu l'as fait pour COMLEAM

5.1 COMLEAM

COMLEAM is a software developed at HSR (HOCHSCHULE FÜR TECHNIK RAPPERSWIL) for modeling the leaching of hazardous substances from vertical and horizontal building elements exposed to weather conditions and their entry into the receiving environment. Its major purpose is to offer a tool for analyzing and forecasting the leaching behavior of building materials, allowing for informed decisions about material selection, construction procedures, and environmental risk assessment. COMLEAM models the leaching process and gives information about how it might affect nearby soil, groundwater, or surface water by using the material properties, weather information, and exposure conditions (Burkhardt et al., 2020). The software is based on a dynamic simulation of time-limited leaching from construction materials, buildings, and cities using lab or field data and the occurrence of substances in surface waters. Figure 6 illustrates the representation of the COMLEAM, showing the water and substance flow from a component to the target compartment (soil or receiving surface water).

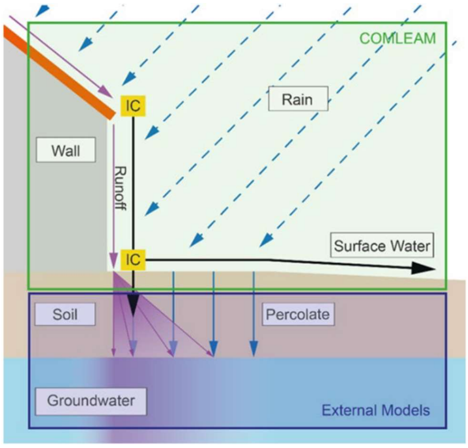


Figure 6. COMLEAM model representation (Burkhardt et al., 2020)

5.1.1 Model Structure

COMLEAM represents a platform for estimating leaching and environmental exposure for predefined or user-defined boundary conditions (substances, emission functions) and scenarios. The software is divided into user-managed modules (weather, geometry, material and substance, emission function) whose data is merged in the calculation core, and the results are read from the database automatically (Figure 7). The results are presented in a report that includes the most important simulation parameters, details about the geometry, materials, and runoff coefficients, emission function parameters and initial concentrations, water and substance balances for each component, and emissions into surface water.

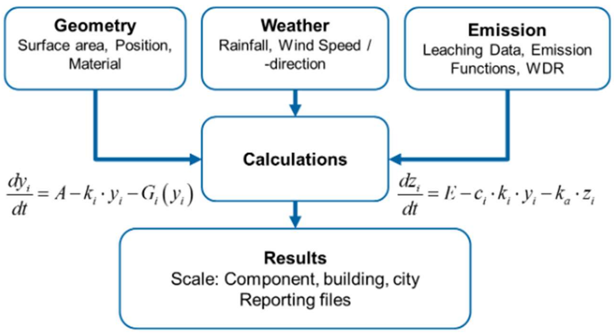


Figure 7. COMLEAM Structure

The four COMLEAM modules are presented and described in depth in the following sections.

5.1.1.1 Weather data module

Precipitation is used to compute the water impinging on horizontal components, while driving rain is used for vertical components. Weather data are in hourly resolution, as it's a requirement for WDR (wind driven rain) calculation.

Commenté [AB7]: Expliciter ce que c'est

5.1.1.2 Geometry Module

The geometry module assigns a specific identification number to each building and describes them in terms of orientation, dimensions, and materials. Each component characterizes a building:

- size
- orientation (in degrees from North)
- ground angle (between 0° for horizontal and 90° for vertical members)
- proportion of material type (in %) (Each component can be made of different building materials such as glass, plastic, mineral building materials, etc., and the relative area fraction is assigned. For example, a building component reflecting a building façade may include a surface proportion of 20% glass and 80% mineral plaster).

5.1.1.3 Material and substance

A material describes a construction product. Subtypes (e.g., plaster, paint, fair-faced concrete) and a substance (e.g., Diuron, DCOIT) with the initial contents are assigned to each material type (e.g., metal, wood, glass, plastic). The subtypes include specific runoff coefficients, which describe the runoff-effective portion of precipitation as a summary loss of water absorption of the material, evaporation, and rebound of the water.

5.1.1.4 Emission data module

In COMLEAM, emission data can be implemented via measured data, which describe the substance emission [mg/m^2] as a function of the accumulated amount of water [L/m^2], from which COMLEAM is able to parametrize emission functions. Otherwise, the emission functions must be directly parametrized on COMLEAM through parameters that were previously derived by regression from the measured data (field / laboratory).

5.1.1.4.1 Emission functions

The relationship between a component's outflow and the resulting emissions is mapped as a key aspect of the modeling in the emission module. An emission function is used to characterize this connection. When assessing building items or chemicals, realistic and believable simplifications are needed, which is what the emission functions do (Tietje et al., 2018). The emission function describes the cumulative amount of substance emitted by a component (mg/m²) as a function of the cumulative amount of runoff

(L/m²). (Burkhardt et al., 2020) mentioned in the COMLEAM manual the conditions that a function should have to be representative of the emission of substances from the surface:

- "The function increases monotonically;
- Start at the origin (i.e., at the point (0;0));
- The slope of the function must constantly decrease (monotonically decreasing);
- The quantity released is smaller than the initial amount in the component;
- The function must be fitted to the available data as best as possible (least squares);
- The function should be as universal as possible, i.e., various materials and substances should be described by the parameters without changing the shape of the function."

COMLEAM provides six functions of type T as follows:

- Logarithmic function
- Michaelis-Menten kinetics
- Langmuir desorption function
- Limited growth function
- Diffusion-controlled function

For all emission functions the following applies:

$$E(t) = c_0 . E_T(t)$$

Where

$E(t)$ cumulated Emission quantity per square meter area [mg/m²]

c_0 initial applied amount per square meter area [mg/m²]

$E_T(t)$ dimensionless emission function (0 ≤ $E_T(t)$ ≤ 1) of type T

The mathematical definition of the emission functions and their parameters, as well as an explanation of their advantages and disadvantages, are provided in the sections that follow.

5.1.1.4.1.1 Logarithmic function

In COMLEAM, the logarithmic emission function is represented as follows:

$$E_{log}(q) = a_{char} \times \ln\left(1 + 1.72 \frac{q}{q_{char}}\right)$$

Where

q_{char}: characteristic discharge [Lm²]

a_{char} : characteristic substance fraction representing the proportion of the applied biocide quantity emitted up to the characteristic discharge q_{char} [dimensionless]

$q = q(t)$: Cumulative runoff water volume [L/m²]

In order to develop a comprehensive understanding of the physical meaning of these parameters, we will refer to the examples provided by (Tietje et al., 2018). Given that achar = 0.5, the characteristic discharge q_{char} represents the "half-value discharge," which corresponds to the quantity of water required for half of the applied substance to be released. Another example is assuming an initial applied quantity of c_0 = 1000 mg/m². Consider the logarithmic emission function with parameters a_{char} = 0.01 for the characteristic substance percentage and q_{char} = 10 L/m² for the characteristic discharge. Once a q_{char} of 10 L/m² has flowed off the façade, the total emitted amount from the façade up to that point is E = c_0. a_{char} = 1000*0.01 = 10 mg/m². The logarithmic function demonstrates that 50% of the initial amount is released after a runoff quantity of 1.69*10^9 L/m² is reached.

5.1.1.4.1.2 Langmuir desorption & Michaelis-Menten kinetics functions

The Langmuir function, named after Irving Langmuir, is a mathematical model used in surface science, catalysis, and other adsorption processes. It describes the adsorption behavior of molecules on surfaces, illustrating the relationship between adsorbate amount and gas or liquid phase concentration. The Langmuir sorption-desorption function is also used in soil mass transport modeling to measure the equilibrium between substance sorption and desorption under limited sorption sites. It is utilized to estimate the maximum possible emission amount, which may be determined using the following expression:

$$E_{Langmuir}(q) = a_{Langmuir} \cdot \frac{b_{Langmuir} \cdot q}{1 + b_{Langmuir} \cdot q}$$

The Michaelis-Menten equation is equivalent to the Langmuir equation; it differs only by using the inverse of a parameter. It is employed in pharmacokinetics to determine the rate at which a medication is released throughout the body. The Michaelis-Menten equation is expressed as follows:

$$E_{MM}(q) = a_{MM} \times \frac{q}{K_{MM} + q}$$

The parameters, shown in Table 6, have a physical meaning in terms of the proportion of the applied amount that is released and the discharge quantity, at which point half of the emission has occurred.

Table 6. Langmuir emission function and Michaelis-Menten kinetics parameters

Parameter	Unit	Range	Meaning
$a_{MM} = a_{langmuir}$	dimensionless	0<a<1	Proportionality factor for the proportion of the applied quantity that is available for emission.
$b_{Langmuir}$	m²/l	$b_{Langmuir} > 0$	$b_{Langmuir} = \frac{1}{K_{MM}}$

K_{MM}	L/m²	$K_{MM} > 0$	$K_{MM} = q_{1/2}$ is the discharge quantity after which half of the emission has taken place
q= q(t)	L/m²	q>0	Cumulative runoff water volume

The Langmuir function is based on several assumptions (Ye et al., 2021):

- Adsorption occurs on a homogeneous surface with a finite number of identical adsorption sites;
- Total adsorption occurs when all the sites are covered by a monolayer of adsorbed substance;
- Each site can receive 1 molecule, all sites are equivalent, and the surface is without roughness;
- The occupation of the surrounding sites does not influence adsorption on a site;
- The surface does not undergo any structural changes upon adsorption.

The Langmuir emission function has a favorable aspect as it functions as a desorption function in emission analysis, making use of two separate physical features. By conceptualizing it as desorption, it enables a thorough comprehension of the complete emission process. Nevertheless, there is a significant problem associated with it: the Langmuir emission function has a tendency to forecast somewhat lower emissions over prolonged time periods in comparison to the logarithm function. It is important to take this constraint into account when selecting an emission modeling method, since it might affect the precision of forecasts, particularly in situations where longer time periods are important (Tietje et al., 2018).

5.1.1.4.1.3 Limited growth function

The limited growth function presupposes a continuous decrease in emissions. It is expressed as follows:

$$E_{LG}(q) = a_{LG}(1 - e^{-b_{LG} \cdot q})$$

The parameters, shown in Table 7, have physical significance as they consider the percentage a_{LG} of the applied amount used for emission and the discharge quantity $q_{1/2}$, which represents the point at which half of the emission has occurred.

Table 7. Limited growth function parameters

Parameter	Unit	Range	Comments
a_{LG}	Dimensionless	$0 < a_{LG} < 1$	Proportionality factor for the proportion of the applied quantity that is available for emission.
b_{LG}	m²/L	$b_{LG} > 0$	$b_{LG} = \frac{\ln(2)}{q_{1/2}} \approx \frac{0.69}{q_{1/2}}$ is calculated from the discharge quantity after which half of the emission has taken place.
q= q(t)	L/m²	q>0	Cumulative runoff water volume

One advantage of the function is its simplicity, since it just has two arguments that have obvious and easily understandable physical interpretations. The model accurately replicates the known emission patterns, especially when emissions demonstrate a declining tendency over time. In addition, the constrained growth function is designed to be user-friendly, utilizing an exponential function that simplifies its implementation. Nevertheless, it is important to recognize some constraints. The function may not be appropriate for conventional experimental data, thereby limiting its usefulness in some situations. Furthermore, as compared to the logarithmic function, the restricted growth model tends to forecast somewhat lower emissions over extended periods. This suggests that effectively capturing emission patterns, particularly when a long-term view is crucial, may provide issues (Tietje et al., 2018).

5.1.1.4.1.4 Diffusion-controlled function

Diffusion is the spontaneous and random movement of particles under a concentration gradient from a high concentration area to a low concentration area. The diffusion-controlled function is a mathematical representation of diffusion-governed processes in which the rate is primarily determined by the rate of diffusion. It highlights the importance of diffusion in influencing a process's overall rate. However, it is a simplification that does not account for other factors that may impact the process, such as electrostatic interactions or molecule crowding. Nonetheless, it provides a valuable framework for understanding and studying systems in which diffusion plays a dominant role in determining the pace of the process.

Scientists widely use this function in the domain of substance leaching to represent the release or migration of molecules from a solid material into a surrounding medium, such as soil or water. In this application, it specifies the rate at which the substance diffuses through the medium and reaches the surrounding environment. The specific process under consideration determines the overall form of a diffusion-controlled function. In COMLEAM, the diffusion-controlled function has the following form:

$$E_{Diff}(q) = a_{Diff} \cdot \sqrt{q}$$

Where

a_{Diff} $[\frac{m}{\sqrt{L}}]$: parameter calculated from the runoff amount q1/2 after which half of the emission has occurred.

$q = q(t)$: Cumulative runoff water volume [L/m²]

5.1.1.4.1.5 Linear emission function

When the emission remains constant over time, the linear emission function is used. This occurs when the emissions are significantly negligible in comparison to the quantity of the substance existing on the component, such as copper facades. Here, the value of c_0 is not specified; it is equal to 1. The emission function corresponding to this is a cumulative function, given in mg/m², that exhibits linearity with the emission rate a_{linear} :

$$E_{linear}(t) = a_{linear} \times q(t)$$

Where

a_{linear} : parameter of the linear emission function [mg/l]

$q(t)$: Cumulative runoff water volume [L/m²]

5.1.1.4.2 Calibration and validation

(Tietje et al., 2018) investigated the uncertainties in emission calculation procedures to understand their effects in various scenarios for the Terbutryn. The study was done on the Terbutryn since they had reliable data on this substance leaching as well as its functional relationship, which is similar to other compounds such as diuron, OIT, DCOIT, carbendazim, isoproturon, and IPBC (Burkhardt et al., 2012). They recommended a mathematical form for the emission function based on their results. They also answered questions about the best emission function for describing water runoff and emissions, the factors and parameters influencing results, and how assumptions and simplifications affect long-term risk assessment of substance emissions. The simulations and evaluation of the emission functions were based on three distinct data sets, which are detailed in (Appendix h,Table 11), along with the initial conditions and primary results. Appendix i, Figure 11 shows the results of the field study in Zurich with terbutryn free (approx. 1 year) and terbutryn encapsulated (approx. 2 years) and Ober-Ramstadt (RMI) with terbutryn free (approx. 1 year).

Commenté [AB8]: Juste sur la terbutryn ? si oui préciser et éventuellement généraliser ou critiquer

5.1.1.4.2.1 Emission functions adjustments

The function adjustments were performed based on non-linear least squares. The function parameters for the emission functions are selected to minimize the total sum of squared differences between the measurements and the function values. The logarithmic emission function accurately represents cumulative runoff emission data, with visual perception confirming its average match (Appendix i, Figure 12). The slope of the emission function determines the actual emission, which agrees well with the slope of the data. The region at the end of measurements is crucial for curve fitting, as it determines how the function extrapolates the data. The fitted function shows a high agreement with the log emission function, but slightly overestimates the trend towards the end of the experiment. Compared to the logarithmic emission function, the limited growth emission function gives a more accurate picture of data sets. However, when it comes to computing the growth boundary, it greatly understates emissions, which means it can't be used to guess what future emissions will be (Appendix i, Figure 13). The curve fitting of the diffusion-driven emission function shows that it is not good for accurately describing actual emission data. This is because it predicts too many emissions in this range and the upper range of the measuring time (Appendix i, Figure 14). The Michaelis-Menten function provides a highly precise representation of runoff emission data at both the beginning and end of the measurement period (Appendix i, Figure 15). However, when extended, the functions underestimate the observed trend in the data, making them unsuitable for accurately predicting future emissions and their environmental presence.

5.1.1.4.2.2 COMLEAM simulation

In order to compare the emission functions, particularly with respect to how accurately the functions can track the simulated emission levels, Tietje et al. (2018) simulated the emission of terbutryn at the same sites as the field measurements. The simulation settings are shown in (Appendix h, Table 12). The results are shown in Figure 8. The double-log linear function, shown in Figure 8, won't be discussed since it is a more complicated function, not defined in COMLEAM and not advised to be used based on.

The simulation results for Zurich Terbutryn free data show that over 1.5 years, the emission functions were simulated to estimate the trend of emissions. The results showed that the simulations with limited growth emission functions led to an underestimation of emissions. The logarithm function balanced cumulative emissions at the end of the field test (365 days) minimally higher than the real measured data, but with a tendency to overestimate long-term emission prediction. The Michaelis-Menten emission function showed the best agreement, while the diffusion-driven emission function most strongly underestimated the increase in cumulative emissions between 60 and 260 days and total emissions. However, an underestimation cannot be attributed to the emission function alone. Other reasons for underestimation include the emission function itself, the estimation of wind direction or speed by the ISO standard, and the weather data not completely fitting the discharges. The RMI data were also relatively well approximated, but the diffusion-based function most significantly overestimates the effective washout amount. For the data from the experiments with encapsulated terbutryn, the agreement with the data in the first year was extremely good. However, after a certain range of different emission functions, the logarithmic function was the best fit, this time together with the diffusion function.

Commenté [AB9]: C'est quoi ?

Tietje et al. (2018) also investigated the impact of each function on forecasting terbutryn emissions in plaster containing encapsulated terbutryn over a span of two years. The parameters of the emission functions are determined using the data from the first year, while the emission trend for the second year is projected using the available meteorological data. The findings indicate that the estimation of emissions in the initial year is almost as precise as in other instances. In the second prediction phase of the extrapolation, the range of outcomes for the emission functions experiences a substantial expansion. The limited growth function reaches a plateau at the end of the first year, leading to the most underestimated emissions in the second year. The Michaelis-Menten emission function results in an underestimation of emissions during the extrapolation phase without reaching a plateau following the adjustment period. The logarithmic emission function has the highest level of agreement with the observed data, leading to the least amount of overestimation in the extrapolation. If the only criterion for selecting an emission function for "conservative" extrapolation were the quality of extrapolation in the period from 300 to 600 days following the start of the simulation, both the diffusion-driven and logarithmic emission functions would be very suitable. Nevertheless, the rate of growth of the diffusion-driven emission function surpasses that of the logarithmic emission function.

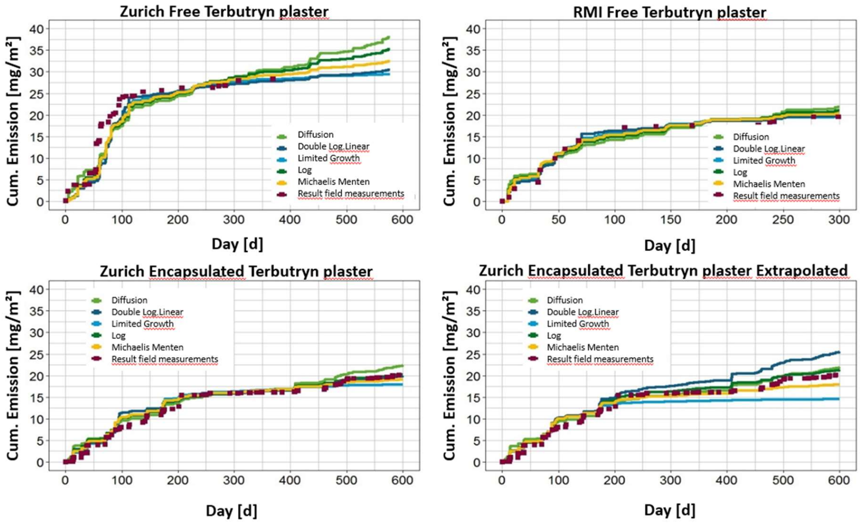


Figure 8. Simulation results Zurich terbutryn free (top left), RMI terbutryn free (top right), Zurich terbutryn encapsulated (bottom left) and Zurich terbutryn encapsulated extrapolation (bottom right) (Tietje et al., 2018)

5.1.2 Hydrologic model

The hydrologic model in COMLEAM refers to the mathematical representation used to study and predict the behavior of water, starting from rainfall until reaching the receiving environment, passing by the building envelope. It helps analyze the movement and distribution of water across different components, more specifically the building envelope as well as the receiving media (soil, surface water).

5.1.2.1 Runoff calculation

Runoff is the quantity of water that flows over the components as a result of rain reaching horizontal or vertical components. Due to several factors, the amount of water that runs off the surface does not match the amount that hits it. It is anticipated to be smaller because of splashes, evaporation, and material characteristics (diffusion of water across the material's surface). The amount of runoff for a component is calculated as follows, depending on the amount of rain reaching the surface and a runoff coefficient that accounts for losses on the surface:

Commenté [AB10]: Splashes ?

$$q(t) = \psi . r(t) \quad \text{or} \quad q(t) = \psi . r_{SR}(t)$$

Where

$q(t)$: cumulated amount of runoff (L/m²)

ψ : runoff coefficient – dimensionless $0 < \psi \leq 1$

$r_{SR}(t)$: for vertical components, cumulated amount of wind driven rain (WDR) (L/m²)

$r(t)$: for horizontal component, cumulated amount of precipitation (L/m²)

The amount of rain reaching a component depends on whether the component is vertical or horizontal. Horizontal components receive the actual rainfall directly, while vertical ones receive rain driven by the wind, which will be discussed in the following section.

5.1.2.2 Wind driven rain WDR

Wind-driven rain (WDR) is the main source of moisture present on the vertical components of structures. It describes the amount of rain that hits vertical building components, and its calculation in COMLEAM involves considering precipitation, wind speed, and wind direction, following the guidelines outlined in the ISO-15927 standard part 3 entitled "Hygrothermal performance of buildings: calculation and presentation of climatic data, Part 3: Calculation of a driving rain index for vertical surfaces.".

The ISO-15927 standard is one of the most frequently used models for wind-driven rain calculation. It is a semi-empirical model, which is a mathematical model that combines theoretical principles with empirical data based on measurements to describe or predict a phenomenon. The standard does not explicitly mention Eq. 1, called the WDR relationship; however, the combination of equations mentioned in the standard leads to it. Based on the WDR relationship, the WDR is calculated with the location factor α, the amount of precipitation r, the wind speed w, and the angle γ between the building component and wind direction (Figure 9)using the following formula:

$$r_{SR} = \alpha . r^{0.88} . w . \cos(\gamma) \quad \textbf{Eq.1}$$

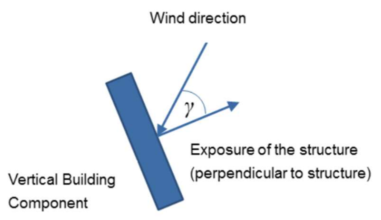


Figure 9. Angle γ in the wind driven rain (WDR) formula

Where

r : amount of precipitation (L/m²)

w : wind speed (m/s)

γ : angle between the component and the wind direction

α (Location factor) $= \frac{2}{9} C_R C_T O W$, where $C_R, C_T, O, and\ W$ are correction factors that corresponds to:

- The roughness coefficient C_R considers the variation in mean wind speed at the site owing to upstream roughness of the terrain and elevation above ground. It is calculated using the following formulas:

$$C_r(z) = K_r \times \ln\left(\frac{z}{z_0}\right) \mid z \geq z_{min} \quad \text{or} \quad C_r(z) = K_r \times \ln\left(\frac{z_{min}}{z_0}\right) \mid z < z_{min}$$

With

K_r: terrain factor (-)

z: Component height above the ground (m)

z_0: aerodynamic roughness length (m)

z_{min}: minimum height (m)

Except for z, all the other parameters (K_r, z_0, z_{min}) are provided by the ISO standard, see Table 8.

Table 8. Terrain categories and related parameters – ISO standard

Terrain category	Description	K_R	z_0	z_{min}
I	Rough open sea; lake shore with at least 5 km open water upwind and smooth flat country without obstacles	0,17	0,01	2
II	Farm land with boundary hedges, occasional small farm structures, houses or trees	0,19	0,05	4
III	Suburban or industrial areas and permanent forests	0,22	0,3	8
IV	Urban areas in which at least 15 % of the surface is covered with buildings of average height exceeding 15 m	0,24	1	16

- The C_T coefficient refers to the terrain topography coefficient, which reflects the influence of the location topography at the site on the calculation of WDR (Burkhardt et al., 2020). It considers the rise in average wind speed around isolated hills and escarpments. It comes into play when the wind approaches the slope of the hill or escarpment and when the building is positioned either "more than halfway up the slope of a hill" or "within 1.5 times the height of the cliff from the base of a cliff." The C_T value varies from 1.0 for gently sloping upstream slopes (less than 5% inclination) to a maximum of 1.6 for buildings situated at the crest of steep cliffs or escarpments (Blocken and Carmeliet, 2010).
- The obstruction factor O reflects the influence of the presence of an obstacle on the amount of rain hitting the surface. Table 9 gives the values of O as a function of the distance of the obstacle from the vertical component.

Table 9. Obstruction factor - ISO Standard

Distance of obstruction from wall m	Obstruction factor *O*
from 4 to 8	0,2
over 8 to 15	0,3
over 15 to 25	0,4
over 25 to 40	0,5
over 40 to 60	0,6
over 60 to 80	0,7
over 80 to 100	0,8
over 100 to 120	0,9
over 120	1,0

- The wall factor W is, by definition, "ratio of the quantity of water hitting a wall to the quantity passing through an equivalent unobstructed space''. This factor considers the type of the vertical component (the wall) in terms of height/ roof overhang. The standard provides different wall factors for six different wall configurations (Table 10).

Table 10. Wall factors as per the ISO standard (Blocken and Carmeliet, 2010)

Description of wall	Average value	Distribution
Two-storey gable	0,4	0,5 0,4 0,3 0,3 0,3 0,2
Three-storey gable	0,3	0,5 0,4 0,3 0,3 0,3 0,3 0,3 0,3 0,2
Multi-storey building with flat roof (pitch < 20°)	0,2 for a ten-storey building, for example, but with a higher intensity at top	0,5 for top 2,5 m 0,2 for remainder
Two-storey wall with eaves	0,3	Pitched roof (≥20°) typical overhang: 350 mm 0,3 0,3 0,3
Three-storey wall with eaves	0,4	Pitched roof typical overhang: 350 mm 0,4 0,4 0,4
Two-storey building with flat roof (pitch < 20°)	0,4	0,5 0,4 0,2

5.1.2.2.1 ISO-15927-3 Limitations

Referring to (Blocken and Carmeliet, 2010), the ISO standard includes some warnings related to the reliability and applicability of the standard, which includes:

1. The standard cannot be used for mountainous regions featuring steep cliffs or deep gorges.
2. The standard is particularly applied to climates similar to the UK climate; however, the standard does not specify any criteria to tell whether an area has the same or a different climate as the UK.
3. The standard cannot be used for regions in which snow or hail are the governing time of precipitation.
4. The standard mentions the need to assess the representativeness of calculated values from a meteorological station to a building located at a distant site.
5. The standard is not suitable for areas where severe convective storms, characterized by brief episodes of heavy precipitation such as showers or thunderstorms lasting less than 1 hour, account for more than 25% of the annual rainfall.
6. Rain penetration into cracks, as well as windows and doors edges, is dependent on brief heavy rainy events along with strong wind.

Thus, to achieve a better and more reliable calculation of WDR reaching a component, it is crucial to take into account these limitations while employing ISO 15927-3 and enhance it with additional site-specific data and professional opinion.

5.2 TEB model

The SURFEX modeling platform, an externalized code that represents surface processes created by Météo-France, includes the TEB (Town Energy Balance) model (Masson, 2000). TEB is a surface scheme that models the exchange of energy, radiation, and water between urban surfaces, soils, and the atmosphere using the soil-vegetation-atmosphere transfer scheme: ISBA (Lemonsu et al., 2012; Masson, 2000). This model can be run either coupled to other meteorological models or in off-line mode, forced by observed atmospheric data. It is based on a regular mesh with a resolution ranging from several tens to several hundreds of meters. Each grid cell in the model represents a city street in the area, characterized by its width, building height, construction materials, roof and facade color and insulation, and the proportion of windows, etc. (CNRS, 2022). In TEB, the urban environment is represented by three compartments: buildings (roofs and walls), roads (streets, sidewalks, and parking lots), and gardens (permeable surfaces of bare or vegetated soil) (Figure 10).

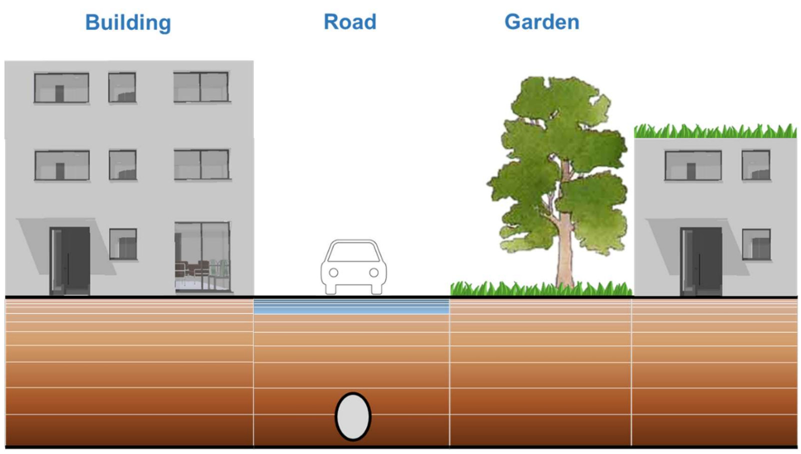


Figure 10. Schematic representation of the canyon street with the three compartments "building, road and garden"

Initially, TEB was exclusively dedicated to artificial surfaces, based on the concept of a canyon, developed by (Oke, 1987), where a town is represented by a roof, a road, and two opposite walls (Lemonsu, 2003; Masson, 2000; Masson et al., 2002). New versions were developed to integrate the vegetation into the urban scheme. The TEB-Veg model incorporates various aspects of vegetation, including vegetation within the street and urban green spaces (Lemonsu et al., 2012) using the ISBA model (Boone, 2000), green roofs in urban design (De Munch et al., 2013), and tall vegetation within the street (Redon et al., 2017). The urban geometry is simplified in the model, focusing on processes at a neighborhood scale rather than directly simulating all the features of individual buildings or streets. These simplifications result in the description of a town in the form of an idealized infinite street made up of a road and vegetation surrounded on either side by buildings of a defined height. The improved version of TEB-Veg, TEB-Hydro, was introduced to provide an extensive picture of the hydrological activities occurring in the urban subsoil (Bernard et al., 2021; Stavropulos-Laffaille et al., 2021, 2018). The various hydrological and energy processes involved in this model are well described by Stavropulos-Laffaille et al. (2018) and Stavropulos-Laffaille et al. (2021).

6 Conclusion

Biocides are essential in the building envelope, including roofs, facades, foundations, terraces, and surrounding areas, to inhibit the development of fungi, algae, and other damaging microorganisms. Various types of roofing materials, including polymeric, mineral, and wooden, utilize biocides to enhance the livability of structures. Film-preserving biocides are crucial for façade preservation, especially when they have thermal insulation systems, as they create an environment conducive to their proliferation. Termites, a species of insect that consumes timber, can invade construction materials and cause damage to building components due to their high moisture requirements. Biocides are subject to diverse regulatory frameworks. The BPR, which is controlled by the ECHA, governs biocides in the European Union. The multifarious applications of biocidal products are underscored by their classification into distinct categories, including disinfectants, preservatives, arthropod control, and antifouling agents.

Leaching from construction materials is a significant source of biocide contamination in urban waters, as rain transports biocides into soil, surface rivers, and streams. Stormwater pollution is particularly concerning because it often collects in sewage systems, is released into surface water bodies, or infiltrates into groundwater. Various environmental compartments, including urban sewage systems, stormwater runoff, soil, surface water, and urban wastewater treatment plant effluents, contain detected biocides. The leaching process involves rainwater diffusion, desorption of biocidal molecules, concentration gradients, hydrolysis or photolysis, and transportation by surface water, which is washed away by runoff. The release of biocides from construction materials is controlled by chemical and physical processes, with biocides exhibiting first flush behavior and desorption. Various factors, including interactions between the matrix and the chemical properties of the active substances, pH, structure properties, permeability, porosity, and exposure orientation, influence the leaching of biocides. Different biocides can have big differences in their water solubility and partitioning coefficients. Emission rates and total emissions go up as water solubility goes up and down as partitioning coefficients go down. The structural similarity between active substances and materials, including the existence of functional groups, exerts an influence on their interactions. Monolithic materials exhibit diffusion-controlled release mechanisms, while granular materials rely on percolation. Permeability and porosity also play a role in the release process. Exposure orientation significantly influences the amount of biocides leached into runoff. Environmental factors like rain intensity, total precipitation, wind direction, temperature, and UV irradiation also influence the leaching process. The amount of water in contact with exposed surfaces is a critical factor, with the emission of biocides mainly related to the amount of water reaching the surface. UV irradiation can cause the photodegradation of active substances in biocides.

Adding the COMLEAM model to the TEB model helps us understand the complicated interactions between biocides and building materials, including how they leak into the environment. COMLEAM uses measured data to implement emission data, which describes substance emission as a function of water accumulated. These functions map the relationship between a component's outflow and resulting emissions. There are six different kinds of emission functions in COMLEAM. These are logarithmic, Michaelis-Menten kinetics, Langmuir desorption, limited growth, and diffusion-controlled functions. COMLEAM uses the hydrologic model to analyze and forecast water dynamics from precipitation to the environment that receives the water as it traverses the building envelope. The runoff calculation is based on the amount of water that flows over components as a result of rain reaching horizontal or vertical components. Wind-driven rain (WDR) is the main source of moisture on vertical components of structures, and its calculation in COMLEAM considers precipitation, wind speed, and wind direction based on the ISO-15927 standard. As the research goes on, looking into biocide modeling in water discharge from building facades and roofs could help us learn more about how these substances are spread out and build up inside buildings for the future. The evaluation and comparison of various emission functions within the COMLEAM model underscore the significance of opting for suitable models in order to generate precise predictions. The TEB model, integrated into SURFEX, simulates energy, radiation, and water exchange between urban surfaces, soils, and the atmosphere. Originally designed for artificial surfaces, it represents towns as canyons, evolving to include vegetation by coupling with the ISBA model. The model simplifies urban geometry to focus on neighborhood-scale processes, utilizing a regular mesh with resolutions ranging from tens to hundreds of meters. Each grid cell represents a city street with specified characteristics. This approach strikes a balance between computational efficiency and accuracy, making TEB suitable for studying urban energy and water dynamics.

7 Future work

Having acquired a comprehensive understanding of biocides, including their properties, presence on building exteriors, and leaching processes, along with the two tools that will be used (COMLEAM, TEB), we will now proceed to the next steps of representing the urban area in all its aspects, covering the various building characteristics (age, usage type, construction material, orientation) and the diverse biocidal content on building components, as well as the frequency of renovations.

First, in order to move forward in the modeling process, it is crucial to choose exemplary compounds that will represent other biocides present in the building envelope. The choice of these exemplary substances will be made following a meticulous review based on several factors: choice of biocidal usage, Claudia Paijens thesis findings, market study, and availability of leaching data. The chosen methodology will be based on the dynamics at a mesh level, with the aim of determining the appropriate means to characterize this spatial unit. It will rely upon two distinct categories of land usage, specifically single-family housing and multi-family housing. Various techniques for expressing the mesh are examined, and the study proceeds by assessing the mean values at this scale. The examination of mesh sensitivity is conducted to comprehend its influence on the outcomes. Following the analysis of this step, the focus shifts to stabilization, with the goal of identifying the number of meshes at which the data reaches a consistent overall average. By employing a systematic methodology, it becomes feasible to investigate and comprehend fluctuations and patterns at many levels.

For now, we will start by trying to assign an initial façade age which is a crucial parameter in determining the amount of biocidal substances present on the facades in the initial condition. Obtaining complete data on the historical distribution of building façade coating ages may be difficult. Therefore, our initial approach is to treat the area building by building, specify a period that takes into account the longest possible renovation period (the oldest plaster, render, or paint we can get), and then specify a renovation period for each building based on probabilistic distribution laws depending on building type, location, type of use, etc. After that, we are planning to study the sensitivity to spatial scale and the sensitivity to simplification of description by testing the number of buildings required for statistical effects to be smoothed out for the same land use and the level of modeling details required.

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Commenté [AB11]: Deux fois la même référence Paijens 2019. Tu ferais mieux de citer ses articles en anglais.

Commenté [RS12R11]:

9 Appendices

a. Biocidal active substances used in PT02 (Disinfectants and algaecides)

Substance name	CAS no.	Approval start date	Approval end date	Approval/Assessment status
Dialuminium chloride pentahydroxide	12042-91-0			Commission decision (participant withdrawal)
Reaction mass of titanium dioxide and silver chloride	-			Commission decision (participant withdrawal)
1,2-benzisothiazol-3(2H)-one (BIT)	2634-33-5			Initial application for approval in progress
2,2-dibromo-2-cyanoacetamide (DBNPA)	10222-01-2			Initial application for approval in progress

2-Phenoxyethanol	122-99-6			Initial application for approval in progress
6-(phthalimido)peroxyhexanoic acid (PAP)	128275-31-0			Initial application for approval in progress
Active bromine generated from sodium bromide and calcium hypochlorite	-			Initial application for approval in progress
Active bromine generated from sodium bromide and chlorine	-			Initial application for approval in progress
Active bromine generated from sodium bromide and sodium hypochlorite	-			Initial application for approval in progress
Active bromine generated from sodium bromide by electrolysis	-			Initial application for approval in progress
Active chlorine generated from chloride of ambient water by electrolysis	-			Initial application for approval in progress
active chlorine generated from magnesium chloride hexahydrate and potassium chloride by electrolysis	-			Initial application for approval in progress
Active chlorine generated from sodium chloride and pentapotassium bis(peroxymonosulphate) bis(sulphate)	-			Initial application for approval in progress
Alkyl (C12-16) dimethylbenzyl ammonium chloride (ADBAC/BKC (C12-16))	68424-85-1			Initial application for approval in progress
Alkyl (C12-18) dimethylbenzyl ammonium chloride (ADBAC (C12-18))	68391-01-5			Initial application for approval in progress
Alkyl (C12-C14) dimethyl(ethylbenzyl)ammonium chloride (ADEBAC (C12-C14))	85409-23-0			Initial application for approval in progress
Alkyl (C12-C14) dimethylbenzylammonium chloride (ADBAC (C12-C14))	85409-22-9			Initial application for approval in progress
Bromochloro-5,5-dimethylimidazolidine-2,4-dione (BCDMH/Bromochlorodimethylhydantoin)	32718-18-6			Initial application for approval in progress
Bronopol	52-51-7			Initial application for approval in progress
chlorine dioxide	10049-04-4			Initial application for approval in progress
Chlorine dioxide generated from sodium chlorate and hydrogen peroxide in the presence of a strong acid	-			Initial application for approval in progress
Chlorine dioxide generated from sodium chlorite by acidification	-			Initial application for approval in progress
Chlorine dioxide generated from sodium chlorite by electrolysis	-			Initial application for approval in progress
Chlorine dioxide generated from sodium chlorite by oxidation	-			Initial application for approval in progress
Chlorine dioxide generated from Tetrachlorodecaoxide complex (TCDO) by acidification	-			Initial application for approval in progress
Copper	7440-50-8			Initial application for approval in progress

D-gluconic acid, compound with N,N″-bis(4-chlorophenyl)-3,12-diimino-2,4,11,13-tetraazatetradecanediamidine(2:1) (CHDG)	18472-51-0			Initial application for approval in progress
Didecyldimethylammonium chloride (DDAC (C8-10))	68424-95-3			Initial application for approval in progress
Dimethyloctadecyl[3-(trimethoxysilyl)propyl]ammonium chloride	27668-52-6			Initial application for approval in progress
Ethanol	64-17-5			Initial application for approval in progress
Ethylene oxide	75-21-8			Initial application for approval in progress
Formic acid	64-18-6			Initial application for approval in progress
Free radicals generated in situ from ambient air or water	-			Initial application for approval in progress
Glycolic acid	79-14-1			Initial application for approval in progress
Glyoxal	107-22-2			Initial application for approval in progress
hydrogen peroxide released from sodium percarbonate	-			Initial application for approval in progress
Magnesium monoperoxyphthalate hexahydrate (MMPP)	84665-66-7			Initial application for approval in progress
Monolinuron	1746-81-2			Initial application for approval in progress
N-(3-aminopropyl)-N-dodecylpropane-1,3-diamine (Diamine)	2372-82-9			Initial application for approval in progress
Ozone generated from oxygen	-			Initial application for approval in progress
Pentapotassium bis(peroxymonosulphate) bis(sulphate)	70693-62-8			Initial application for approval in progress
Peracetic acid generated from 1,3-diacetyloxypropan-2-yl acetate and hydrogen peroxide	-			Initial application for approval in progress
Peracetic acid generated from tetraacetylethylenediamine and hydrogen peroxide	-			Initial application for approval in progress
Performic acid generated from formic acid and hydrogen peroxide	-			Initial application for approval in progress
Poly(oxy-1,2-ethanediyl), α-[2-(didecylmethylammonio)ethyl]- .omega.- hydroxy-, propanoate (salt) (Bardap 26)	94667-33-1			Initial application for approval in progress
Polymer of N-Methylmethanamine (EINECS 204-697-4 with (chloromethyl) oxirane (EINECS 203-439-8)/Polymeric quaternary ammonium chloride (PQ Polymer)	25988-97-0			Initial application for approval in progress
Pyridine-2-thiol 1-oxide, sodium salt (Sodium pyrithione)	3811-73-2			Initial application for approval in progress
Pyrithione zinc (Zinc pyrithione)	13463-41-7			Initial application for approval in progress

Quaternary ammonium compounds, benzyl-C12-18-alkyldimethyl, salts with 1,2-benzisothiazol-3(2H)-one 1,1-dioxide	68989-01-5			Initial application for approval in progress
reaction mass of N,N-didecyl-N-(2-hydroxyethyl)-N-methylammonium propionate and N,N-didecyl-N-(2-(2-hydroxyethoxy)ethyl)-N-methylammonium propionate and N,N-didecyl-N-(2-(2-(2-hydroxyethoxy)ethoxy)ethyl)-N-methylammonium propionate	-			Initial application for approval in progress
Reaction products of aluminium trihydroxide and hydrochloric acid and aluminium and water	-			Initial application for approval in progress
Reaction products of paraformaldehyde and 2-hydroxypropylamine (ratio 1:1)	25254-50-6			Initial application for approval in progress
Reaction products of paraformaldehyde and 2-hydroxypropylamine (ratio 3:2)	-			Initial application for approval in progress
Reaction products of: glutamic acid and N-(C12-C14-alkyl)propylenediamine (Glucoprotamin)	164907-72-6			Initial application for approval in progress
Salicylic acid	69-72-7			Initial application for approval in progress
Silver	7440-22-4			Initial application for approval in progress
Silver borophosphate glass	-			Initial application for approval in progress
Silver chloride	7783-90-6			Initial application for approval in progress
Silver nitrate	7761-88-8			Initial application for approval in progress
Silver phosphate glass	308069-39-8			Initial application for approval in progress
silver phosphoborate glass	-			Initial application for approval in progress
Silver zinc zeolite	130328-20-0			Initial application for approval in progress
Sodium dichloroisocyanurate dihydrate	51580-86-0			Initial application for approval in progress
Symclosene	87-90-1			Initial application for approval in progress
Tosylchloramide sodium (Tosylchloramide sodium - Chloramin T)	127-65-1			Initial application for approval in progress
Troclosene sodium	2893-78-9			Initial application for approval in progress
5-chloro-2-(4-chlorphenoxy)phenol (DCPP)	3380-30-1	01/12/2016	30/11/2026	Approved
Active chlorine generated from sodium chloride by electrolysis	-	01/07/2022	30/06/2032	Approved
Active chlorine released from calcium hypochlorite	7778-54-3	01/01/2019	31/12/2028	Approved
Active chlorine released from chlorine	7782-50-5	01/01/2019	31/12/2028	Approved

Active chlorine released from hypochlorous acid	-	01/07/2022	30/06/2032	Approved
Active chlorine released from sodium hypochlorite	7681-52-9	01/01/2019	31/12/2028	Approved
Amines, N-C10-16-alkyltrimethylenedi-, reaction products with chloroacetic acid	139734-65-9	01/01/2018	31/12/2027	Approved
Biphenyl-2-ol	90-43-7	01/07/2017	30/06/2027	Approved
Calcium dihydroxide/calcium hydroxide/caustic lime/hydrated lime/slaked lime	1305-62-0	01/05/2018	30/04/2028	Approved
Calcium magnesium oxide/dolomitic lime	37247-91-9	01/05/2018	30/04/2028	Approved
Calcium magnesium tetrahydroxide/calcium magnesium hydroxide/hydrated dolomitic lime	39445-23-3	01/05/2018	30/04/2028	Approved
Calcium oxide/lime/burnt lime/quicklime	1305-78-8	01/05/2018	30/04/2028	Approved
Chlorocresol	59-50-7	01/05/2018	30/04/2028	Approved
Citric acid	77-92-9	01/03/2018	28/02/2028	Approved
Copper sulphate pentahydrate	7758-99-8	01/07/2015	30/06/2025	Approved
Didecyldimethylammonium chloride(DDAC)	7173-51-5	01/02/2024	31/01/2034	Approved
Formaldehyde	50-00-0	01/02/2022	31/01/2025	Approved
Glutaral (Glutaraldehyde)	111-30-8	01/10/2016	30/09/2026	Approved
Hydrogen peroxide	7722-84-1	01/02/2017	31/01/2027	Approved
L-(+)-lactic acid	79-33-4	01/05/2019	30/04/2029	Approved
Mixture of 5-chloro-2-methyl-2H- isothiazol-3-one (EINECS 247-500-7) and 2-methyl-2H-isothiazol-3-one (EINECS 220-239-6) (Mixture of CMIT/MIT)	55965-84-9	01/07/2017	30/06/2027	Approved
Nonanoic acid, Pelargonic acid	112-05-0	01/10/2015	30/09/2025	Approved
Peracetic acid	79-21-0	01/10/2017	30/09/2027	Approved
Peracetic acid generated from tetra-acetylethylenediamine (TAED) and sodium percarbonate	-	01/01/2019	31/12/2028	Approved
polyhexamethylene biguanide hydrochloride with a mean number-average molecular weight (Mn) of 1415 and a mean polydispersity (PDI) of 4.7 (PHMB(1415;4.7))	1802181-67-4	01/11/2019	31/10/2026	Approved
polyhexamethylene biguanide hydrochloride with a mean number-average molecular weight (Mn) of 1600 and a mean polydispersity (PDI) of 1.8 (PHMB(1600;1.8))	27083-27-8	01/07/2017	30/06/2024	Approved
Propan-1-ol	71-23-8	01/05/2019	30/04/2029	Approved
Propan-2-ol	67-63-0	01/07/2016	30/06/2026	Approved
Reaction mass of peracetic acid and peroxyoctanoic acid	-	01/04/2022	31/03/2032	Approved
Hydrochloric acid	-	01/05/2014	30/04/2024	Approved Renewal in progress
Cinnamaldehyde/3-phenyl-propen-2-al(Cinnamic aldehyde)	104-55-2			Cancelled application

(ethylenedioxy)dimethanol (Reaction products of ethylene glycol with paraformaldehyde (EGForm))	3586-55-8			No longer supported
active bromine generated from ozone and bromide of natural water and sodium bromide	-			No longer supported
Active chlorine generated from hydrochloric acid by electrolysis	-			No longer supported
active chlorine generated from magnesium chloride hexahydrate by electrolysis	-			No longer supported
Active chlorine generated from potassium chloride by electrolysis	-			No longer supported
active chlorine generated from sodium chloride and pentapotassium bis(peroxymonosulphate) bis(sulphate) and sulphamic acid	-			No longer supported
Active Chlorine: manufactured by the reaction of hypochlorous acid and sodium hypochlorite produced in situ	-			No longer supported
Cetylpyridinium chloride	123-03-5			No longer supported
Chloramin B	127-52-6			No longer supported
Chlorine dioxide	10049-04-4			No longer supported
Chlorine dioxide generated from sodium chloride by electrolysis	-			No longer supported
Chlorine dioxide generated from sodium chlorite and sodium persulfate	-			No longer supported
Peracetic acid generated by perhydrolysis of N-acetylcaprolactam by hydrogen peroxide in alkaline conditions	-			No longer supported
Peroxyoctanoic acid	33734-57-5			No longer supported
Silver	7440-22-4			No longer supported
Sodium 2-biphenylate	132-27-4			No longer supported
Sodium bromide	7647-15-6			No longer supported
Sodium p-chloro-m-cresolate	15733-22-9			No longer supported
Tetrachlorodecaoxide complex (TCDO)	92047-76-2			No longer supported
Tetrahydro-1,3,4,6-tetrakis(hydroxymethyl)imidazo[4,5-d]imidazole-2,5 (1H,3H)-dione (TMAD)	5395-50-6			No longer supported
Tetrakis(hydroxymethyl)phosphonium sulphate (2:1) (THPS)	55566-30-8			No longer supported
2-Butanone, peroxide	1338-23-4			Not approved
2-Propenoic acid, 2-methyl-, butyl ester, polymer with butyl 2-propenoate and methyl 2-methyl-2-propenoate (CAS nr: 25322-99-0)/ Polymeric quaternary ammonium bromide (PQ Polymer)	-			Not approved
Clorophene (Chlorophene)	120-32-1			Not approved
Silver copper zeolite	130328-19-7			Not approved

Silver sodium hydrogen zirconium phosphate	265647-11-8			Not approved
Silver zeolite	130328-18-6			Not approved
Triclosan	3380-34-5			Not approved

b. Biocidal active substances used in PT06 (Preservatives for products during storage)

Substance name	CAS	Approval start date	Approval end date	Approval/Assessment status
7a-ethyldihydro-1H,3H,5H-oxazolo[3,4-c]oxazole (EDHO)	7747-35-5			Commission decision (participant withdrawal)
cis-1-(3-chloroallyl)-3,5,7-triaza-1-azoniaadamantane chloride (cis CTAC)	51229-78-8			Commission decision (participant withdrawal)
Methenamine 3-chloroallylochloride (CTAC)	4080-31-3			Commission decision (participant withdrawal)
Reaction mass of titanium dioxide and silver chloride	-			Commission decision (participant withdrawal)
Sodium N-(hydroxymethyl)glycinate	70161-44-3			Commission decision (participant withdrawal)
(benzyloxy)methanol	14548-60-8			Initial application for approval in progress
(ethylenedioxy)dimethanol (Reaction products of ethylene glycol with paraformaldehyde (EGForm))	3586-55-8			Initial application for approval in progress
1,2-benzisothiazol-3(2H)-one (BIT)	2634-33-5			Initial application for approval in progress
1,3-bis(hydroxymethyl)-5,5-dimethylimidazolidine-2,4-dione (DMDMH)	6440-58-0			Initial application for approval in progress
2,2-dibromo-2-cyanoacetamide (DBNPA)	10222-01-2			Initial application for approval in progress
2,2′,2″-(hexahydro-1,3,5-triazine-1,3,5-triyl)triethanol (HHT)	4719-04-4			Initial application for approval in progress
2,2′-dithiobis[N-methylbenzamide] (DTBMA)	2527-58-4			Initial application for approval in progress
2-butyl-benzo[d]isothiazol-3-one (BBIT)	4299-07-4			Initial application for approval in progress
2-methyl-2,3-dihydro-1,2-thiazol-3-one hydrochloride	26172-54-3			Initial application for approval in progress
2-methyl-2H-isothiazol-3-one (MIT)	2682-20-4			Initial application for approval in progress
2-octyl-2H-isothiazol-3-one (OIT)	26530-20-1			Initial application for approval in progress
5-Chloro-2-methyl-2H-isothiazol-3-one (CIT)	26172-55-4			Initial application for approval in progress
Benzyl Alcohol	100-51-6			Initial application for approval in progress
Bronopol	52-51-7			Initial application for approval in progress
Didecyldimethylammonium chloride (DDAC (C8-10))	68424-95-3			Initial application for approval in progress
Didecyldimethylammonium chloride(DDAC)	7173-51-5			Initial application for approval in progress
Dodecylguanidine monohydrochloride	13590-97-1			Initial application for approval in progress
Ethanol	64-17-5			Initial application for approval in progress
Formic acid	64-18-6			Initial application for approval in progress
Hexa-2,4-dienoic acid (Sorbic acid)	110-44-1			Initial application for approval in progress

Monochloramine generated from ammonium carbamate and a chlorine source	-			Initial application for approval in progress
N-(3-aminopropyl)-N-dodecylpropane-1,3-diamine (Diamine)	2372-82-9			Initial application for approval in progress
p-[(diiodomethyl)sulphonyl]toluene	20018-09-1			Initial application for approval in progress
Pyridine-2-thiol 1-oxide, sodium salt (Sodium pyrithione)	3811-73-2			Initial application for approval in progress
Pyrithione zinc (Zinc pyrithione)	13463-41-7			Initial application for approval in progress
Reaction products of paraformaldehyde and 2-hydroxypropylamine (ratio 1:1)	25254-50-6			Initial application for approval in progress
Reaction products of paraformaldehyde and 2-hydroxypropylamine (ratio 3:2)	-			Initial application for approval in progress
Silver chloride	7783-90-6			Initial application for approval in progress
sulfur dioxide released from sodium metabisulfite	7446-09-5			Initial application for approval in progress
Tetrahydro-1,3,4,6-tetrakis(hydroxymethyl)imidazo[4,5-d]imidazole-2,5 (1H,3H)-dione (TMAD)	5395-50-6			Initial application for approval in progress
Tetrakis(hydroxymethyl)phosphonium sulphate (2:1) (THPS)	55566-30-8			Initial application for approval in progress
2-bromo-2-(bromomethyl)pentanedinitrile (DBDCB)	35691-65-7	01/01/2018	31/12/2027	Approved
3-iodo-2-propynylbutylcarbamate (IPBC)	55406-53-6	01/07/2015	30/06/2025	Approved
Biphenyl-2-ol	90-43-7	01/07/2017	30/06/2027	Approved
Chlorocresol	59-50-7	01/05/2018	30/04/2028	Approved
Glutaral (Glutaraldehyde)	111-30-8	01/10/2016	30/09/2026	Approved
Hydrogen peroxide	7722-84-1	01/02/2017	31/01/2027	Approved
L-(+)-lactic acid	79-33-4	01/11/2023	31/10/2033	Approved
MBIT	2527-66-4	01/07/2018	30/06/2028	Approved
Mixture of 5-chloro-2-methyl-2H- isothiazol-3-one (EINECS 247-500-7) and 2-methyl-2H-isothiazol-3-one (EINECS 220-239-6) (Mixture of CMIT/MIT)	55965-84-9	01/07/2017	30/06/2027	Approved
N-(trichloromethylthio)phthalimide (Folpet)	133-07-3	01/01/2016	31/12/2025	Approved
Peracetic acid	79-21-0	01/10/2017	30/09/2027	Approved
2-Phenoxyethanol	122-99-6			Cancelled application
Peracetic acid generated from tetra-acetylethylenediamine (TAED) and sodium percarbonate	-			Cancelled application
Sodium Azide	26628-22-8			Cancelled application
N,N′-methylenebismorpholine (MBM)	5625-90-1	01/04/2017	31/03/2022	Expired
Performic acid generated from formic acid and hydrogen peroxide	-			No longer supported
Potassium 2-biphenylate	13707-65-8			No longer supported
Silver nitrate	7761-88-8			No longer supported
Sodium 2-biphenylate	132-27-4			No longer supported
Sodium p-chloro-m-cresolate	15733-22-9			No longer supported
Tetrahydro-3,5-dimethyl-1,3,5-thiadiazine-2-thione (Dazomet)	533-74-4			No longer supported
2-Butanone, peroxide	1338-23-4			Not approved
4,4-dimethyloxazolidine	51200-87-4			Not approved

polyhexamethylene biguanide hydrochloride with a mean number-average molecular weight (Mn) of 1415 and a mean polydispersity (PDI) of 4.7 (PHMB(1415;4.7))	1802181-67-4			Not approved
polyhexamethylene biguanide hydrochloride with a mean number-average molecular weight (Mn) of 1600 and a mean polydispersity (PDI) of 1.8 (PHMB(1600;1.8))	27083-27-8			Not approved

c. Biocidal active substances used in PT07 (Film preservatives) (ECHA, 2023c)

Substance name	CAS	Approval start date	Approval end date	Approval/Assessment status
Reaction mass of titanium dioxide and silver chloride	-			Commission decision (participant withdrawal)
2-butyl-benzo[d]isothiazol-3-one (BBIT)	4299-07-4			Initial application for approval in progress
2-octyl-2H-isothiazol-3-one (OIT)	26530-20-1			Initial application for approval in progress
2-thiazol-4-yl-1H-benzoimidazole (Thiabendazole)	148-79-8			Initial application for approval in progress
3-(4-isopropylphenyl)-1,1-dimethylurea/ Isoproturon	34123-59-6			Initial application for approval in progress
3-iodo-2-propynylbutylcarbamate (IPBC)	55406-53-6			Initial application for approval in progress
4,5-Dichloro-2-octylisothiazol-3(2H)-one (4,5-Dichloro-2-octyl-2H-isothiazol-3-one (DCOIT))	64359-81-5			Initial application for approval in progress
Benzoic acid	65-85-0			Initial application for approval in progress
Biphenyl-2-ol	90-43-7			Initial application for approval in progress
Dimethyloctadecyl[3-(trimethoxysilyl)propyl]ammonium chloride	27668-52-6			Initial application for approval in progress
Diuron	330-54-1			Initial application for approval in progress
Free radicals generated in situ from ambient air or water	-			Initial application for approval in progress
p-[(diiodomethyl)sulphonyl]toluene	20018-09-1			Initial application for approval in progress
Pyridine-2-thiol 1-oxide, sodium salt (Sodium pyrithione)	3811-73-2			Initial application for approval in progress
Pyrithione zinc (Zinc pyrithione)	13463-41-7			Initial application for approval in progress
Silver borophosphate glass	-			Initial application for approval in progress
Silver chloride	7783-90-6			Initial application for approval in progress
Silver phosphate glass	308069-39-8			Initial application for approval in progress
silver phosphoborate glass	-			Initial application for approval in progress
Silver zinc zeolite	130328-20-0			Initial application for approval in progress
Terbutryn	886-50-0			Initial application for approval in progress
1-[[2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]methyl]-1H-1,2,4-triazole (Propiconazole)	60207-90-1	01/12/2016	30/11/2026	Approved
Carbendazim	10605-21-7	01/02/2022	31/01/2025	Approved

Dichloro-N-[(dimethylamino)sulphonyl] fluoro-N-(ptolyl)methanesulphenamide (Tolylfluanid)	731-27-1	01/01/2018	31/12/2027	Approved
Fludioxonil	131341-86-1	01/04/2018	31/03/2028	Approved
N-(trichloromethylthio)phthalimide (Folpet)	133-07-3	01/10/2016	30/09/2026	Approved
tebuconazole	107534-96-3	01/07/2015	30/06/2025	Approved
Azoxystrobin	131860-33-8	01/11/2018	31/10/2025	Approved Other updates in progress
N-(Dichlorofluoromethylthio)-N′,N′-dimethyl-N-phenylsulfamide (Dichlofluanid)	1085-98-9			No longer supported
Silver nitrate	7761-88-8			No longer supported
Sodium 2-biphenylate	132-27-4			No longer supported
2-Propenoic acid, 2-methyl-, butyl ester, polymer with butyl 2-propenoate and methyl 2-methyl-2-propenoate (CAS nr: 25322-99-0)/ Polymeric quaternary ammonium bromide (PQ Polymer)	-			Not approved
Cu-HDO	312600-89-8			Not approved
Silver copper zeolite	130328-19-7			Not approved
Silver sodium hydrogen zirconium phosphate	265647-11-8			Not approved
Silver zeolite	130328-18-6			Not approved
Triclosan	3380-34-5			Not approved

d. Biocidal active substances used in PT08 (wood preservatives) (ECHA, 2023c)

Substance name	CAS	Approval start date	Approval end date	Approval-Assessment status
N-Didecyl-N-dipolyethoxyammonium borate/Didecylpolyoxethylammonium borate (Polymeric betaine)	214710-34-6			Initial application for approval in progress
(RS)-α-cyano-3phenoxybenzyl-(1RS)-cis, trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate (Cypermethrin)	52315-07-8	01/06/2015	31/05/2025	Approved
2-octyl-2H-isothiazol-3-one (OIT)	26530-20-1	01/01/2018	31/12/2027	Approved
4-bromo-2-(4-chlorophenyl)-1-ethoxy methyl-5-trifluoromethylpyrrole-3-carbonitrile (Chlorfenapyr)	122453-73-0	01/05/2015	30/04/2025	Approved
Alkyl (C12-16) dimethylbenzyl ammonium chloride (ADBAC/BKC (C12-16))	68424-85-1	01/02/2015	31/01/2025	Approved
Coco alkyltrimethylammonium chloride (ATMAC/TMAC)	61789-18-2	01/05/2018	30/04/2028	Approved
Cu-HDO	312600-89-8	01/09/2015	31/08/2025	Approved
Didecyldimethylammonium chloride(DDAC)	7173-51-5	01/02/2015	31/01/2025	Approved
Granulated copper	7440-50-8	01/01/2017	31/12/2026	Approved
hydrogen cyanide	74-90-8	01/10/2014	30/09/2024	Approved

Penflufen	494793-67-8	01/02/2019	31/01/2029	Approved
Permethrin	52645-53-1	01/05/2016	30/04/2026	Approved
Poly(oxy-1,2-ethanediyl), α-[2-(didecylmethylammonio)ethyl]- .omega.-hydroxy-, propanoate (salt) (Bardap 26)	94667-33-1	01/01/2018	31/12/2027	Approved
Potassium (E,E)-hexa-2,4-dienoate (Potassium Sorbate)	24634-61-5	01/12/2016	30/11/2026	Approved
Creosote	8001-58-9	01/05/2013	31/10/2029	Approved Other updates in progress
1-[[2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]methyl]-1H-1,2,4-triazole (Propiconazole)	60207-90-1	01/04/2010	31/12/2023	Approved Renewal in progress
3-iodo-2-propynylbutylcarbamate (IPBC)	55406-53-6	01/07/2010	31/07/2025	Approved Renewal in progress
4,5-Dichloro-2-octylisothiazol-3(2H)-one (4,5-Dichloro-2-octyl-2H-isothiazol-3-one (DCOIT))	64359-81-5	01/07/2013	30/06/2023	Approved Renewal in progress
Basic Copper carbonate	12069-69-1	01/02/2014	31/01/2024	Approved Renewal in progress
Boric acid	10043-35-3	01/09/2011	28/02/2024	Approved Renewal in progress
Copper (II) oxide	1317-38-0	01/02/2014	31/01/2024	Approved Renewal in progress
Copper hydroxide	20427-59-2	01/02/2014	31/01/2024	Approved Renewal in progress
DDACarbonate	894406-76-9	01/02/2013	31/07/2025	Approved Renewal in progress
Disodium tetraborate pentahydrate	12179-04-3	01/09/2011	28/02/2024	Approved Renewal in progress
etofenprox	80844-07-1	01/02/2010	31/10/2026	Approved Renewal in progress
K-HDO	66603-10-9	01/07/2010	31/12/2026	Approved Renewal in progress
sulfuryl fluoride	2699-79-8	01/01/2009	31/12/2023	Approved Renewal in progress
tebuconazole	107534-96-3	01/04/2010	30/06/2026	Approved Renewal in progress
Tetrahydro-3,5-dimethyl-1,3,5-thiadiazine-2-thione (Dazomet)	533-74-4	01/08/2012	31/01/2025	Approved Renewal in progress
Trichoderma harzianum strain T-720	67892-31-3			Cancelled application
(E)-1-(2-Chloro-1,3-thiazol-5-ylmethyl)-3-methyl-2-nitroguanidine (Clothianidin)	210880-92-5	01/02/2010	31/01/2020	Expired
2-thiazol-4-yl-1H-benzoimidazole (Thiabendazole)	148-79-8	01/07/2010	30/06/2020	Expired
Bifenthrin	82657-04-3	01/02/2013	31/01/2023	Expired

Boric oxide	1303-86-2	01/09/2011	31/08/2021	Expired
Cyproconazole	94361-06-5	01/11/2015	31/10/2020	Expired
Dichloro-N-[(dimethylamino)sulphonyl] fluoro-N-(ptolyl)methanesulphenamide (Tolylfluanid)	731-27-1	01/10/2011	30/09/2021	Expired
Disodium octaborate tetrahydrate	12280-03-4	01/09/2011	31/08/2021	Expired
Disodium tetraborate	1330-43-4	01/09/2011	31/08/2021	Expired
Disodium tetraborate decahydrate	1303-96-4	01/09/2011	31/08/2021	Expired
Fenoxycarb	72490-01-8	01/02/2013	31/01/2023	Expired
fenpropimorph	67564-91-4	01/07/2011	30/06/2021	Expired
flufenoxuron	101463-69-8	01/02/2014	31/01/2017	Expired
N-(Dichlorofluoromethylthio)-N′,N′-dimethyl-N-phenylsulfamide (Dichlofluanid)	1085-98-9	01/03/2009	28/02/2019	Expired
Thiacloprid	111988-49-9	01/01/2010	31/12/2019	Expired
thiamethoxam	153719-23-4	01/07/2010	30/06/2020	Expired
N-(3-aminopropyl)-N-dodecylpropane-1,3-diamine (Diamine)	2372-82-9			Not approved

e. Biocidal active substances used in PT09 (Fibre, rubber and polymerised materials preservatives)

Substance name	CAS no.	Approval start date	Approval end date	Approval/Assessment status
(benzothiazol-2-ylthio)methyl thiocyanate (TCMTB)	21564-17-0			Initial application for approval in progress
1,2-benzisothiazol-3(2H)-one (BIT)	2634-33-5			Initial application for approval in progress
1-[[2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl]methyl]-1H-1,2,4-triazole (Propiconazole)	60207-90-1	01/06/2015	31/05/2025	Approved
2-butyl-benzo[d]isothiazol-3-one (BBIT)	4299-07-4			Initial application for approval in progress
2-octyl-2H-isothiazol-3-one (OIT)	26530-20-1			Initial application for approval in progress
2-thiazol-4-yl-1H-benzoimidazole (Thiabendazole)	148-79-8			Initial application for approval in progress

3-iodo-2-propynylbutylcarbamate (IPBC)	55406-53-6			Initial application for approval in progress
4,5-Dichloro-2-octylisothiazol-3(2H)-one (4,5-Dichloro-2-octyl-2H-isothiazol-3-one (DCOIT))	64359-81-5			Initial application for approval in progress
Allyl isothiocyanate	57-06-7			Initial application for approval in progress
Azoxystrobin	131860-33-8	01/11/2018	31/10/2025	Approved Other updates in progress
Benzoic acid	65-85-0			Initial application for approval in progress
Biphenyl-2-ol	90-43-7			Initial application for approval in progress
Bronopol	52-51-7			No longer supported
Carbendazim	10605-21-7			Not approved
Chlorine dioxide generated from sodium chlorite by acidification	-			Initial application for approval in progress
Chlorocresol	59-50-7	01/05/2018	30/04/2028	Approved
Cu-HDO	312600-89-8			Not approved
Dimethyloctadecyl[3-(trimethoxysilyl)propyl]ammonium chloride	27668-52-6			Initial application for approval in progress
Dimethyltetradecyl[3-(trimethoxysilyl)propyl]ammonium chloride	41591-87-1			Initial application for approval in progress
Fludioxonil	131341-86-1	01/04/2018	31/03/2028	Approved
Free radicals generated in situ from ambient air or water	-			Initial application for approval in progress
Metam-sodium	137-42-8			No longer supported
N-(trichloromethylthio)phthalimide (Folpet)	133-07-3	01/10/2016	30/09/2026	Approved
p-[(diiodomethyl)sulphonyl]toluene	20018-09-1			Initial application for approval in progress
polyhexamethylene biguanide hydrochloride with a mean number-average molecular weight (Mn) of 1415 and a mean polydispersity (PDI) of 4.7 (PHMB(1415;4.7))	1802181-67-4			No longer supported
polyhexamethylene biguanide hydrochloride with a mean number-average molecular weight (Mn) of 1600 and a mean polydispersity (PDI) of 1.8 (PHMB(1600;1.8))	27083-27-8			Not approved
Potassium 2-biphenylate	13707-65-8			No longer supported
Potassium dimethyldithiocarbamate	128-03-0			Commission decision (participant withdrawal)
Pyridine-2-thiol 1-oxide, sodium salt (Sodium pyrithione)	3811-73-2			Initial application for approval in progress
Pyrithione zinc (Zinc pyrithione)	13463-41-7			Initial application for approval in progress

Reaction mass of chloromethyl hexyl cyanocarbonodithioimidate and bromomethyl hexyl cyanocarbonodithioimidate and dihexyl cyanocarbonodithioimidate	-			Cancelled application
Reaction mass of titanium dioxide and silver chloride	-			Commission decision (participant withdrawal)
Silver	7440-22-4			No longer supported
Silver	7440-22-4			No longer supported
Silver adsorbed on silicon dioxide	-			Initial application for approval in progress
Silver borophosphate glass	-			Initial application for approval in progress
Silver chloride	7783-90-6			Initial application for approval in progress
Silver copper zeolite	130328-19-7			Initial application for approval in progress
Silver nitrate	7761-88-8			Initial application for approval in progress
Silver phosphate glass	308069-39-8			Initial application for approval in progress
silver phosphoborate glass	-			Initial application for approval in progress
Silver sodium hydrogen zirconium phosphate	265647-11-8			Initial application for approval in progress
Silver zeolite	130328-18-6			Initial application for approval in progress
Silver zinc zeolite	130328-20-0			Initial application for approval in progress
Sodium 2-biphenylate	132-27-4			No longer supported
Sodium dimethyldithiocarbamate	128-04-1			Initial application for approval in progress
Sodium p-chloro-m-cresolate	15733-22-9			No longer supported
sulfur dioxide released from sodium metabisulfite	7446-09-5			Initial application for approval in progress
Terbutryn	886-50-0			Initial application for approval in progress
Thiram	137-26-8			No longer supported
Triclosan	3380-34-5			Not approved

f. Biocidal active substances used in PT10 (Construction material preservatives)

Substance name	CAS no.	Approval start date	Approval end date	Approval/Assessment status
1,2-benzisothiazol-3(2H)-one (BIT)	2634-33-5			No longer supported
2-butyl-benzo[d]isothiazol-3-one (BBIT)	4299-07-4			Initial application for approval in progress

2-octyl-2H-isothiazol-3-one (OIT)	26530-20-1			Initial application for approval in progress
2-thiazol-4-yl-1H-benzoimidazole (Thiabendazole)	148-79-8			Initial application for approval in progress
3-(4-isopropylphenyl)-1,1-dimethylurea/ Isoproturon	34123-59-6			Initial application for approval in progress
3-iodo-2-propynylbutylcarbamate (IPBC)	55406-53-6			Initial application for approval in progress
4,5-Dichloro-2-octylisothiazol-3(2H)-one (4,5-Dichloro-2-octyl-2H-isothiazol-3-one (DCOIT))	64359-81-5			Initial application for approval in progress
Alkyl (C12-16) dimethylbenzyl ammonium chloride (ADBAC/BKC (C12-16))	68424-85-1			Initial application for approval in progress
Alkyl (C12-18) dimethylbenzyl ammonium chloride (ADBAC (C12-18))	68391-01-5			Initial application for approval in progress
Alkyl (C12-C14) dimethyl(ethylbenzyl)ammonium chloride (ADEBAC (C12-C14))	85409-23-0			Initial application for approval in progress
Alkyl (C12-C14) dimethylbenzylammonium chloride (ADBAC (C12-C14))	85409-22-9			Initial application for approval in progress
Azoxystrobin	131860-33-8	01/11/2018	31/10/2025	Approved Other updates in progress
Biphenyl-2-ol	90-43-7			Initial application for approval in progress
Carbendazim	10605-21-7	01/02/2022	31/01/2025	Approved
Cu-HDO	312600-89-8			Not approved
Didecyldimethylammonium chloride (DDAC (C8-10))	68424-95-3			Initial application for approval in progress
Didecyldimethylammonium chloride(DDAC)	7173-51-5			Initial application for approval in progress
Diuron	330-54-1			Initial application for approval in progress
Fludioxonil	131341-86-1	01/04/2018	31/03/2028	Approved
Nonanoic acid, Pelargonic acid	112-05-0			Not approved
p-[(diiodomethyl)sulphonyl]toluene	20018-09-1			Initial application for approval in progress
Pine ext.	94266-48-5			Not approved
Poly(oxy-1,2-ethanediyl), α-[2-(didecylmethylammonio)ethyl]- .omega.-hydroxy-, propanoate (salt) (Bardap 26)	94667-33-1			Initial application for approval in progress
Potassium 2-biphenylate	13707-65-8			No longer supported
Pyridine-2-thiol 1-oxide, sodium salt (Sodium pyrithione)	3811-73-2			Initial application for approval in progress
Pyrithione zinc (Zinc pyrithione)	13463-41-7			Initial application for approval in progress
Pythium oligandrum, Chromista - Stramenopila	-	01/01/2016	31/12/2025	Approved
reaction mass of N,N-didecyl-N-(2-hydroxyethyl)-N-methylammonium propionate and N,N-didecyl-N-(2-(2-hydroxyethoxy)ethyl)-N-methylammonium propionate and N,N-didecyl-N-(2-(2-(2-hydroxyethoxy)ethoxy)ethyl)-N-methylammonium propionate	-			Initial application for approval in progress

Reaction mass of titanium dioxide and silver chloride	-			Commission decision (participant withdrawal)
Silver chloride	7783-90-6			No longer supported
Sodium 2-biphenylate	132-27-4			No longer supported
tebuconazole	107534-96-3	01/07/2015	30/06/2025	Approved
Terbutryn	886-50-0			Initial application for approval in progress

g. Biocidal active substances used in PT18 (Insecticide, Acaricides & other Biocidal Products against Arthropods)

Substance name	CAS	Approval start date	Approval end date	Approval-Assessment status
(1,3,4,5,6,7-hexahydro-1,3-dioxo-2H-isoindol-2-yl)methyl (1R-trans)-2,2-dimethyl-3-(2-methylprop-1-enyl)cyclopropanecarboxylate (d-Tetramethrin)	1166-46-7			Initial application for approval in progress
(RS)-3-Allyl-2-methyl-4-oxocyclopent-2- enyl-(1R,3R;1R,3S)-2,2-dimethyl-3-(2- methylprop-1-enyl)-cyclopropanecarboxy late (mixture of 4 isomers 1R trans, 1R:1R trans, 1S: 1R cis, 1R: 1R cis,1S 4:4:1:1) (d-Allethrin)	231937-89-6			Initial application for approval in progress
2-methyl-4-oxo-3-(prop-2-ynyl)cyclopent-2-en-1-yl 2,2-dimethyl-3-(2-methylprop-1-enyl)cyclopropanecarboxylate (Prallethrin)	23031-36-9			Initial application for approval in progress
4-bromo-2-(4-chlorophenyl)-1-ethoxy methyl-5-trifluoromethylpyrrole-3-carbonitrile (Chlorfenapyr)	122453-73-0			Initial application for approval in progress
Chrysanthemum cinerariaefolium extract from open and mature flowers of Tanacetum cinerariifolium obtained with supercritical carbondioxide	89997-63-7			Initial application for approval in progress
Chrysanthemum cinerariaefolium, extract from open and mature flowers of Tanacetum cinerariifolium obtained with hydrocarbon solvents	89997-63-7			Initial application for approval in progress
Cyanamide	420-04-2			Initial application for approval in progress
Geraniol	106-24-1			Initial application for approval in progress
Margosa extract from cold-pressed oil of the kernels of Azadirachta Indica extracted with super-critical carbon dioxide	84696-25-3			Initial application for approval in progress
Silicic acid, aluminium magnesium sodium salt	12040-43-6			Initial application for approval in progress
Sodium dimethylarsinate (Sodium Cacodylate)	124-65-2			Initial application for approval in progress
Tetramethrin	7696-12-0			Initial application for approval in progress
(E)-1-(2-Chloro-1,3-thiazol-5-ylmethyl)-3- methyl-2-nitroguanidine (Clothianidin)	210880-92-5	01/10/2016	30/09/2026	Approved
.alpha.-cyano-3-phenoxybenzyl2,2-dimethyl-3-(2-methylprop-1-enyl)cyclopropanecarboxylate (Cyphenothrin)	39515-40-7	01/02/2020	31/01/2030	Approved
.alpha.-cyano-4-fluoro-3-phenoxybenzyl3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate (Cyfluthrin)	68359-37-5	01/03/2018	28/02/2028	Approved
[1.alpha.(S*),3.alpha.]-(.alpha.)-cyano-(3-phenoxyphenyl)methyl3-(2,2-dichlor-oethenyl)-2,2-dichlorovinyl)-2,2-dimethyl-cyclopropanecarboxylate (alpha-Cypermethrin)	26046-85-5	01/07/2016	30/06/2026	Approved
[2,4-Dioxo-(2-propyn-1-yl)imidazolidin-3-yl]methyl(1R)-cis-chrysanthemate;[2,4- Dioxo-(2-propyn-1-yl)imidazolidin-3-yl] methyl(1R)-trans-chrysanthemate (Imiprothrin)	51-03-6	01/07/2019	30/06/2029	Approved

1R-trans phenothrin	67375-30-8	01/09/2015	31/08/2025	Approved
2-(2-butoxyethoxy)ethyl 6-propylpiperonyl ether (Piperonyl butoxide/PBO)	72963-72-5	01/07/2018	30/06/2028	Approved
Abamectin	71751-41-2	01/07/2013	30/06/2023	Approved
Acetamiprid	135410-20-7	01/02/2020	31/01/2027	Approved
Bacillus sphaericus 2362, strain ABTS-1743	143447-72-7	01/07/2016	30/06/2026	Approved
Bacillus thuringiensis subsp. israelensis, strain SA3A	-	01/07/2016	30/06/2026	Approved
Bacillus thuringiensis subsp. kurstaki, strain ABTS-351	-	01/03/2017	28/02/2027	Approved
Bendiocarb	22781-23-3	01/02/2014	31/01/2024	Approved
Decanoic acid	334-48-5	01/09/2015	31/08/2025	Approved
diflubenzuron	35367-38-5	01/02/2015	31/01/2025	Approved
epsilon-Momfluorothrin	1065124-65-3	01/07/2017	30/06/2027	Approved
etofenprox	80844-07-1	01/07/2015	30/06/2025	Approved
fipronil	120068-37-3	01/10/2013	30/09/2023	Approved
hydrogen cyanide	74-90-8	01/10/2014	30/09/2024	Approved
Kieselgur (diatomaceous earth)	61790-53-2	01/11/2018	31/10/2028	Approved
Margosa extract from the kernels of Azadirachta Indica extracted with water and further processed with organic solvents	84696-25-3	01/05/2014	30/04/2024	Approved
N-cyclopropyl-1,3,5-triazine-2,4,6-triamine (Cyromazine)	66215-27-8	01/01/2018	31/12/2027	Approved
Octanoic acid	124-07-2	01/09/2015	31/08/2025	Approved
Permethrin	52645-53-1	01/05/2016	30/04/2026	Approved
pyriproxyfen	95737-68-1	01/02/2015	31/01/2025	Approved
Pyrogenic, synthetic amorphous, nano, surface treated silicon dioxide	68909-20-6	01/11/2018	31/10/2028	Approved
S-Methoprene	65733-16-6	01/09/2015	31/08/2025	Approved
Synthetic amorphous silicon dioxide (nano)	112926-00-8	01/11/2015	31/10/2025	Approved
thiamethoxam	153719-23-4	01/02/2015	31/01/2025	Approved
Transfluthrin	118712-89-3	01/11/2015	31/10/2025	Approved
(RS)-α-cyano-3phenoxybenzyl-(1RS)-cis, trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate (Cypermethrin)	52315-07-8	01/06/2020	31/05/2030	Approved Other updates in progress
1-(3,5-dichloro-4-(1,1,2,2-tetrafluoroethoxy)phenyl)-3-(2,6-difluorobenzoyl) urea (Hexaflumuron)	86479-06-3	01/04/2017	30/09/2024	Approved Renewal in progress
Aluminium phosphide releasing phosphine	20859-73-8	01/02/2012	31/07/2024	Approved Renewal in progress
Bacillus thuringiensis subsp. israelensis Serotype H14, Strain AM65-52	-	01/10/2013	30/09/2023	Approved Renewal in progress
deltamethrin	52918-63-5	01/10/2013	30/09/2023	Approved Renewal in progress
Dinotefuran	165252-70-0	01/06/2015	30/11/2024	Approved Renewal in progress
imidacloprid	138261-41-3	01/07/2013	30/06/2023	Approved Renewal in progress
Indoxacarb (enantiomeric reaction mass S:R 75:25)	-	01/01/2010	30/06/2024	Approved Renewal in progress
lambda-cyhalothrin	91465-08-6	01/10/2013	30/09/2023	Approved Renewal in progress
Magnesium phosphide releasing phosphine	12057-74-8	01/02/2012	31/07/2024	Approved Renewal in progress
Metofluthrin	240494-71-7	01/05/2011	31/10/2023	Approved Renewal in progress
Spinosad	168316-95-8	01/11/2012	30/04/2025	Approved Renewal in progress
sulfuryl fluoride	2699-79-8	01/07/2011	31/12/2023	Approved Renewal in progress

Carbon dioxide	124-38-9	01/11/2012	31/10/2022	Expired
Nitrogen	7727-37-9	01/09/2011	31/08/2021	Expired
(RS)-3-Allyl-2-methyl-4-oxocyclopent-2- enyl (1R,3R)-2,2-dimethyl-3-(2-methyl prop-1-enyl)-cyclopropanecarboxylate (mixture of 2 isomers 1R trans: 1R/S only 1:3) (Esbiothrin)	260359-57-7			Not approved
1-ethynyl-2-methylpent-2-enyl 2,2-dimethyl-3-(2-methylprop-1-enyl)cyclopropanecarboxylate (Empenthrin)	54406-48-3			Not approved
2-chloro-N-[[[4-(trif luoromethoxy) phenyl]amino]carbonyl]benzamide (Triflumuron)	64628-44-0			Not approved
S-[(6-chloro-2-oxooxazolo[4,5-b]pyridin-3(2H)-yl)methyl] O,O-dimethylthiophosphate (Azamethiphos)	35575-96-3			Cancelled application
3-phenoxybenzyl (1R)-cis,trans-2,2- dimethyl-3-(2-methylprop-1-enyl)cyclopropanecarboxylate (d-Phenothrin)	188023-86-1			No longer supported
bacillus sphaericus	143447-72-7			No longer supported
bacillus thuringiensis subsp. israelensis, serotype H14	-			No longer supported
Chrysanthemum cinerariaefolium, ext.	89997-63-7			No longer supported
Esfenvalerate/(S)-.alpha.-Cyano-3-phenox ybenzyl (S)-2-(4-chlorophenyl)-3-methylbutyrate (Esfenvalerate)	66230-04-4			No longer supported
Pyrethrins and Pyrethroids	8003-34-7			No longer supported

h. Study characteristics & COMLEAM simulation settings

Table 11. Study characteristics (Tietje et al., 2018)

Site	Zurich, free Terbutryn	Ober-Ramstadt (RMI), free Terbutryn	Zurich, encapsulated Terbutryn
Material	plaster	plaster	plaster
Substance	terbutryn free	terbutryn free	terbutryn encapsulated
Tested component	field trial west-facing test façade with a size of 1.3 m² (height 1.8 m)	field trial: test body with a size of 0.8 m² (height 1.25 m, width 0.6 m)	field trial: total of 6 m² of façade area (3 m high)
Measurement period	369 days	308 days	615 days
C_0	2250 mg/m²	1036 mg/m²	1400 mg/m²
Precipitation amount	978 mm	607 mm	1756 mm
Water quantity	56 L/m²	58 L/m²	78 L/m²
Number of analysed samples	35	16	99
Terbutryn emission quantity	28 mg/m² (1.24 % of the initial amount)	20 mg/m² (1.93 % of the initial amount)	20 mg/m² (1.43 % of the initial amount)
Precipitation amount	978 mm	607 mm	1756 mm
Water quantity	56 L/m²	58 L/m²	78 L/m²
Number of analysed samples	35	16	99
Terbutryn emission quantity	28 mg/m² (1.24 % of the initial amount)	20 mg/m² (1.93 % of the initial amount)	20 mg/m² (1.43 % of the initial amount)

Table 12. COMLEAM simulation settings (Tietje et al., 2018)

Parameter	Zürich Terbutryn free	Zürich Terbutryn encapsulated	RMI Terbutryn free Weather
Geometry	Area: 1.3 m²	Area: 6 m²	Area: 0.8 m²
	Exposure: 270 °	Exposure: 275 °	Exposure: 180 °
	Ground angle: 90 °	Ground angle: 90 °	Ground angle: 90 °

Material with applied amount c_0	Plaster without color with c_0 = 2250 mg/m²	Plaster without color with c_0 = 1400 mg/m²	Plaster without color with c_0 = 1036 mg/m²
Runoff coefficient	1.0	1.0	1.0
Rain impact parameter	CR: 0.72	CR: 0.72	CR: 0.72
	CT: 1	CT: 1	CT: 1
	O: 0.6	O: 0.6	O: 1
	W: 0.55	W: 0.55	W: 0.5

i. Emission functions adjustment results

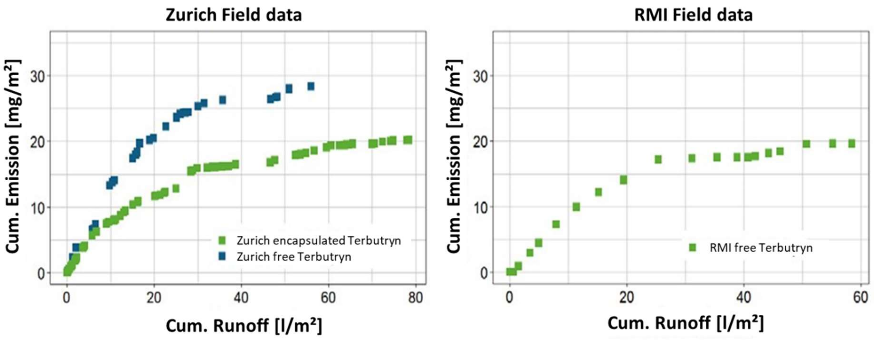

Figure 11. Results of the field study Zurich with terbutryn free (approx. 1 year) and terbutryn encapsulated (approx. 2 years) and Ober-Ramstadt with terbutryn free (approx. 1 year)

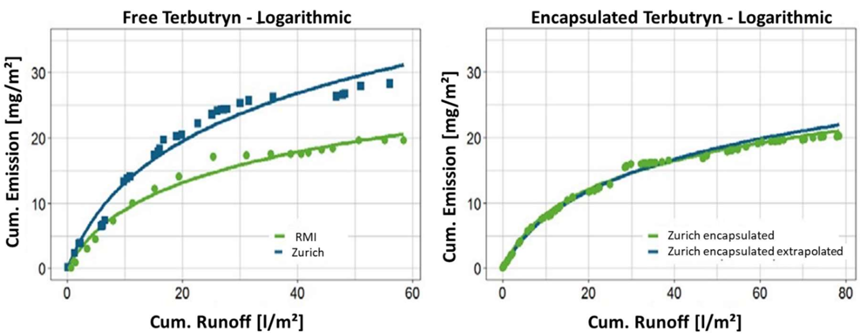

Figure 12. Curve fit of the logarithmic emission function at the sites Zurich (terbutryn free) and RMI (terbutryn free) (left), as well as Zurich (terbutryn encapsulated) when fitting to all data and when extrapolating the second half of the data (right; experimental period almost two years)

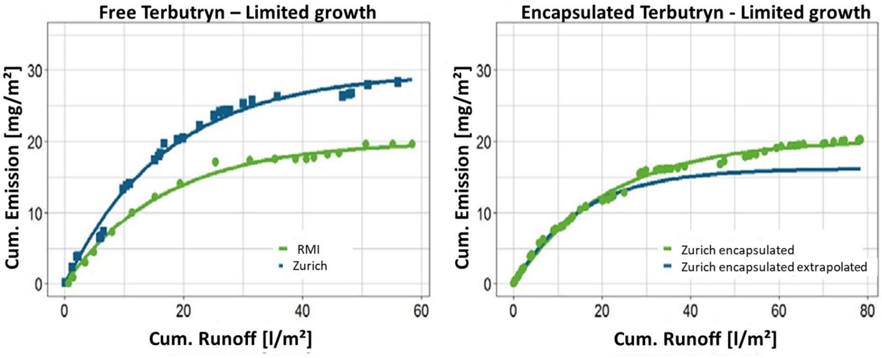

Figure 13. Curve fit of the emission function limited growth at the Zurich (terbutryn free) and RMI (terbutryn free) sites (left), and Zurich (terbutryn encapsulated) when fitted to all data and when extrapolating the second half of the data (right)

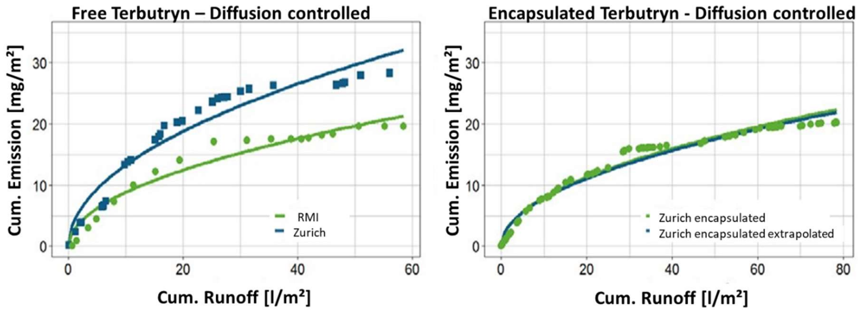

Figure 14. Curve fit of the diffusion-driven emission function at the Zurich (terbutyn free) and RMI (terbutryn free) sites (left), and Zurich (terbutryn encapsulated) when fitted to all data and when extrapolating the second half of the data (right)

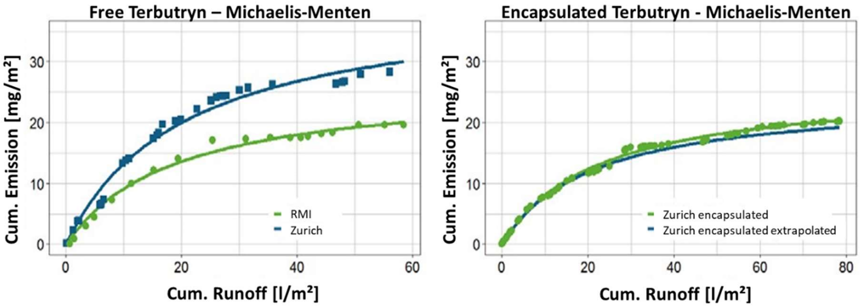

Figure 15. Curve fit of the Langmuir or Michaelis-Menten emission function at the Zurich (terbutryn free) and RMI (terbutryn free) sites (left), and Zurich (terbutryn encapsulated) when fitted to all data and when extrapolating the second half of the data (right).