

#### INERIS-DRC-02-25582-ECOT-VMi-n°02DR0270.doc

# Supplement to the methodology for risk evaluation of biocides

Emission scenario document for biocides used as masonry preservatives.

(product type 10)

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#### 1 INTRODUCTION

The purpose of this document is to provide a realistic scenario for the release into the environment of biocides used as masonry preservatives.

Mosses, lichen, and algae can disfigure surfaces of buildings (face, roof ...). They are a contributing factor to spalling and erosion of building materials. Like all plants, they hold and attract moisture, storing it within the surface on which they are growing, giving rise to problems of water penetration. They release various organic acids, which can degrade building materials.

Thus, masonry preservatives (product type 10) are products used for preservation and remedial treatment of masonry or construction materials (except wood preservatives, which are covered by product type 8) for the control of microbiological and algal attack (Danish Environmental Protection Agency, 2001).

Examples of products covered by this product type are products for the preservation (to protect or/and to cure) of mortar, concrete, concrete additives, baked clay, slate and other building materials (Bioexpo, 1998).

Four WIPO (World Intellectual Property Organization) treaties created classification systems which organise information concerning inventions, trademarks, and industrial designs into indexed, manageable structures for easy retrieval.

Among these classifications, there is the Nice classification, which concerns the International Classification of Goods and Services for the purposes of the registration of marks. The following goods are listed:

- ✓ Brickwork preservatives, except paints and oils
- ✓ Cement preservatives, except paints and oils
- ✓ Masonry preservatives, except paints and oils
- ✓ Preservatives for tile, except paints and oils

(WIPO, 2001)

There are biocides which are added at low concentration to additives for mortar or concrete. These biocides are used to protect additives themselves and not building materials. These biocides are covered by product type 6 "in-can preservatives" but their releases to the environment can also be performed according to the present scenario.

Moreover, biocides are added to paint, mortar or roughcast which are used on buildings. These biocides protect both product and building against mosses, lichen, algae... These biocides belong to product type 7 "film preservatives" as well. They will be taken into account in this document.

The treatment with biocides generally takes place in-situ. So, the releases can occur during the treatment or during the service life of building materials.

Note: Surface biocides are also used on slipways and causeways to remove/prevent the growth of algae. A specific scenario should be developed for this use. But as these biocides belong to both antifouling and masonry preservatives, this scenario does not elaborated in this document.

The scenarios in this report are presented in the following way:

#### Input

[Variable/parameter] [Symbol] [Unit] S/D/O/P

These parameters are the input to the scenario. The S, D, O or P classification of a parameter indicates the status:

- S Parameter must be present in the input data set for the calculation to be executed (there has been no method implemented in the system to estimate this parameter; no default value is set).
- D Parameter has a standard value (most defaults can be changed by the user)
- O Parameter is the output from another calculation (most output parameters can be overwritten by the user with alternative data).
- P Parameter value can be chosen from a "pick-list" of values.
- Default or output parameter is closed and cannot be changed by the user.

# **Output**

[Symbol] [Description]

#### **Intermediate calculations**

Parameter description (Unit)

[Parameter = equation] (Equation no.)

End calculations

[Parameter = equation] (Equation no.)

#### 2 TYPES OF BUILDING MATERIALS

In masonry, there are four basic materials: cement, clay, concrete and mortar. These materials are treated in the following sections.

#### 2.1 Cement

Cement is a finely ground, non-metallic, inorganic powder which when mixed with water forms a paste that sets and hardens. This hydraulic hardening is primarily due to the formation of calcium silicate hydrates as a result of the reaction between mixing water and the constituents of the cement. In the case of aluminous cements hydraulic hardening involves the formation of calcium aluminate hydrates.

Cement is a basic material for building and civil engineering construction. In Europe, the use of cement and concrete (a mixture of cement, aggregates, sand and water) in large civic works can be traced back to antiquity.

In 1995 world production of cement was up to 1 420 million tonnes. Table 1 shows distribution of cement production in the world (IPPC, 2000).

Table 1: World cement production by geographic regions in 1995 (IPPC, 2000)

Regions	Distribution of cement
	production (%)
China	30
Japan	7
Other Asia	23
EU	12
Other Europe	6
USA	5
Other America	8
Africa	4
Former USSR	4
Oceania	1

In 1995, EU cement production was 172 million tonnes and consumption 168 million tonnes. 23 million tonnes of cement were imported and 27 million tonnes exported.

The world's five largest cement producers are the four West European groups: Holderbank, Lafarge, Heidelberger and Italcementi, together with Cemex from Mexico (IPPC, 2000).

In 1995, there were 252 installations producing cement clinker and finished cement in the EU. In addition there are a further 68 grinding plants (mills) without kilns.

<u>Table 2</u>: Number of cement plants in EU countries 1995 (IPPC, 2000)

Country	Cement plants (with kilns)	Cement plants (cement mills only)
Austria	11	1
Belgium	5	3
Denmark	1	-
Finland	2	-
France	38	5
Germany	50	20
Greece	8	-
Ireland	2	-
Italy	64	29
Luxembourg	1	1
Netherlands	1	2
Portugal	6	1
Spain	37	5
Sweden	3	-

United Kingdom	23	1
Total	252	68

Note: In recent year typical kiln size has come to be around 3000 tonnes per day. (IPPC, 2000)

In France, the production of cement is about 20 Mt in 1999 (Cimbéton, 2000).

The basic chemistry of the cement manufacturing process begins with the decomposition of calcium carbonate  $(CaCO_3)$  at about  $900^{\circ}C$  to leave calcium oxide (CaO, lime) and liberate gaseous carbon dioxide  $(CO_2)$ ; this process is known as calcination. This is followed by the clinkering process in which the calcium oxide reacts at high temperature (typically 1  $400 - 1500^{\circ}C$ ) with silica, alumina, and ferrous oxide to form the silicates, aluminates, and ferrites of calcium which form the clinker. The clinker is then ground or milled together with gypsum and other additives to produce cement.

There are four main processing routes for the manufacture of cement: the dry, semi dry, semi wet and wet processes.

At present, about 78% of Europe's cement production comes from dry process kilns, a further 16% of production is accounted for by semi dry and semi wet process kilns, with a remainder of European production – of about 6%- now coming from wet process kilns. The choice of manufacturing process is primarily based on the nature of the available raw materials.

The percentages of each type of cement supplied to domestic markets in 1994 are:

- Portland composite cement 44%
- Portland cement 43%
- Blastfurnace 7%
- Pozzolanic cement 5%
- Other cement 1%

(IPPC, 2000)

#### 2.2 Baked clay materials

Building materials produced with baked clay are essentially tiles and brickwork. Tiles are the constituents of roofing which ensure watertightness of roof. According to the application, there are different types of brickwork.

Clay is one of the most abundant mineral materials on earth. However, clay for the production of building material must possess some specific properties and characteristics. To satisfy modern production requirements, clays must have plasticity which permits them to be shaped or molded when mixed with water. Thus, additives like sand and limestone are added to give special properties. The manufacturing procedure to produce baked clay materials is divided in 6 steps:

Storage raw material: it is a common practice to store enough raw material to insure continuous operation regardless of weather conditions. Normally several storage areas (one for each source) are provided to permit some blending of the clays. Blending produces more uniform raw materials, helps to control colour and permits some control over raw material suitability for manufacturing a given type of unit.

- **Preparation:** The clay is crushed to break up large chunks and remove stones, after which large grinding wheels, weighting 4 to 8 tons each, revolve in a circular pan, grinding and mixing the raw material.
- **Forming:** The first step in the forming process produces a homogeneous, plastic mass ready for molding. It is most commonly achieved by adding water to the clay in a pug mill, a mixing chamber which contains one or more revolving shafts with blades.
- **Drying:** When wet clay units come from molding or cutting machines, they contain from 7 to 30% moisture, depending upon the forming method. Before the firing process begins, most of this water is evaporated in dryer chambers at temperature ranging from 38°C to 204°C. Drying time, which varies with the different clays, is usually from 24 hours to 48 hours. The drying must be constant so as not to change properties of materials.
- **Firing and cooling:** Firing requires from 40 to 150 hours depending upon kiln type and other variables. The heat conditions in each zone are carefully controlled. Firing may be divided into six general stages: water-smoking (evaporating free water, 204°C), dehydration (from 149°C to 982°C), oxidation (from 538°C to 982°C), vitrification (from 871°C to 1316°C), flashing and cooling. The key to the firing process is to control the temperature in the kiln so that incipient fusion and partial vitrification are complete but viscous fusion is avoided.
- **Drawing:** it is the process of unloading a kiln after cooling. At this stage, units are stored, graded, packaged and taken to a storage yard or loaded onto rail cars or trucks for delivery.

#### 2.3 Concrete

The production of concrete ready for use was about 34 million m³ in 2000 in France (SNBPE Syndicat National du Béton Prêt à l'Emploi, 2001).

Concrete is an homogeneous blend of cement, water, air, granulates and, often, additives.

<u>Table 3</u>: Composition of concrete

Components	Water	Air	Cement	Granulates
Volume (%)	14-22	1-6	7-14	60-78
Weight (%)	5-9		9-18	63-85

The composition varies according to the required properties of the concrete. (Cimbéton, 2000).

#### 2.4 Mortar

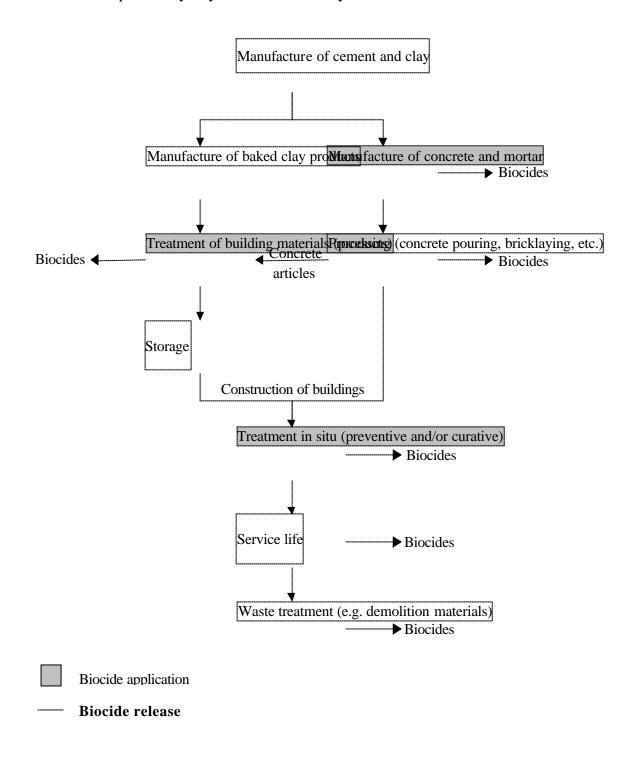
The difference between mortar and concrete is that in mortar there are no granulates. Mortar is used for fixing, blocking, joints, coat, screed...

#### **3 MAIN PROCESSES**

# 3.1 Life cycle of building materials and application of biocides

The application and releases of biocides during each life stage of building materials is presented in Figure 1. It should be noted that the figure has been simplified somewhat. The stage of construction of buildings has not been represented by a box.

However, in principle some treated building materials may end up in construction and demolition waste and some – presumably very small – releases may occur.



<u>Figure 1</u>: Biocides applications and releases during the life of building materials

#### 3.2 Manufacture of cement and clay

According to Cembureau (The European Cement Association, letter 06/03/01), Vicat (letter 06/03/01), Lafarge Ciments (personal communication 06/06/01), Ciments de l'Adour (letter 09/10/01) and Ciments d'Origny (letter 24/10/01), biocides are not used in the cement production process.

At the extraction and during processing of clay no biocides are added (Briquetterie d'Allonne letter 15/10/01).

#### 3.3 Manufacture of baked clay products

According to CTTB (Centre Technique des Tuiles et Briques, mail 13/03/01), for baked clay (tiles and bricks), biocides are not used during the stage of production.

#### 3.4 Manufacture of concrete and mortar

In general, according to professionals, no biocides are added to concrete because micro-organisms develop rarely in alkaline medium.

So, the amount of biocidal products on the market for inclusion into concrete and mortar is assumed to be small.

According to a small survey in France (2001), only one product was counted to be mixed into fresh mortar via water. The active substance of this product is diuron, its concentration in the product is below 15%.

On the other hand, additives added to concrete and mortar during the production may contain biocides. Concentrations are about < 0.5% in additives, i.e. a few ppm in building materials. These biocides are usually in-can preservatives, used to protect additives during their storage (Syndicat National des Adjuvants pour bétons et mortiers SYNAD, letter 19/04/01).

During microbiological attack, there is a release of organic or mineral acid formed by the biochemical processes. The acids initially formed react with hydrated cement and gradually dissolve it. At the beginning, the alkaline solution of hydrated cement neutralises acid. But, the continuous action of bacteria involves an attack in depth.

Because of the roughness of concrete, surface cleaning is ineffective, therefore it is necessary to incorporate biocides in concrete. (Neville, A.M., 2000). This objective is often achieved by the use of potent antibacterial, fungicidal, insecticidal admixtures in the mix (Ramachandran, 1984).

A survey of the literature shows that few investigators have been interested in the subject of the use of chemical admixtures to inhibit microbiological attack. There is, therefore, a dearth of information relating to the effects of the chemicals used on other properties of concrete. Accordingly, no field practice, proprietary products and standards covering their use exist (Ramachandran, 1984).

It has been reported that a great rate of destruction of the micro-organisms occurs both on the surface and within the matrix of the concrete when bactericidal admixtures are used. However, the effects are reported to be transient and not always completely effective. Effectiveness of the admixture is dependent on the degree of water solubility of the materials and the method of incorporation into the mix. Water soluble materials when added via the mix water, are said to be readily leached from the concrete and soon lose their effectiveness (Ramachandran, 1984).

#### 3.5 Treatment of shaped building materials

It is not clear yet whether these articles are surface treated with biocidal products after production and before being marketed.

But according to limited information, no biocides are used during storage of baked clay products (Briqueterie d'Allonne, personal communication, 2001).

#### 3.6 Treatment of buildings in situ (preventive and curative)

#### 3.6.1 Types of substrates and active substances

Many surfaces, made of building materials (cement, concrete, stone or baked clay) such as roofs, walls, façades, paths, terraces, etc. are affected by the development of mosses, lichen, and algae after some time. In general, the development of mosses, lichen and algae occurs outdoors.

Some biocides are able to protect and/or cure building materials. Materials which have been found most effective in imparting bactericidal and fungicidal properties include polyhalogenated phenols, sodium benzoate, benzalkonium chloride and copper compounds. Other materials used as liquid admixtures for preventing bacterial growth include sodium-o-phenylphenate and alkyldimethyl benzyl ammonium chloride (Ramachandran, 1984).

Other substances used are often quaternary ammonium compounds, copper oxyquinolate and derivatives, bromoacetic acid, benzalkonium chloride, sodium hydroxide and sodium hypochloride (CERIB, letter 09/04/01; specification sheets of products). Table 4 presents several products used to treat surfaces (results of a small survey in France in 2001 and in the United Kingdom in 2002, further details are presented in the Annex n°3).

<u>Table 4</u>: Examples of biocidal active substances and amounts applied on surfaces identified in France and in the United Kingdom in 2001-2002 (further details are presented in the Annex n°3)

Active substance	Concentration	Support	Application	Post-application	Amount of
	in product				product applied
Quaternary ammonium salts		Soil, walls, roofs		After 1 or 2 days, clean with high pressure sprayer	0.2 l/m²
Bromoacetic acid Benzalkonium chloride	≤ 2.5% ≤ 2.5%	Stones	Roller, brush, sprayer	After at least 2 hours, rinse with water at low pressure	$0.15 - 0.3 \text{ l/m}^2$
Bromoacetic acid Benzalkonium chloride	≤ 2.5% ≤ 2.5%	Soils, walls, roofs, terraces	Roller, brush, sprayer	After at least 2 hours, rinse with water at low pressure	$0.15 - 0.2 \text{ l/m}^2$
Alkylcocodiméthylbenzyl ammonium chloride	1%	Roofs, terraces, tennis-court	Roller, sprayer	Preventive action: no rinse <u>Curative action</u> : after 1 week, clean with high pressure sprayer	0.5 l/m²
Quaternary ammonium, biguanide	10%	Façades, walls, soils	Sprayer, brush, roller	No rinse	
Quaternary ammonium (alkylcocodimethylbenzyl ammonium chloride, sodium hydroxyde)	1 – 5%	,	High pressure cleaner, brush, sprayer	Cleaning high pressure sprayer	
Lauryldimethylbenzylam monium bromide	2.5 – 10%				0.1 – 0.15 l/m²
Sodium hypochlorite	5%	Buildings	Sprayer	High pressure rinse (50 – 70 bars) after 30 minutes	200 g/m²
Quaternary ammonium salts		Façades, roofs, soils, terraces	Low pressure sprayer, brush, roller	No rinse (preventive action)	0.2 – 1 l/m²
Sodium hypochloride, fatty amine derivative		Façades, roofs, soils, terraces	Dripping sprayer, brush, roller	After 2 hours, rinse (curative action)	0.2 – 1 l/m²
Formaldehyde, Glyoxal Glutaral, Didecyldimethylammoniu m chloride	2.5 – 10%	Façades, roofs, tennis- court	Sprayer	No rinse	
Benzalkonium chloride	16%	Façade	Sprayer, roller	No rinse	
Dodecylamine salicylate and lactate	0.5 – 5%	All surface		No rinse	
Triazine derivatives and isothiazolone derivatives	1%	Façade, roof, tennis-court	Sprayer, roller, brush	No rinse	
2-phenyphenol Benzalkonium chloride	0.25% 0.99%	Building	Sprayer	Rinse	

Active substance	Concentration in product	Support	Application	Post-application	Amount of product applied
Permethrin Tri(hexyleneglycol) biborate	0.2% 3.3%	Building	Sprayer	Rinse	
Benzalkonium chloride Biphenyl-2-ol	2% 1%	Façade, roof, building	Sprayer, roller	No rinse	
Alkyldimethylbenzyl ammonium chloride	20%	Façade, roof, building, monument	Sprayer, roller	No rinse	
Benzalkonium chloride	1%	Façade, roof, monument, building	Roller	No rinse	
Benzalkonium chloride	5%	Façade, roof	Sprayer, roller	Rinse	
Disodium octaborate tetrahydrate	10%	Building	Sprayer	No rinse	
IPBC	0.2%	Building	Sprayer	No rinse	

Note: Most of the products quoted in Table 4 are considered as disinfectants. In general, they are used for a curative treatment and not preventive.

The active substances mentioned in Table 4 are given as examples. In other countries they may not be authorised.

A questionnaire was been also sent to paint companies. Table 5 presents results of this survey. In general, the application is done once at any season. After application of these products, there is no rinse. These products are also covered by product type 7 "film preservatives" but they are considered in the same way as masonry preservatives even if the formulation types and leaching behaviour will be different.

<u>Table 5</u>: Examples of biocidal active substances used by paint professionals

Active substance	Support	Application	Remarks	
Quaternary ammonium	Façade, roof, monument,	Sprayer (0-1	Used as preventive/curative	
compounds (20-50%)	building, road, tennis-court	bar), roller	treatment before painting	
Bromoacetic acid (2%)	Façade, roof, monument,	Sprayer (0-1	Used as preventive/curative	
Benzalkonium chloride	building, tennis-court	bar), roller	treatment before painting	
(25%)				
Tributyltin naphtenate (1%	Façade, roof, monument,	Sprayer (0-1	Used as preventive/curative	
Sn)	building	bar), roller	treatment before painting	
Quaternary ammonium				
MI/MCI (1.3%)	Façade, roof, monument,	Roller	Product added to the paint	
Octyl isothiazolone (5-	building			
10%)				
Diuron (10-25%)	Façade, roof, building	Roller	Ready to use paint (contains	
Carbendazim (2.5-10%)			already a biocidal additive)	
Octyl isothiazolone (2.5-			-	
10%)				

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#### 3.6.2 Application of the biocides

Chemicals used as masonry preservatives are usually applied in an aqueous solution.

The same treatment is applied for preventive and curative action. Nevertheless, in the case of curative action, building materials are, before treatment, cleaned to a maximum with a high pressure cleaner. According to Table 4, the application of the product is performed with long bristle rollers, sprayers, brush sprayers, high pressure cleaners or a wash. For some products, there is a rinse after application. The rinse is in general performed with a high pressure sprayer. But sometimes a low pressure sprayer is recommended.

In order to have a preventive action, the treatment must generally be carried out once a year. The treatment can be done at any season (further details are presented in the Annex n°3). The treatment may be performed by professionals or non-professionals.

To assess releases, two application methods to be considered: sprayer and roller. The releases during application with a brush are considered to be covered by those estimated for application with a roller. A scenario has been developed for each mode of application. And since there is sometimes a rinse after application of the biocidal product (further details are presented in the Table 4), these releases must be assessed too.

#### 4 RELEASE ESTIMATION

#### 4.1 Releases during the treatment

Industrial processes are considered to be continuous while in-situ treatments (both professional and by the public at large) are considered discontinuous (unless otherwise indicated, it is considered that the emissions occur within one day).

#### 4.1.1 Industrial sites

As follows from Chapter 3, there is usually no treatment at industrial sites.

However, paints and additives (for concrete or mortar) may contain biocides which protect the paints and additives (in-can preservatives). So, during the manufacture of concrete, mortar or paints, releases of biocides may occur at industrial sites.

For the formulation of paints, the release scenario developed for industrial substances (IC 14) can be used. No specific information is available for the releases during formulation of concrete or mortar.

#### 4.1.2 Treatment in situ

Whichever the building type, there are two major ways of application: sprayer and roller. Emission routes which should be considered are:

SPRAYING Drift: Emission to air

Deposition on soil (leaching to groundwater)

Emission to water

Runoff: Emission to soil (leaching to groundwater)

Emission to water

ROLLING Spilling Emission to soil (leaching to groundwater)

& BRUSHING & Dripping Emission to water

A building to be treated can be located either in the countryside or within a city.

If it is located within a city, the losses due to spray-drift or dripping (or runoff) are likely to fall upon paved ground and washed with rain to the sewer system. In a more rural setting, the losses are more likely to end up on unpaved soil. These two settings are therefore distinguished throughout this document.

In this document, only roofs and façades are taken into account because they are representative for the different types of surfaces to be treated.

Actually other substrates can be treated with masonry preservatives such as terraces, monuments, streets, pavements, pathways, tennis-courts... No default values for these substrates have been derived yet. In a first approach, the releases will be considered to be similar as for a roof or a façade. And the scenarios for the roof and the façade can be modified if additional information becomes available.

The following table shows receiving compartments of releases due to treatment and rinse according to building type, place and method of application. For the rinse, the releases are the same as for the treatment performed with a sprayer.

<u>Table 6</u>: Receiving compartments of release of biocides during on-site application and rinse

		SPRAYING		ROLLING/	RINSE	
SUBSTRATE	PLACE	Drift	Runoff	BRUSHING	Drift	Runoff
ROOF	Countryside	Soil	Soil	Soil	Soil	Soil
		Air			Air	
	City	Water	Water	Water	Water	Water
		Air			Air	
FACADE	Countryside	Soil	Soil	Soil	Soil	Soil
		Air			Air	
	City	Water	Water	Water	Water	Water
		Air			Air	

#### 4.1.2.1 *Sprayer*

During the application with a sprayer, two phenomena occur: drift and runoff.

Drift is the movement of spray droplets beyond the target zone. Hewitt (2000) considers pesticide drift to be the movement of pesticide through the air at the time of pesticide application or soon thereafter from the target site to any non- or off-target site, excluding pesticide movements by erosion, migration, volatility or wind-blown soil particles after application. The same definition can be taken for the drift of biocidal substances.

Droplets less than 50 microns in diameter remain suspended in the air indefinitely or until they evaporate (further details are presented in the Annex  $n^{\circ}1$ ). So, a fraction of the product can be lost to the air compartment.

Because of runoff the product can reach either the soil compartment, if the building is located in the countryside, or rainwater if the building is located in the city. An additional fraction of the product can be lost to these compartments because of the deposition of droplets due to drift. But in the countryside, spray drift and runoff do not reach the same soil. Runoff reaches soil adjacent to the treated surface. Spray drift reaches soil further away from the surface (cf. Figure 2).

Thus,  $E_{\text{local\_soil(d)}}$  represents the local emission of a substance during application (with sprayer) to soil distant to the treated surface. And  $E_{\text{local\_soil(a)}}$  represents the local emission of a substance during application (with sprayer or roller) to soil adjacent to the treated surface.

Note: Emissions to soil may subsequently reach the ground water.

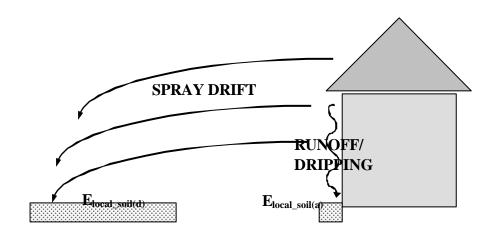


Figure 2: Soils exposed by runoff or dripping and spray drift

According to Annex n° 1, the only data and models available about spray drift concern pesticides used in agriculture. Table 24 shows for hops the fraction of lost product by spray drift. This fraction is about 40%. However, the used pressure ranged between 16 and 29 bars. So, during application of biocides onto buildings, the fraction of lost product may be lower since the used pressure is usually below or up to 3 bars (further details are presented in the Annex n°3, Table 28 & Table 29). On the other hand, according to French and British companies, the fraction of product which drifts ranges between 1 and 10% (further details are presented in the Annex n°3, Table 28 & Table 29).

As a worst case, in a very first approach, a figure of 10% is proposed as a default value (Fdrift = 0.1) for the present scenario.

For the phenomenon of runoff, only data from a small survey performed in France (2001) and in the United Kingdom (2002) are available (further details are presented in the Annex  $n^{\circ}3$ ). For companies, the fraction of product lost during application due to runoff is below or up to 20%. Thus, as a worst case, in a very first approach, a figure of 20% is proposed as a default value (Frunoff = 0.2).

Note: these figures are based on survey data which is not supported by raw data or published academic references.

#### 4.1.2.2 Roller or brush

During rolling and brushing, product losses can occur due to spilling and dripping. Default values for losses during application are presented in Table 7. These are based on those estimated for the application of wood preservatives (OECD, 2002).

Table 7: Default values for losses during brushing

	Fdripping
Professionals	0.03
Amateurs	0.05

These losses will end up in soil adjacent to the treated surface (cf. Figure 2), if the building is located in the countryside and the soil is not protected with a plastic cover. If the building is located within an urban area, then losses will enter rainwater drains.

Note: Emissions to soil may subsequently reach the ground water.

#### 4.1.2.3 Substrates

Representative types of surfaces to be treated are roofs and façades. The dimensions chosen are those already proposed in the scenario for wood preservatives (OECD, 2002).

Thus, for the roof, the scenario describes a roof with a pitch of 25 °, the length is 17.5 m and the width is 7.5 m (cf. Figure 3). The total treated area is 145 m². In this scenario, it is considered that the roof does not have a gutter. So, the releases due to dripping and runoff end up in soil if the roof is located in the countryside or in rainwater if it is located in the city.

Remark: In countries where most houses have a roof-gutter, the scenario can be easily adapted: all releases due to dripping and runoff are collected and end up in rainwater. The rainwater may be discharged to an STP or directly to surface water.

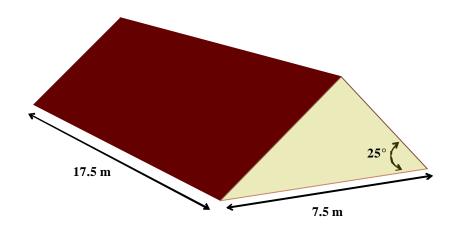


Figure 3: Schematic drawing of the roof

For the façade, the model house already proposed in OECD (2002) is used. The height of the house is 2.5 m and the perimeter is 50 m (17.5 m long and 7.5 m wide), see Figure 4. So, the total treated surface of the house is 125 m<sup>2</sup>.

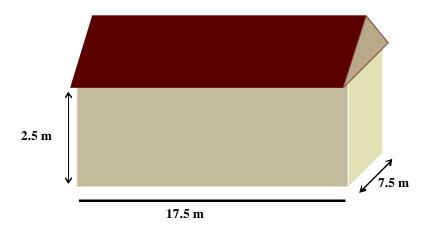


Figure 4: Schematic drawing of the house

For both roof and façade it is assumed that the treatment takes place within one day.

# 4.1.2.4 Dimensions of the receiving soil

In the countryside, spray drift and runoff or dripping do not reach the same soil. Runoff or dripping reaches soil adjacent to the treated house. However, the deposition of spray drift reaches soil further away from the house (cf. Figure 2).

The default values for the size of the soil receiving runoff and dripping are: 10 cm distance from the house and 10 cm deep. The perimeter of house is 50 m. So, the volume soil adjacent to the house is 0.50 m<sup>3</sup> (Vsoil(a)) (OECD, 2002) (cf. Figure 5).

To define the dimension of soil distant to treated surface, the important parameters are: wind speed, height of release and settling velocity. The average wind speed measured at 10 m height is given in Table 8 for 5 European countries.

Table 8: Average wind speed measured at 10 m height (Power Technology Centre, 2001)

Countries	Average wind speed (m.s <sup>-1</sup> )
Spain	4
Belgium	4
Netherlands	5.5
Germany	3
France	3.5
Average	4

According to Annex n°3 about a small survey performed in France (2001) and in the United Kingdom (2002), spraying is done with flat fan nozzles, full cone nozzles or hollow cone nozzles at a pressure ranging between 0 and 3 bars.

For a pressure of 3 bars, Table 20 (Annex n°1) gives a Volume Median Diameter between 220 and 850 microns depending on the type of nozzles. Besides according to Table 21 (Annex n°1) and the diameter of droplets, the settling velocity is between 0.83 m.s<sup>-1</sup> (droplets of 220 microns) and 2.46 m.s<sup>-1</sup> (droplets of 850 microns). As a worst case, the value of 2.46 m.s<sup>-1</sup> will be used as a default value: the receiving compartment will be smaller and so the concentration of the active substance in soil will be higher.

The height of release is the height of the façade (2.5 m) added to the height of the roof (1.75 m), i.e. 4.25 m, taking into account both treatment of the roof and the façade.

The distance travelled by drift can be calculated as:

$$S = \frac{U \times H}{V}$$

where S = Drift [m]

U = Wind speed [m.s<sup>-1</sup>]

H = Height of release [m]

V = Settling velocity [m.s<sup>-1</sup>]

The width of the receiving compartment is:

$$\frac{4\times4.25}{2.46}$$
 = 6.9m

Figure 5 below shows the dimensions of the receiving soil around the treated house. This includes the soil distance from the house and the volume of soil receiving the treatment fluid (Vsoil(d)) which according to the worked example is 54.1 m<sup>3</sup>.

Note: the volume of the receiving compartment might be modified if measured data are available.

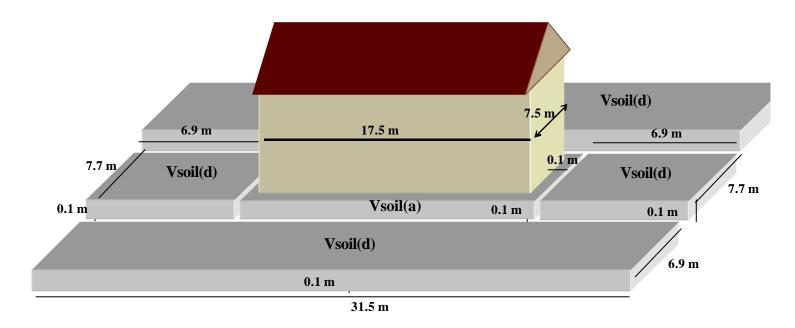


Figure 5: Dimensions of the receiving environmental compartment

Note: if the treatment is performed only on a façade, the height of release is 2.5 m and not 4.25 m. So, the volume soil distant to the house will be 27.3 m<sup>3</sup>.

#### 4.2 Releases during service life

After application, emissions to soil from the treated structure may occur due to frequent wetting by rainfall and subsequent leaching from the treated substrate.

The estimation of emissions from treated products during their service life should be based on standardised leaching tests. These tests should allow to determine the quantity of an active ingredient, leached out of the product due to rainfall, per surface and time.

The leaching tests should be performed with similar philosophy as for wood preservatives (OECD, 2002).

The dimensions of the receiving soil compartment would be the same as those for runoff and dripping (i.e. 0.5 m<sup>3</sup>).

Note: If biocidal products are used during the making of building materials and during their storage, releases can occur during their service life. The estimation of these releases will also be assessed with leaching tests.

#### 4.3 Disposal of material

At the end of their life, building materials can be either landfilled or recycled to produce new building materials. So, releases can occur in the landfills or during their new service life. In this document, these potential releases are not taken into account. However, specific scenarios might have to be developed in the future.

#### 5 RELEASE SCENARIOS

In this chapter, methods are provided to calculate the concentration of an active ingredient or any other substance included in the formulation in the local environment. The calculations proposed here do not take into account removal processes of the substances from the receiving compartment due to for example degradation, volatilisation or leaching to ground water. The calculated concentrations are initial concentrations. Proposals to take into account removal processes are described in Annex n°2.

#### 5.1 Releases during the treatment of building materials in an industrial setting

In first approach, it is not necessary to develop a emission scenario for calculating the releases from the making of building materials and their storage. According to available information, biocides are not added during the production and the storage of building materials.

#### 5.2 Releases during the treatment in situ

# 5.2.1 Treatment of a roof by sprayer

In most cases manual spraying is applied for the treatment in situ. The emission scenario is presented in Table 9.

The roof and the façade can be treated at the same time. Therefore, the same scenario is used twice and the results are added.

During application on the roof, there is a fraction of the product which is lost due to runoff. If the treated surface is located in the countryside, this fraction reaches soil adjacent to treated surface (cf. Figure 2). If it is located in the city, the fraction ends up in the storm water.

Furthermore, the application of the product involves also releases into the air because of spray drift. Releases to air are not taken into account in this document, as they give rise to very short exposure on a local scale.

But after spray drift, there can also be deposition of droplets onto soil. If the treated surface is located in the countryside, the spray drift reaches soil at some distance to the treated surface (cf. Figure 2) which is often unpaved. The dimensions of the receiving compartment are proposed in Chapter 4.1.2.3.

If the treated surface is located in a city, the soil is often paved. So, the releases reach storm water and can be added to the releases due to runoff.

Note: the amount of treated houses in the same period in a city of 10 000 inhabitants (TGD default) is not known. A provisional default value of not more than one house per day is assumed.

Table 9: Emission scenario for calculating the releases from a roof treated by sprayer

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Treated area of a roof per day	$AREA_{roof}$	$m^2.d^{-1}$	145	D
Volume of product applied on area	Vform	$l.m^{-2}$		S
Fraction of active substance in product	Fform	-		S
Density of product	RHOform	kg.m <sup>-3</sup>	1000	S
Fraction of product lost during application by spray drift (cf. §. 4.1.2.1)	Fdrift	- 0.1		D
Fraction of product lost during application due to runoff (cf. § 4.1.2.1)	Frunoff	-	0.2	D
Soil volume distant to treated surface (cf. § 4.1.2.3)	Vsoil(d)	$m^3$	54.1	D
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	$m^3$	0.5	D
Bulk density of wet soil	RHOsoil	kg.wwt.m <sup>-3</sup>	1 700	D
Output:				
Local emission of active substance during application due to spray drift	$Elocal_{spray\_drift,roof}$	kg.d <sup>-1</sup>		
Local emission of active substance during application due to runoff	Elocal <sub>runoff,roof</sub>	kg.d <sup>-1</sup>		
Local emission of active substance during application (sprayer) to storm water	Elocal <sub>spray,roof,water</sub>	kg.d <sup>-1</sup>		
Local concentration of active ingredient in soil (distant to treated surface) resulting from application	$Clocal_{spray, roof, soil(d)} \\$	kg.kg <sub>wwt</sub> -1		
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application	Clocal <sub>spray,roof,soil(a)</sub>	kg.kg <sub>wwt</sub> -1		

# Model calculation

$$Elocal_{spray\_drift,roof} = AREA_{roof} \cdot Vform \cdot Fform \cdot RHOform \cdot Fdrift \cdot 10^{-3}$$
 (1)

The local concentration of the active ingredient in soil is only calculated for a treated surface located in the countryside.

Releases to water are collected by the storm water system. A common collection system for waste water and storm water is considered by default in this scenario. So, the local concentration in surface water can be calculated according to the general methods described in the TGD. However, if the collection system for storm water and waste water is separated, emissions are released directly to surface water, and the general methods described in the TGD need to be adapted for the calculation of the local concentration considering that the storm water will not be treated in a sewage treatment plant.

#### In the countryside

$$Clocal_{spray,roof,soil(d)} = \frac{Elocal_{spray\_drift,roof}}{Vsoil(d) \times RHOsoil}$$
(3)

$$Clocal_{spray,roof,soil(a)} = \frac{Elocal_{runoff,roof}}{Vsoil(a) \times RHOsoil}$$
(4)

#### In a city

$$Elocal_{spray,roof,water} = Elocal_{runoff,roof} + Elocal_{spray\_drift,roof}$$

$$(5)$$

# 5.2.2 Treatment of a façade by sprayer

As for the treatment of a roof, the treatment of a façade by sprayer involves two phenomena: spray drift and runoff.

Releases to air are not taken into account in this document, as they give rise to very short exposure on a local scale.

However, for the deposition after spray drift and runoff, the releases reach either the soil (if the treated surface is located in the countryside) or storm water (if the treated surface is located in a city).

Table 10: Emission scenario for calculating the releases from a façade treated by sprayer

Variable/parameter	Symbol	Unit	Default	S/D/O/P	
Input:					
Treated area of a façade per day	AREA <sub>façade</sub>	m <sup>2</sup> .d <sup>-1</sup> 125		D	
Volume of product applied on area	Vform	l.m <sup>-2</sup>		S	
Fraction of active substance in product	Fform	-	-		
Density of product	RHOform	kg.m <sup>-3</sup>	1000	S	
Fraction of product lost during application by spray drift (cf. § 4.1.2.1)	Fdrift	-	- 0.1		
Fraction of product lost during application due to runoff (cf. § 4.1.2.1)	Frunoff	-	0.2	D	
Soil volume distant to surface treated (cf. § 4.1.2.3)	Vsoil(d)	m <sup>3</sup>	54.1*	D	
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	m <sup>3</sup>	0.5	D	
Bulk density of wet soil	RHOsoil	kg <sub>wwt</sub> .m <sup>-3</sup>	1 700	D	
Output:					
Local emission of active substance during application due to spray drift	Elocal <sub>spray_drift,façade</sub>	kg.d <sup>-1</sup>			
Local emission of active substance during application due to runoff	Elocal <sub>runoff,façade</sub>	kg.d <sup>-1</sup>			
Local emission of active substance during application (sprayer) to storm water	Elocal <sub>spray,façade,water</sub>	kg.d <sup>-1</sup>			
Local concentration of active ingredient in soil (distant to treated surface) resulting from application	$Clocal_{spray,façade,soil(d)} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>			
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application  * This default value considers that the facade		kg.kg <sub>wwt</sub> -1			

<sup>\*</sup> This default value considers that the façade and the roof are treated the same day. Otherwise, if only the façade is treated, the soil volume distant to treated façade is  $27.3 \text{ m}^3$ .

#### Model calculation

$$Elocal_{spray\_drift,façade} = AREA_{façade}$$
 'Vform 'Fform 'RHOform 'Fdrift ' $10^{-3}$  (6)

$$Elocal_{runoff,façade} = AREA_{façade} \cdot Vform \cdot Fform \cdot RHOform \cdot Frunoff \cdot 10^{-3}$$
 (7)

If the treated surface is located in the countryside, spray drift and runoff won't reach the same soil. Runoff reaches soil adjacent to the treated surface. The deposition after spray drift reaches soil further away from the surface (cf. Figure 2).

The local concentration in surface water can be calculated according to the method described in chapter 5.2.1.

## In the countryside

$$Clocal_{spray,façade,soil(d)} = \frac{Elocal_{spray\_drift,façade}}{Vsoil(d) \times RHOsoil}$$
(8)

$$Clocal_{spray, façade, soil(a)} = \frac{Elocal_{runoff, façade}}{Vsoil(a) \times RHOsoil}$$
(9)

#### In a city

$$Elocal_{spray,façade,water} = Elocal_{spray\_drift,façade} + Elocal_{runoff,façade}$$
 (10)

As the roof and façade are often treated at the same time, the local emissions can be added as shown in Table 11.

Table 11: Emission scenario for calculating the releases from a house treated by sprayer

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Local emission of active substance during application (sprayer) to storm water	Elocal <sub>spray,façade,water</sub>	kg.d <sup>-1</sup>		O
Local concentration of active ingredient in soil (distant to treated surface) resulting from application	$Clocal_{spray,façade,soil(d)}$	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application	$Clocal_{spray,façade,soil(a)} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Local emission of active substance during application (sprayer) to storm water	Elocal <sub>spray,roof,water</sub>	kg.d <sup>-1</sup>		O
Local concentration of active ingredient in soil (distant to treated surface) resulting from application	$Clocal_{spray, roof, soil(d)} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application	1 3, , , , ,	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Output:				
Local emission of active substance to storm water from a house treated by sprayer	Elocal <sub>spray,water</sub>	kg.d <sup>-1</sup>		
Total concentration of active ingredient in soil distant to treated surface resulting from application (sprayer)	$Clocal_{spray, soil(d)}$	kg.kg <sub>wwt</sub> <sup>-1</sup>		
Total concentration of active ingredient in soil adjacent to treated surface resulting from application (sprayer)	Clocal <sub>spray,soil(a)</sub>	kg.kg <sub>wwt</sub> <sup>-1</sup>		

## Model calculation

The local concentration in surface water can be calculated according to the method described in chapter 5.2.1.

# In the countryside

$$Clocal_{spray,soil(d)} = Clocal_{spray,roof,soil(d)} + Clocal_{spray,facade,soil(d)}$$

$$(11)$$

$$Clocal_{spray,soil(a)} = Clocal_{spray,façade,soil(a)} + Clocal_{spray,roof,soil(a)}$$
 (12)

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## In a city

$$Elocal_{spray,water} = Elocal_{spray,roof,water} + Elocal_{spray,façade,water}$$
(13)

# 5.2.3 Treatment of a façade with a roller or a brush

During brushing, product losses are due to drips and spills. These losses will end up in soil adjacent to the façade, if the soil is not paved or protected (countryside) or storm water if the soil is paved (city). The dimensions are the same than for runoff due to spray-application (cf. § 4.1.2.3).

Table 12: Emission scenario for calculating the releases from a façade treated with roller or brush

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Treated area of a façade per day	$AREA_{façade}$	$m^2.d^{-1}$	125	D
Volume of product applied on area	Vform	$l.m^{-2}$		S
Fraction of active substance in product	Fform	-		S
Density of product	RHOform	kg.m <sup>-3</sup>	1000	S
Fraction of product lost during application due to dripping (cf. Table 7)	Fdripping	-		P
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	$m^3$	0.5	D
Bulk density of wet soil	RHOsoil	kg <sub>wwt</sub> .m <sup>-3</sup>	1 700	D
Output:				
Local emission of active substance during application	Elocal <sub>drip,roll,façade</sub>	kg.d <sup>-1</sup>		
Local emission of active substance during application (roller) to storm water	Elocal <sub>roll,façade,water</sub>	kg.d <sup>-1</sup>		
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application	$Clocal_{roll,façade,soil(a)} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>		

#### Model calculation

$$Elocal_{drip,roll,façade} = AREA_{façade} \text{ 'Vform 'Fform 'RHOform 'Fdripping '10}^{3}$$
(14)

If the treated surface is located in the countryside, releases reach the soil adjacent to the treated surface. So, the local concentration in soil can be calculated.

The local concentration in surface water can be calculated according to the method described in chapter 5.2.1.

#### In the countryside

$$Clocal_{roll,façade,soil(a)} = \frac{Elocal_{drip,roll,façade}}{Vsoil(a) \times RHOsoil}$$
(15)

#### In a city

$$Elocal_{roll,façade,water} = Elocal_{drip,roll,façade}$$
(16)

#### 5.2.4 Treatment of a roof with a roller or a brush

During brushing, product losses are due to drips and spills. These losses will end up in the soil adjacent to the treated surface if it is located in the countryside or in the storm water if the treated surface is located in the city. The local concentration in surface water can be calculated according to the method described in chapter 5.2.1. The local concentration of the substance in soil adjacent to the treated surface is calculated according to the model described below.

Table 13: Emission scenario for calculating the releases from a roof treated with roller or brush

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Treated area of a roof per day	$AREA_{roof}$	$m^{2}.d^{-1}$	145	D
Volume of product applied on area	Vform	l.m <sup>-2</sup>		S
Fraction of active substance in product	Fform	-		S
Density of product	RHOform	kg.m <sup>-3</sup>	1000	S
Fraction of product lost during application due to dripping (cf. Table 7)	Fdripping	-		P
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	m <sup>3</sup>	0.5	D
Output:				
Local emission of active substance to rainwater during application	$Elocal_{drip,roll,roof} \\$	kg.d <sup>-1</sup>		
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application	$Clocal_{roll,roof,soil(a)}$	kg.kg <sub>wwt</sub> <sup>-1</sup>		
Local emission of active substance during application (roller) to storm water	$Elocal_{roll,roof,water}$	kg.d <sup>-1</sup>		

#### Model calculation

$$Elocal_{drip,roll,roof} = AREA_{roof} \times Vform \times Fform \times RHOform \times Fdripping \times 10^{-3}$$
(17)

# In the countryside

$$\overline{Clocal_{roll,roof,soil(a)}} = \frac{Elocal_{drip,roll,roof}}{Vsoil(a) \times RHOsoil}$$
(18)

## In a city

$$Elocal_{roll,roof,water} = Elocal_{drip,roll,roof}$$
(19)

As the roof and façade are often treated at the same time, the local emissions can be added as shown in Table 14.

Table 14: Emission scenario for calculating the releases from a house treated with roller or brush

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				_
Local emission of active substance during application (roller) on a roof to waste water	, , , , , , , , , , , , , , , , , , ,	kg.d <sup>-1</sup>		О
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application	$Clocal_{roll,roof,soil(a)}$	kg.kg <sub>wwt</sub> <sup>-1</sup>		О
Local emission of active substance during application (roller) on a façade to waste water	Elocal <sub>roll,façade,water</sub>	kg.d <sup>-1</sup>		O
Local concentration of active ingredient in soil (adjacent to treated surface) resulting from application		kg.kg <sub>wwt</sub> <sup>-1</sup>		О
Output:				
Local emission of active substance to storm water from a house treated by roller	Elocal <sub>roll,water</sub>	kg.d <sup>-1</sup>		
Total concentration of active ingredient in soil adjacent to treated surface resulting from application	, , ,	kg.kg <sub>wwt</sub> <sup>-1</sup>		

## Model calculation

The local concentration in surface water can be calculated according to the method described in chapter 5.2.1.

# In the countryside

$$Clocal_{roll,soil(a)} = Clocal_{roll,façade,soil(a)} + Clocal_{roll,roof,soil(a)}$$
(20)

# In a city

$$Elocal_{roll,water} = Elocal_{roll,roof,water} + Elocal_{roll,façade,water}$$
(21)

## 5.2.5 Releases during rinse

# 5.2.5.1 Fraction of product lost during rinse

The rinse is an optional stage which is used for cleaning the treated surface. The rinse is often performed with a high pressure sprayer (further details are presented in the Annex n°3). Between the application stage and the rinse, the applied substances can degrade, evaporate or be fixed onto the substrate. This fraction of the applied quantities (Felim) will not be washed off during the rinse. Data on the elimination processes will normally not be available and therefore in a first approach Felim will be set to zero.

Furthermore the fraction of the product already lost during application has to be taken into account.

<u>Table 15</u>: Fraction of product lost during rinse

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Fraction of product eliminated between application and rinse	Felim	-	0	D
Fraction of product lost during application by spray drift (cf. § 4.1.2.1)	Fdrift	-	0.1	D
Fraction of product lost during application due to runoff (cf. § 4.1.2.1)	Frunoff	-	0.2	D
Fraction of product lost during application due to dripping (cf. Table 7)	Fdripping	-		Р
Output:				
Fraction of product lost during rinse	Frinse	-		

#### Model calculation

## Rinse after treatment with sprayer

$$Frinse = 1 - Fdrift - Frunoff - Felim$$
 (22)

#### Rinse after treatment with roller or brush

$$Frinse = 1 - Fdripping - Felim (23)$$

### 5.2.5.2 Rinse with high pressure sprayer

During the rinse, there are releases due to runoff and due to spray drift. 70 - 80% of runoff and 20 - 30% of spray drift can be assumed (Kärcher, Personal communication, 2001).

Table 16 gives local emissions of active substance during rinse due to spray drift and runoff.

Table 16: Emission scenario for calculating the releases from rinsing a house after treatment

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Quantity treated area of a façade per day	$AREA_{façade}$	$m^2.d^{-1}$	125	D
Quantity treated area of a roof per day	$AREA_{roof}$	$m^2.d^{-1}$	145	D
Quantity of product initially applied on area	Vform	$l.m^{-2}$		S
Fraction of active substance in product	Fform	-		S
Density of product	RHOform	kg.m <sup>-3</sup>	1000	S
Fraction of product lost during rinse (cf. 5.2.5.1)	Frinse	-		O
Fraction of rinsing solution lost during rinse due to runoff	Frunoff <sub>rinse</sub>	-	0.75	D
Fraction of rinsing solution lost during rinse by spray drift	Fdrift <sub>rinse</sub>	-	0.25	D
Output:				
Local emission of active substance to air during application due to drift	$Elocal_{rinse,drift,roof}$	kg.d <sup>-1</sup>		
Local emission of active substance during application due to runoff	Elocal <sub>rinse,runoff,roof</sub>	kg.d <sup>-1</sup>		
Local emission of active substance to air during application by spray drift	Elocal <sub>rinse,drift,façade</sub>	kg.d <sup>-1</sup>		
Local emission of active substance during application due to runoff	Elocal <sub>rinse,runoff,façade</sub>	kg.d <sup>-1</sup>		

#### Model calculation

$$Elocal_{rinse,drift,roof} = AREA_{roof}$$
 'Vform 'Fform 'RHOform 'Fdrift\_{rinse}' Frinse '10<sup>-3</sup> (24)

$$Elocal_{rinse,drift,façade} = AREA_{façade}$$
 'Vform 'Fform 'RHOform 'Fdrift<sub>rinse</sub> 'Frinse '10<sup>-3</sup> (25)

$$Elocal_{rinse,runoff,roof} = AREA_{roof}$$
 'Vform 'Fform 'RHOform 'Frunoff\_{rinse}' Frinse '10<sup>-3</sup> (26)

$$Elocal_{rinse,runoff,façade} = AREA_{façade}$$
 'Vform 'Fform 'RHOform 'Frunoff\_{rinse} 'Frinse '10<sup>-3</sup> (27)

# 5.2.5.3 Releases and local concentration of active substance into the different compartments

According to the small survey performed in France and in England (further details are presented in the Annex  $n^{\circ}$ 3), the rinse is done with a high pressure sprayer. According to Annex  $n^{\circ}$ 1, the droplet size is almost the same than during application with a sprayer. So, the dimensions of the receiving compartment are also the same.

Table 17: Emission scenario for calculating local concentrations due to the rinse of a house

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Local emission of active substance from roof during application due to spray drift (cf. § 5.2.5.2)	, , ,	kg.d <sup>-1</sup>		O
Local emission of active substance from roof during application due to runoff (cf. § 5.2.5.2)	Elocal <sub>rinse,runoff,roof</sub>	kg.d <sup>-1</sup>		О
Local emission of active substance from façade during application by spray drift (cf. § 5.2.5.2)	Elocal <sub>rinse,drift,façade</sub>	kg.d <sup>-1</sup>		О
Local emission of active substance from façade during application due to runoff (cf. § 5.2.5.2)	Elocal <sub>rinse,runoff,façade</sub>	kg.d <sup>-1</sup>		O
Soil volume distant to surface treated (cf. § 4.1.2.3)	Vsoil(d)	$m^3$	54.1	D
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	m <sup>3</sup>	0.5	D
Bulk density of wet soil	RHOsoil	kg <sub>wwt</sub> .m <sup>-3</sup>	1 700	D
Output:				
Local emission of active substance to storm water during rinse	Elocal <sub>rinse,water</sub>	kg.d <sup>-1</sup>		
Local concentration of active ingredient in soil distant to treated surface resulting from rinse	$Clocal_{rinse, soil(d)}$	kg.kg <sub>wwt</sub> <sup>-1</sup>		
Local concentration of active ingredient in soil adjacent to treated surface	Clocal <sub>rinse,soil(a)</sub>	kg.kg <sub>wwt</sub> -1		

Model calculation

#### In the countryside

$$Clocal_{rinse,soil(d)} = \frac{\left(Elocal_{rinse,drift,façade} + Elocal_{rinse,drift,roof}\right)}{Vsoil(d) \times RHOsoil}$$
(28)

$$Clocal_{rinse,runoff,façade} + Elocal_{rinse,runoff,roof})$$

$$Vsoil(a) \times RHOsoil$$
(29)

## In a city

In a first approach, the resulting concentrations in soil and emissions to water after application and after rinsing can be added if the application and the rinse are performed the same day.

Note: If the rinse and the application are not performed the same day, for storm water, only the releases from the rinse need to be taken into account.

## 5.3 Release during service life

The releases assessment during service life is valid for both film preservatives and masonry preservatives used outdoors.

The releases during service life are potential emissions to the environment resulting only from leaching of a substance from treated substrates. The calculations proposed here do not take into account removal processes of the substance from the receiving compartment due to, for example, to degradation, volatilisation, leaching to ground water. Such removal processes are considered in the calculations proposed in Annex n°2.

The cumulative quantity of an active ingredient ( $Q_{leach,time}$ ), emitted to the relevant environmental compartment due to leaching from a treated substrate, within a certain period of service, is considered for assessment ( $kg.m^{-2}$ ).

The soil adjacent to the treated surface is the only compartment where a substance can accumulate. So, in this chapter, only the releases due to leaching of a substance from a treated house located in the countryside is taken into account.

The estimation of Q<sub>each,time</sub> should preferably be based on representative data from well-designed and standardised leaching tests. In this scenario, the contact medium is rainwater whereas the receiving environmental compartment is soil. The dimensions of this compartment are the same than the receiving compartment of runoff and dripping (Vsoil(a)).

A leaching test should be performed with similar philosophy as for wood preservatives (OECD, 2002).

 $Q_{\text{leach,time}}$  is calculated on the basis of this leaching test.

Two different time windows are considered:

- $Q_{leach,time1}$ : time1 = 30 days for an initial assessment, and
- $Q_{leach,time2}$ : time2 > 30 days for a longer assessment period

The 30 day cut-off is proposed in order to be coherent with a typical life cycle period of soil organisms in the effect assessment for PEC/PNEC determination (OECD, 2002). A longer assessment period is necessary for substances which accumulate in soil.

The releases during service of a house (cf. Table 18) result from a façade and a roof.

<u>Table 18</u>: Emission scenario for calculating the releases during service life of a house

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Leachable area of a façade proposed in the relevant scenarios	$AREA_{façade}$	m²	125	D
Leachable area of a roof proposed in the relevant scenarios	$AREA_{roof}$	m²	145	D
Duration of the initial assessment period	time1	d	30	D
Duration of the long-term assessment period	time2	d		D
Cumulative quantity of an active ingredient, leached over the initial assessment period	$Q_{leach,timel}$	kg.m <sup>-2</sup>		S
Cumulative quantity of an active ingredient, leached over a longer assessment period	Qleach,time2	kg.m <sup>-2</sup>		S
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	$m^3$	0.5	D
Bulk density of wet soil	RHOsoil	kg <sub>wwt</sub> .m <sup>-3</sup>	1 700	D
Output:				
Concentration in local soil at the end of the initial assessment period	$Clocal_{soil(a),leach,time1} \\$	kg.kg <sup>-1</sup>		
Concentration in local soil at the end of a longer assessment period	$Clocal_{soil(a),leach,time2}$	kg.kg <sup>-1</sup>		

Model calculation

$$Clocal_{soil(a),leach,time1} = \frac{Q_{leach,time1} \times \left(AREA_{façade} + AREA_{roof}\right)}{Vsoil(a) \times RHOsoil}$$
(31)

$$Clocal_{soil(a),leach,time2} = \frac{Q_{leach,time2} \times \left(AREA_{façade} + AREA_{roof}\right)}{Vsoil(a) \times RHOsoil}$$
(32)

# 5.4 Example of calculation

# 5.4.1 Releases during the treatment in situ

In these examples, only releases due to the treatment by sprayer are calculated. The calculation of releases due to treatment by roller or brush is considered to be similar.

In all calculation, it is considered that the product used for the treatment is the fourth product of the Table 4.

# 5.4.1.1 Treatment of a roof by sprayer

 $AREA_{roof} = 145 \text{ m}^2$ 

 $V form = 0.5 l.m^{-2}$ 

Fform = 0.01

RHOform =  $1000 \text{ kg.m}^{-3}$ 

Fdrift = 0.1

Frunoff = 0.2

 $Vsoil(d) = 54.1 \text{ m}^3$ 

 $Vsoil(a) = 0.5 \text{ m}^3$ 

RHOsoil =  $1700 \text{ kg}_{\text{wwt}} \cdot \text{m}^{-3}$ 

$$\begin{split} Elocal_{spray\_drift,roof} &= 145 \times 0.5 \times 0.01 \times 1000 \times 0.1 \times 10^{\text{-3}} = 0.0725 \text{ kg.d}^{\text{-1}} \\ Elocal_{runoff,roof} &= 145 \times 0.5 \times 0.01 \times 1000 \times 0.2 \times 10^{\text{-3}} = 0.145 \text{ kg.d}^{\text{-1}} \end{split}$$

#### In the countryside

Clocal<sub>spray,roof,soil(a)</sub> = 
$$0.0725 / (54.1 \times 1700) = 788 \mu g.kg_{wwt}^{-1}$$
  
Clocal<sub>spray,roof,soil(a)</sub> =  $0.145 / (0.5 \times 1700) = 170 \text{ mg.kg}_{wwt}^{-1}$ 

#### In a city

$$Elocal_{spray,roof,water} = 0.0725 + 0.145 = 0.2175 \text{ kg.d}^{-1}$$

## 5.4.1.2 Treatment of façade by sprayer

 $AREA_{roof} = 125 \text{ m}^2$ 

 $V form = 0.5 l.m^{-2}$ 

Fform = 0.01

 $RHOform = 1000 \text{ kg.m}^{-3}$ 

Fdrift = 0.1

Frunoff = 0.2

 $Vsoil(d) = 54.1 \text{ m}^3$ 

 $Vsoil(a) = 0.5 \text{ m}^3$ 

RHOsoil =  $1700 \text{ kg}_{\text{wwt}} \cdot \text{m}^{-3}$ 

$$\begin{split} Elocal_{spray\_drift,façade} &= 125 \times 0.5 \times 0.01 \times 1000 \times 0.1 \times 10^{\text{-3}} = 0.0625 \ kg.d^{\text{-1}} \\ Elocal_{runoff,façade} &= 125 \times 0.5 \times 0.01 \times 1000 \times 0.2 \times 10^{\text{-3}} = 0.125 \ kg.d^{\text{-1}} \end{split}$$

## In the countryside

$$\begin{aligned} & Clocal_{spray,façade,soil(d)} = 0.0625 \: / \: (54.1 \times 1700) = 679 \: \mu g. k g_{wwt}^{-1} \\ & Clocal_{spray,façade,soil(a)} = 0.125 \: / \: (0.5 \times 1700) = 147 \: mg. k g_{wwt}^{-1} \end{aligned}$$

#### In a city

$$Elocal_{spray,façade,water} = 0.0625 + 0.125 = 0.1875 \text{ kg.d}^{-1}$$

# 5.4.1.3 Treatment of a house by sprayer

## In the countryside

$$Clocal_{spray,soil(d)} = 788 + 679 = 1467 \mu g.kg_{wwt}^{-1}$$
  
 $Clocal_{spray,soil(a)} = 170 + 147 = 317 \text{ mg.kg}_{wwt}^{-1}$ 

# In a city

$$Elocal_{spray,water} = 0.2175 + 0.1875 = 0.405 \text{ kg.d}^{-1}$$

# 5.4.1.4 Releases during rinse of house after treatment by sprayer

Fdrift = 0.1

Frunoff = 0.2

Felim = 0

Frinse = 
$$1 - 0.1 - 0.2 - 0 = 0.7$$

 $AERA_{house} = 270 \text{ m}^2$ 

 $V form = 0.5 l.m^{-2}$ 

Fform = 0.01

RHOform =  $1000 \text{ kg.m}^{-3}$ 

 $Frunoff_{rinse} = 0.75$ 

 $Fdrift_{rinse} = 0.25$ 

 $Vsoil(d) = 54.1 \text{ m}^3$ 

 $Vsoil(a) = 0.5 \text{ m}^3$ 

RHOsoil =  $1700 \text{ kg}_{\text{wwt}} \cdot \text{m}^{-3}$ 

Elocal<sub>rinse,drift</sub> = 
$$270 \times 0.5 \times 0.01 \times 1000 \times 0.25 \times 0.7 \times 10^{-3} = 0.236 \text{ kg.d}^{-1}$$
  
Elocal<sub>rinse,runoff</sub> =  $270 \times 0.5 \times 0.01 \times 1000 \times 0.75 \times 0.7 \times 10^{-3} = 0.71 \text{ kg.d}^{-1}$ 

# In the countryside

$$Clocal_{rinse,soil(d)} = 0.236 / (54.1 \times 1700) = 2,57 \text{ mg. kg}_{wwt}^{-1}$$
  
 $Clocal_{rinse,soil(a)} = 0.71 / (0.5 \times 1700) = 835 \text{ mg. kg}_{wwt}^{-1}$ 

#### In a city

$$Elocal_{rinse,water} = 0.236 + 0.71 = 0.946 \text{ kg.d}^{-1}$$

# 5.4.2 Releases during service life

It is not possible to give an example for this scenario since no value for  $Q_{\text{leach, time1}}$  and  $Q_{\text{leach,time2}}$  is available.

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### 7 GLOSSARY

**Clinker:** Nodular manufactured product that is ground to produce cement powder.

**Portland cement:** A hydraulic cement produced by pulverizing clinker consisting essentially of hydraulic calcium silicates and usually containing one or more of the forms of calcium sulfate as an interground addition.

**Pozzolans:** siliceous and aluminous materials that possess little or no cementitious value in themselves but that will-in finely divided form and in the presence of moisture-chemically react with calcium hydroxide at ordinary temperatures to form compounds possessing cementitious properties (e.g. fly ash, silica fume, rice husk, volcanic ash).

#### ANNEX N°1: SPRAY DRIFT

Off target deposition or spray drift may be defined as airborne movement of particles of an applied agricultural or other chemical outside the intended target area, resulting in contamination of a non-target area, with the potential risk of injury or damage to humans, plants, animals, environment or property. (Craig *et al.*, 1998).

The drift<sup>1</sup> of pesticides caused by spraying during application has been known to be a problem for the environment. High fractions of pesticides can be transported through the air and deposited in neighbouring ecosystems. (FOCUS, 1997). This problem can also occur during application of biocidal products on building by spraying. But the only data and models available about spray drift concern pesticides used in agriculture. So, to assess releases, existing models are used.

All agricultural nozzles produce a range of droplet sizes known as the droplet size spectrum. Nozzle type, orifice size and spray pressure are equipment factors that affect the droplet size spectrum. The higher the operating pressure, the smaller the droplet. Conversely, low pressure produces large droplets that may bounce off the target (SDTF,1997 (a) (b)).

Table 19 shows a typical distribution of droplets sizes for flat-fan nozzle when spraying water at two different pressures (North Dakota State University, 1997).

<u>Table 19</u>: Droplet size range for flat-fan nozzle at 1.38 bars et 2.76 bars (North Dakota State University, 1997)

Size range	Percent of total volume			
(microns)	1.38 bar	2.76 bar		
0-21	0.1	0.4		
21-63	3	10.4		
63-105	10.7	20.1		
105-147	16.2	25.4		
147-210	36.7	35.3		
210-294	27.5	7.7		
over 294	5.8	0.7		

Higher pressures create fine droplets; 2.76 bars should be considered the maximum for conventional broadcast spraying.

Indeed, the higher the delivery rate is, the larger the droplets are. And the lower the delivery rate is, the smaller the droplets are. Indeed, nozzles with small orifice produce small drops while large nozzles produce larger drops. Table 20 shows the variation droplet size versus pressure and delivery rate according to the type of nozzles (Spraying Systems Co, 1995).

<sup>&</sup>lt;sup>1</sup> The drift is the movement of spray droplets beyond the target zone.

Table 20: Droplets size versus pressure and delivery rate (Spraying Systems Co, 1995)

Type of	0.7 bar		3 bar		7 bar	
spraying	Delivery	VMD*	Delivery	VMD*	<b>Delivery rate</b>	VMD*
	rate (l/min)	(microns)	rate (l/min)	(microns)	(l/min)	(microns)
Hydraulic	0.02	20	0.03	15		
atomizing spray	0.08	100	30	200	45	400
Thin spraying			0.1	110	0.2	110
	0.83	375	1.6	330	2.6	290
Hollow cone	0.19	360	0.38	300	0.61	200
spray	45	3 400	91	1 900	144	1 260
Flat spray	0.19	260	0.38	220	0.61	190
	18.9	4 300	38	2 500	60	1 400
Full cone spray	0.38	1 140	0.72	850	1.1	500
	45	4 300	87	2 800	132	1 720

<sup>\*</sup> VMD: Volume Median Diameter

The volume Median Diameter (VMD) is a particle size which is based on volumetric measurements. It is defined as the diameter at which half the spray volume is in droplets of larger diameter and the other half of the volume is in smaller droplets.

Droplet size was shown to be the most important factor affecting drift from application. A useful measure for evaluating drift potential is the percentage of spray volume consisting of droplets less than 141 microns in diameter. This value was selected because of the characteristics of the particle-measuring instrument, and because it is close to 150 microns which is commonly considered a point below which droplets are more prone to drift.

However, it is important to know that drift doesn't start and stop at 141 microns. Drift potential continually increases as droplets get smaller than 141 microns, and continually decreases as droplets get bigger (SDTF,1997 (a) (b))

The rate of deposition depends on the diameter of droplets. If the droplet diameter decreases, the velocity of descent will also decrease (cf. Table 21).

<u>Table 21</u>: Settling velocity of droplets (ASM 222, 2001; Spraying Systems Co, 1995)

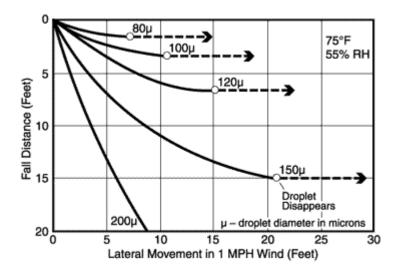
Diameter (microns)	Settling velocity (m.s <sup>-1</sup> )
5 000	3.6
2 000	3.4
1 000	2.7
500	1.9
300	1.21
240	0.93
200	0.73
140	0.44
100	0.26
40	0.049

10	0.003
2	$1.2 \times 10^{-4}$

Droplets less than 50 microns have insufficient momentum for impaction as they remain suspended in the air indefinitely or until they evaporate. (Landers, in press)

Figure 6 shows the trajectories of evaporating spray droplets falling through stable air at 25°C, having 55% relative humidity in a mile per hour crosswind. From this figure, it can be concluded that there is a rapid decrease in drift potential of droplets as they increase to about 150 or 200 microns.

Figure 6: Evaporation rate of water droplets



Nozzle spray angle has an effect on droplet size. Spray angle is the interior angle formed between the outer edges of the spray pattern from a single nozzle. Table 22 shows that nozzle with wider spray angle will produce smaller spray droplets than a nozzle with the same delivery rate but narrower spray angle (North Dakota State University, 1997).

<u>Table 22</u>: Effect of spray angle on droplet size with flat fan nozzles (North Dakota State University, 1997)

Spray angle (°)	Nozzle pressure (bars)				
	1.035	2.76	4.14		
	Volume Median Diameter in microns				
40	900	810	780		
65	600	550	530		
80	450	360	330		
110	390	310	290		

For example, equipment used can be a knapsack sprayer. One type of knapsack sprayer has a hand operated hydraulic pump that forces liquid pesticide through a hose and one or more nozzles. The pump is usually actived by moving a lever. A mechanical agitator plate may be attached to the pump

plunger. Some of these sprayers can generate pressures of approx. 7000 hPa. Capacity of these sprayers is usually approx. 20 L or less (University of Nebraska-Lincoln, 2001)

In addition to the spraying equipment, local meteorological conditions such as the wind speed, relative humidity and temperature also influence the amount of spray drift. To minimise transportation of pesticides via the atmosphere, wind velocity should be as small as possible to get more deposition within the sprayed area (FOCUS, 1997).

There are several factors that play a significant part in the creation and reduction of drift. A summary of the influences of various factors on spray drift is given in Table 23 (North Dakota State University, 1997).

<u>Table 23</u>: Summary of influences of various factors on spray drift (North Dakota State University, 1997)

Factor	More drift	Less drift
Spray particle size	Smaller	Larger
Release height	Higher	Lower
Wind speed	Higher	Lower
Spray pressure	Higher	Lower
Nozzle size	Smaller	Larger
Air temperature	Higher	Lower
Relative humidity	Lower	Higher
Nozzle type	Produce small droplets	Produce larger droplets
Air stabiliy	Vertically stable air	Vertical movement of air
Volatility	Volatile	Non volatile

The dominant processes responsible for drift of pesticides don't depend on pesticide properties but on climatic situations (wind speed, relative humidity in air, air temperature). Consequently, none of the models consider pesticide properties such as water solubility or vapour pressure (FOCUS, 1997).

The distance between nozzle and target (boom height) is also an important factor. Less distance between the droplet release point and the target will reduce spray drift. Less distance means less time to travel from nozzle to target and therefore less drift occurs. Small spray droplets have little inertial energy, so a short distance from nozzle to target increases the chance that the small droplets can reach the target (North Dakota State University, 1993).

In order to quantify the amounts of pesticides transported by spray drift, many studies have been performed in the last few years.

The Spray Drift Task Force (SDTF) is a consortium of 38 agricultural chemical companies established in 1990 in response to US-Environmental Protection Agency (EPA) spray drift data requirements. Data were generated to support the reregistration of approximately 2 000 existing products and the registration of future products from SDTF member companies. The studies were

designed and conducted in consultation with scientists at universities, research institutions, and the US-EPA (SDTF 1997 (a) (b)).

Based on data generated by the SDTF, in a typical orchard airblast application to a 1 200 feet wide grove of oranges, over 99% of the applied active ingredient stays on the crop and less than 0.5% drifts.

Although airblast applications are commonly made in orchards at least 1 200 feet wide, using an application area of this size was not practical. Instead, six row section of orchards were used in the SDTF studies. This design generated data representative of larger orchards because most drift originates from the outer rows.

Because the application area was smaller than for a typical orchard, and because most drift comes from the outer downwind rows, the percentage of active ingredient deposited on the ground downwind of the grove in the SDTF studies was approximately 4%, rather than 0.5%. This percentage of drift is artificially high due to the relative size and location of the application areas.

Even if the surface is not continuous, grove of oranges may be considered as similar surface to wall, roof ... So, the studies made in six row section of orchards may be used. Thus, the most interesting model for vertical surface is about application by spraying on orchard. So, 4% of the applied active ingredient which drifts may be considered as a worst case. (SDTF,1997 (a))

In the years 1988-1992, a number of drift tests have been carried out by the agrochemical industry in co-operation with the authorities for the notification of pesticides in Germany. The objective was to produce drift values to be used for approval of pesticides in Germany. The drift values resulting from the tests take into account specific situations: different crops (vineyard, orchards, hop, vegetable, field) and leaf stages (early or late). A conservative situation has been chosen on the basis of a 90<sup>th</sup> percentile. From all the situations studied, orchards and hops have at a late growth stage the most continuous surface (cf. Table 24). The used pressure ranged between 3 and 17 bars for orchards, and between 16 and 29 bars for hops. Care has to be taken to use these data for this ESD.

Table 24: German drift tables (Ganzelmeier et al., 1995)

Distance from the edge of trees (m)	Orchards (%)	Hops (%)
3	15.73	19.33
5	8.41	11.57
10	3.6	5.77
15	1.81	3.84
20	1.09	1.79
30	0.54	0.56
40	0.32	0.25
50	0.22	0.13

Based on data generated by the SDTF, in a typical full field ground hydraulic application, more than 99.9% of the applied active ingredient stays on the field and less than 0.1% drifts.

Although ground hydraulic applications typically consist of a 1200 feet ( $\approx$  366 m) wide and 1000 feet (305 m) long application area, using fields of this size was not practical. Instead 180 feet ( $\approx$  55 m) wide application area was used in the field studies. Because the application area was smaller than is typical for commercial applications, and because most drift comes from the outer area of the field, the percentage of the active ingredient leaving the field in the SDTF studies was slightly higher than the typical full field application, but was still only about 0.5%.

These studies were performed with flat fan nozzle at approx. 2 800 hPa pressure.

The field can be considered as horizontal surface (terrace, street, pavement, tennis court, pathway...). However, the surface treated would be much smaller than 55 m wide and 305 m long. So, 0.5% of the applied product which drifts is not a worst case. The amount of product which drifts would be higher.

(SDTF,1997 (b))

As nozzle size and pressure used in agriculture and in masonry are very different, the dimensions of the receiving compartments can be calculated with the following general equation.

The distance travelled through by drift can be calculated as:

$$S = \frac{U \times H}{V}$$

where S = Drift [m]

U = Wind speed [m.s<sup>-1</sup>]

H = Height of release [m]

V = Settling velocity [m.s<sup>-1</sup>]

Thanks to this equation, the dimensions of receiving compartment are calculated. (ASM 222, 2001)

#### ANNEX N°2: REMOVAL PROCESSES IN THE RECEIVING COMPARTMENT

In the scenarios described in chapter 5, the concentration in soil ignoring removal processes (degradation, volatilisation or leaching to groundwater) in the receiving compartments is calculated. In this Annex, the removal processes will be estimated according to EU TGD (1996) and taken into account in the estimation of the concentrations in the receiving compartments.

The emission into soil is based on a single emission during application (including the rinse) followed by an average leaching rate from treated substrates in service, the following model (cf. Table 26) could be used. The dimensions of building and the receiving soil according to the different scenarios are described in chapter 4.1.2.3.

Table 25: Total concentration of active ingredient lost during application and rinse

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Total concentration of active ingredient in soil distant to treated surface resulting from application (sprayer)		kg.kg <sub>wwt</sub> -1		O
Total concentration of active ingredient in soil adjacent to treated surface resulting from application (sprayer)	1 2/ 1/	kg.kg <sub>wwt</sub> -1		O
Total concentration of active ingredient in soil adjacent to treated surface resulting from application	, , ,	kg.kg <sub>wwt</sub> -1		O
Local concentration of active ingredient in soil distant to treated surface resulting from rinse	$Clocal_{rinse, soil(d)} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Local concentration of active ingredient in soil adjacent to treated surface resulting from rinse	$Clocal_{rinse, soil(a)} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>		О
Output:				
Total concentration of active ingredient in soil adjacent to treated surface resulting from application and rinse	**	kg.kg <sub>wwt</sub> <sup>-1</sup>		
Total concentration of active ingredient in soil distant to treated surface resulting from application and rinse	**	kg.kg <sub>wwt</sub> -1		

# Model calculation

# If the application is performed with sprayer

$$C_{\text{applic}, \text{soil(a)}} = C_{\text{spray}, \text{soil(a)}} + C_{\text{rinse}, \text{soil(a)}}$$

$$C_{\text{applic}, \text{soil}(d)} = C_{\text{spray}, \text{soil}(d)} + C_{\text{rinse}, \text{soil}(d)}$$

# If the application is performed with roller

$$C_{\text{applic}, \text{soil(a)}} = C_{\text{roll}, \text{soil(a)}} + C_{\text{rinse}, \text{soil(a)}}$$

$$C_{\text{applic}, \text{soil}(d)} = C_{\text{rinse}, \text{soil}(d)}$$

<u>Table 26</u>: Emission scenario for calculating the releases in soil taking into account removal processes

Variable/parameter	Symbol	Unit	Default	S/D/O/P
Input:				
Total concentration of active ingredient in soil adjacent to treated surface resulting from application and rinse	11 / //	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Total concentration of active ingredient in soil distant to treated surface resulting from application and rinse	11 / //	kg.kg <sub>wwt</sub> <sup>-1</sup>		O
Duration of the initial assessment period	time1	d	30	D
Duration of the long-term assessment period	time2	d		D
Cumulative quantity of an active ingredient, leached over the initial assessment period	Qleach,time1	kg.m <sup>-2</sup>		S
Cumulative quantity of an active ingredient, leached over a longer assessment period	Qleach,time2	kg.m <sup>-2</sup>		S
Leachable area proposed in the relevant scenarios	AREA	m²		D
Soil volume adjacent to surface treated (cf. § 4.1.2.3)	Vsoil(a)	$m^3$	0.5	D
Bulk density of wet soil	RHOsoil	kg <sub>wwt</sub> .m <sup>-3</sup>	1 700	D
Soil-water partitioning coefficient	$K_{soil\_water}$	$m^3.m^3$		D
First order rate constant for removal from soil	k	$d^{-1}$		D
Output:				
Average daily emission of active ingredient due to leaching over the initial assessment period	$E_{soil(a),leach,timel}$	kg.d <sup>-1</sup>		
Average daily emission of active ingredient due to leaching over a longer duration	$E_{soil(a),leach,time2} \\$	kg.d <sup>-1</sup>		
Time weighted concentration in local soil over the initial assessment period	$Clocal_{soil(x), time1} \\$	kg.kg <sub>wwt</sub> <sup>-1</sup>		
Time weighted concentration in local soil over a longer duration	$Clocal_{soil(x),time2}$	kg.kg <sub>wwt</sub> <sup>-1</sup>		
Average concentration in soil pore water over the initial assessment period	$Clocal_{pore(x),time1}$	kg.m <sup>-3</sup>		
Average concentration in soil pore water over a longer duration	$Clocal_{pore(x),time2}$	kg.m <sup>-3</sup>		

X represents a (soil adjacent to treated surface) or d (soil distant to treated surface)

#### Model calculation

$$E_{\textit{soil}(a),\textit{leach,time}1} = \frac{\textit{AREA} \times \textit{Q}_{\textit{leach,time}1}}{\textit{time}1}$$

$$E_{soil(a),leach,time2} = \frac{AREA \times Q_{leach,time2}}{time2}$$

$$Clocal_{soil(a),time1} = \frac{E_{soil(a),leach,time1}}{Vsoil(a) \times RHOsoil \times k} + \frac{1}{k \times time1} \left[ C_{applic,soil(a)} - \frac{E_{soil(a),leach,time1}}{Vsoil(a) \times RHOsoil \times k} \right] \times \left( 1 - e^{-timel \times k} \right)$$

$$Clocal_{soil(a),time2} = \frac{E_{soil(a),leach,time2}}{Vsoil(a) \times RHOsoil \times k} + \frac{1}{k \times time2} \left[ C_{applic,soil(a)} - \frac{E_{soil(a),leach,time2}}{Vsoil(a) \times RHOsoil \times k} \right] \times \left( 1 - e^{-time2 \times k} \right)$$

$$Clocal_{pore(a),time1} = \frac{Clocal_{soil(a),time1} \times RHOsoil}{K_{soil\_water}}$$

$$Clocal_{pore(a),time2} = \frac{Clocal_{soil(a),time2} \times RHOsoil}{K_{soil\_water}}$$

$$Clocal_{soil(d),time1} = \frac{C_{applic,soil(d)}}{k \times time1} \times \left(1 - e^{-time1 \times k}\right)$$

$$Clocal_{soil(d),time2} = \frac{C_{applic,soil(d)}}{k \times time2} \times \left(1 - e^{-time2 \times k}\right)$$

$$Clocal_{pore(d),time1} = \frac{Clocal_{soil(d),time1} \times RHOsoil}{K_{soil\_water}}$$

$$Clocal_{pore(d),time2} = \frac{Clocal_{soil(d),time2} \times RHOsoil}{K_{soil\_water}}$$

If for a given product, no in-situ treatment is foreseen, i.e. if only pre-treated building materials is used for the construction of building, only releases due to leaching from the materials are taken into consideration and  $C_{applic,soil(a)} = 0$  and  $C_{applic,soil(d)} = 0$ .

# ANNEX N°3: RESULTS OF THE SURVEY PERFORMED IN FRANCE AND IN THE UNITED KINGDOM

# **IN FRANCE:**

On 4 October 2001, a little survey was been performed. A multiple choice questionnaire sent to 59 companies (applicators, formulator and producer). Answer rate was 28.8% (cf. Table 27).

Table 27: Statistic of survey

	Send	Answer	%
Applicator	12	1	8
Formulator	28	8	28.6
Producer	19	8	42
Total	59	17	28.8

Table 28 presents answers results of survey.

### **IN THE UNITED KINGDOM:**

On April 2002, the same survey was been to 70 British companies. Answer rate was 27.1%. Table 29 presents answers results of survey.

# **CONCLUSION:**

Thus, these surveys show that in general:

- Application is done with sprayer or roller
- Pressure of sprayer is between 0 and 3 bars.
- Nozzle used is flat fan nozzle, full cone nozzle or hollow cone nozzle
- During spraying, the loss by drift is below to 10% and the loss by runoff is below to 20%
- There is sometimes rinse
- The application is done once at any season

<u>Table 28</u>: Results of survey performed in France

Product	Composition	Surfaces	Materials	Application	Pressure	Rinse	Loss	Frequency	Season	Customers/ Remarks
1	Formaldehyde (2,5 à 10%) Glyoxal (2,5 à 10%) Glutaral (2,5 à 10%)	Façade, roof, tennis - court	Concrete, cement, stone, mortar, baked clay, slate	Sprayer	Between 0 and 1 bar	No	< 10% by drift	0,5/an	Spring Summer	-
2	Alkylbenzyldimethyl ammonium chloride	Façade, roof, monument, building, road, tennis -court	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	Between 1 and 2 bars	No	-	< 1/an	Indifferent	Point P
3	Fongicide/algicide	Bricks stored outside during 1 month	Baked clay	1	-	-	-	-	-	-
4	Quaternary ammonium (1%)	Roof, terraces, tennis - court	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	-	No	-	2/an	Spring Autumn	-
5	Benzalkonium chloride (16%)	Façade	Concrete, cement, stone, mortar	Sprayer, roller	Between 0 and 1 bar	No	0	1/an	Indifferent	Pinault, Point p, Tout Faire
6	Benzalkonium chloride (6,4%)	Façade, terraces, balusters	Concrete, cement, stone, mortar, baked clay, slate	High pressure sprayer	-	No	-	1/an	Indifferent	Pinault, Point p, Tout Faire
7	Quaternary ammonium (25-50%) 2-octyl-2H-isothiazole-3-one (2,5-10%)	Façade, roof, monument, building, road, tennis-court	Concrete, cement, stone, baked clay, slate	Sprayer, roller	-	Yes/No	-	1-2/an	Spring Summer	-
8	Dodecylamine salicylate and lactate (0,5-5%)	All surface (cleaning to prepare surface before paint)	Paint, masonry, brick, plaster	-	-	-	-	-	-	-
9 (curative)	Benzyldiméthyl cocoammonium chloride Octylisothiazolone Calcium hypochloride	Façade, roof, monument, building, road, tennis-court	-	Portable sprayer, flat fan nozzle	Between 1 and 2 bars	No	1% by drift	1/an	Spring Summer Autumn	-
10 (préventive)	Fongicide	Façade, roof, monument, building, road, tennis -court	-	Portable sprayer, flat fan nozzle	Between 1 and 2 bars	No	1% by drift	1/an	Spring Summer Autumn	-

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Product	Composition	Surfaces	Materials	Application	Pressure	Rinse	Loss	Frequency	Season	Customers/ Remarks
11 (préventive)	Fongicide	Façade, roof, monument, building, road, tennis-court	-	Portable sprayer, flat fan nozzle	Between 1 and 2 bars	No	1% by drift	1/an	Spring Summer Autumn	-
12	Alkylbenzylammo nium (3,75%)	Façade, roof, monument, building, tennis -court, walls	Concrete, cement, stone, mortar, baked clay, slate, plaster, earthenware	Sprayer, roller, brush	Between 0 and 1 bar	No	< 1% by evaporation 10 à 20% by dripping	1-2/an	Spring Summer	Négoces de matériaux de construction, Gdes surfaces
13	Triazine derivatives and isothiazolone derivatives in solvent (1%)	Façade, roof, monument, building, tennis -court, walls	Concrete, cement, stone, mortar, baked clay, slate, plaster, earthenware	Sprayer, roller, brush	Between 0 and 1 bar	No	5% by evaporation 10 à 20% by dripping	1/an	Indifferent	Négoces de matériaux de construction, Gdes surfaces
14	Sodium hypochloride (C<10% of active chlorine)	Façade, roof, monument, building	Concrete, cement, stone, mortar, baked clay	Sprayer, roller	Between 0 and 1 bar	Yes, >5 bars	< 10% by evaporation < 10% by dripping < 10% by drift	1/an	Indifferent	No wind and 5°C <t<35°c< td=""></t<35°c<>
15	Didécyl-di-méthylammonium chloride (C<5%)	Façade, roof, monument, building	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	Between 0 and 1 bar	Yes, >5 bars	< 10% by evaporation < 10% by dripping < 10% by drift	1/an	Indifferent	No wind and 5°C <t<35°c< td=""></t<35°c<>
16	Diuron (C<15%)	Façade	Mortar	Add with water of mixing	-	No	0	1 for service life of mortar	Indifferent	-
17	1,6-dihydroxy -2,5-dioxahexane (6- 8%) Didécyldimethylammonium chloride (8-12%)	Façade, roof, monument, building, tennis-court, road	Concrete, cement, stone, mortar, baked clay, slate	Roller	-	No	5% by evaporation 20% by dripping 5% by drift	1-2/an	Spring Autumn	-

<u>Table 29</u>: Results of survey performed in the United Kingdom

Product	Composition	Surfaces	Materials	Application	Pressure	Rinse	Loss	Frequency	Season	Customers/ Remarks
1	2-phenyphenol (0.25%) Benzalkonium chloride (0.99%)	Building	Stone	Sprayer	-	Yes	10% by runoff 5% by spray drift	1	Indifferent	-
2	Permethrin (0.2%) Tri(hexylene glycol) biborate (3.3%)	Building	Stone	Sprayer	-	Yes	10% by runoff 5% by spray drift	1	Indifferent	-
3	Benzalkonium chloride (2%) Biphenyl-2-ol (1%)	Façade, roof, building	Concrete, cement, stone, mortar	Sprayer, roller	-	No	-	1	Indifferent	-
4	Alkyl dimethylbenzyl ammonium chloride (20%)	Façade, roof, monument, building	Concrete, cement, stone, mortar, baked clay	Sprayer, roller	Between 1 and 2 bars	No	< 0.1% by runoff < 0.1% by spray drift	1	1	Many
5	Benzalkonium chloride (1%)	Façade, roof, monument, building	Concrete, cement, stone, mortar, baked clay, slate	Roller	-	No	99% by evaporation	1	Spring, Summer, Autumn	Many
6	Benzalkonium chloride (5%)	Façade, roof	Concrete, cement	Sprayer, roller	Between 1 and 2 bars	Yes	2% by evaporation 5% by runoff 5% by spray drift	1	Indifferent	General public
7	Disodium octaborate tetrahydrate (10%) Benzalkonium (2%)	Façade, building	Concrete, cement, stone, mortar, baked clay	Sprayer	Between 0 and 1 bar	No	< 2% by runoff < 1 % by spray drift	1	Indifferent	-
8 (curative)	n alkyldimethylbenzylammonium (1%) 2-phenylphenol (0.25%)	Façade	Concrete, cement, stone	Roller	-	No	< 1% by runoff	1	Spring, summer	Various professional
9	IPBC (0.2%)	Building	Concrete, cement, stone, mortar	Sprayer	Between 0 and 1 bar	No	-	1	Indifferent	Many
10	Disodium octaborate tetrahydrate (10%)	Building	Concrete, cement, stone, mortar	Sprayer	Between 0 and 1 bar	No	-	1	Indifferent	Many
11	Benzalkonium chloride (0.4%) 2-phenyl phenol (0.1%)	Façade, building	Concrete, cement, stone, mortar	Roller	1	No	5% by runoff	Before repainting	Indifferent	Used for trade /professional use
12	Ammonium chloride (0-5%)	Façade, roof, monument, building, street, tennis -court	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	3 bars	No	5% by spray drift	1	Indifferent	-

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Product	Composition	Surfaces	Materials	Application	Pressure	Rinse	Loss	Frequency	Season	Customers/ Remarks
13	Ammonium bromide (2-7%)	Façade, roof, monument, building, street, tennis -court	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	3 bars	No	5% by spray drift	1	Indifferent	-
14	Alkylaryldimethyl benzalkonium chloride (5%)	Façade, roof, monument, building	Concrete, cement, stone, mortar, slate, timber	Sprayer, roller	Between 1 and 2 bars	Yes	< 5% by evaporation 20% by runoff 10% by spray drift	2	Spring, summer	-
15	Benzalkonium chloride (3,37%) 2 phenylphenol (0,85%)	Façade	Concrete, cement, mortar	Sprayer	Between 0 and 1 bar	No	10% by spray drift	1	Indifferent	-
16	Benzalkonium Chloride (3%)	Façade, roof, building, garden paths, patios	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	Between 0 and 3 bars	Yes	-	-	Indifferent	-
17	Benzalkonium Chloride (1,5%) Disodium octaborate (3%)	Façade, roof, building, garden paths, patios	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	Between 0 and 3 bars	Yes	-	-	Indifferent	-
18	Disodium octaborate (10%) Alkyaryl trimethyl ammonium chloride (2%)	Façade, roof, monument, building	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	Between 1 and 2 bars	No	10% by runoff	1	Spring	-
19	Quaternary ammonium compounds (< 1-2%)	Façade, roof, monument, building, street, tennis-court	Concrete, cement, stone, mortar, baked clay, slate	Sprayer, roller	Between 0 and 1 bar	No	-	1	Indifferent	-