Emerging substances in a landfill leachate

POTENTIAL RISKS AT A CLOSED LANDFILL IN THE NETHERLANDS

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Abstract

Risks from emerging contaminants from a closed municipal and industrial landfill in the municipality of Oss, the Netherlands, were studied. The landfill is located in a polder area close to the Meuse River. Both Meuse and polder water levels are managed. Previous studies at the *Oijense Bovendijk* landfill excluded emerging substances, possibly underestimating the risks of the present contamination. Using historical data about the local industries, the potentially present emerging substances were identified. The fate, behaviour and effects of these substance groups were used to determine the risk potential. The groups with the highest risk potential were identified to be antibiotics, hormones, NSAIDs, PFCs and PBDEs. The spreading of the contaminants was calculated and the groundwater is expected to exfiltrate to ditches. Temporal variation in water levels, due to extreme water events has little influence, but might cause an overestimation of the flow velocity. The contamination could result in a risk for the users of the landfill terrain, groundwater or surrounding surface waters. However, due to adsorption and biodegradation most contaminants will not have reached the surface waters. Exact information about the dispersal and risks of the contamination is currently unavailable as the fate and behaviour of emerging substances are not fully understood. Field sampling and further research is necessary to evaluate the risks at the *Oijense Bovendijk* landfill.

This research shows that depending on the history of a landfill there might be emerging substances present, which are not considered in the standard risk analysis of landfills in Dutch policies. These new risks should be taken into account while considering the public and environmental health of the surroundings of old landfills.

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1 Introduction

Groundwater is one of the most precious substances found in the subsurface and is the most extracted raw material. Worldwide 50% of drinking water and 20% of irrigation water comes from groundwater (Zektser & Everett, 2004). Groundwater should therefore be properly protected from both depletion and contamination. Groundwater contamination is often caused by diffuse sources such as agriculture or contaminated surface waters (Zektser & Everett, 2004). The most common point sources are spills, industrial areas, and landfills. The latter may contain a large variety of substances.

Besides the more common soil and groundwater contaminants, such as heavy metals and (chlorinated) hydrocarbons, so called emerging substances are detected in the environment (Lapworth et al., 2012). These substances are defined by the EU as: "… substances that have been detected in the environment, but which are currently not included in routine monitoring programmes at EU level and whose fate, behaviour and (eco)toxicological effects are not well understood." (NORMAN, 2015). Because proper information about these substances is often missing, it is difficult to set an intervention standard. Intervention standards are decided upon after a risk assessment based on the dose-response relationships first described by Paracelsus (1493-1541) in the 16th century. Nowadays, his famous statement is still often quoted: "All substances are poisons; there is none which is not a poison. The right dose differentiates a poison and a remedy" (in: Parasuraman, 2011). Due to the lack of a risk assessments for emerging substances, these might be left out of policy and monitoring programmes (Lapworth et al., 2012). However, not having a standard does not imply that a substance is not harmful.

Emerging substances are of growing concern for human and environmental health. Studies on emerging substances have primarily focussed on their effects on surface water and drinking water. In the 1990s, research started on hormonal disruption of fish in surface waters. Jobling et al. (1998) were the first to document widespread sexual endocrine disruption in wild fish consistent with exposure to hormonally active substances in surface waters in Great Britain. Hormones found in surface water mostly come from sewage water and are not removed by treatment (Mompelat et al., 2008). Hormones and other emerging substances have also been detected in drinking water in western countries (Mompelat et al., 2008). Mompelat et al. (2008) argue that the majority of studies on pharmaceutical products concern wastewater and its treatment, while the risks are higher for drinking water. Besides, some pharmaceutical products are released into the environment directly via human and animal excretion or by landfilling. Nowadays landfills in the developed world are designed to prevent groundwater contamination, but this is not the case at historical landfills and in developing countries (Lapworth et al., 2012).

In the 1990s, several studies reported detection of pharmaceuticals in groundwater near landfills. Holm et al. (1995) detected several sulphonamide antibiotics up to a distance of 150 meter downgradient of a landfill and urged for further research into the spreading and degradation of these products in groundwater. Barnes et al. (2004) studied emerging contaminants in groundwater near landfills of different ages and showed that higher concentrations are found closest to the landfill, but some persistent contaminants can be transported substantial distances. The fate and behaviour of substances in groundwater can differ severely from surface water as substances can adsorb to soil and anaerobic and dark conditions alter the decay (Kümmerer, 2008). Although not all processes are yet understood, Lapworth et al. (2012) concluded that landfills can be a source for emerging contaminants in groundwater for a long time.

Management policies on contamination from landfills in the Netherlands was developed in the 1980s. This policy led to a significant amount of studies and remediation operations. However, these studies were based on the common contaminants. Nowadays, municipalities are the competent authorities for closed landfills, and contamination control falls under the European Water Framework Directive. The municipality of Oss, in the province of *Noord Brabant* in the Netherlands is known for its rich industrial past. Large pharmaceutical and chemical factories were founded here. In the 1950s the commercial waste was disposed of at landfills around the city. The *Oijense Bovendijk* landfill (hereafter OBL) near Oss is suspected to have been used for a large variety of commercial waste (NIPA, 2013). Multiple studies have been conducted on possible contamination and risks at the OBL. In 1984 the first explorative study was conducted leading to more specific studies. The most recent one was conducted by NIPA in 2013 and did not show alarming concentrations of substances, except for arsenic. Besides, contamination was detected in the aquifer below the landfill (NIPA, 2013).

The municipality of Oss is concerned that emerging substances at the OBL have been overlooked in previous studies and might pose risks to users in its surroundings. With over 600 closed landfills in the province of *Noord Brabant* alone, newly identified risks could cause great concern to public and environmental health.

1.1 AIM & RESEARCH QUESTIONS

The aim of this research is to assess the risks posed by emerging substances in groundwater leaching from the *Oijense Bovendijk* landfill.

To achieve this aim, the study was formed around the central research question, namely:

What are the risks to humans and the environment of emerging substances leached from the closed Oijense Bovendijk landfill?

To answer the central research question the study is divided into four sub questions:

1. Which not previously analysed substances are potentially present at the *Oijense Bovendijk* landfill?
2. What are the behaviour, fate and effects of these substances?
3. How has the contamination spread and how was this influenced by varying water levels in the nearby Meuse River and the surrounding polder area?
4. Which contaminants can be present in contaminated media, corresponding to the exposure routes present in the area of the *Oijense Bovendijk* landfill?

The first two sub questions focus on the hazard, while the latter focus on the exposure. The hazard and exposure determine the risk of a contamination.

1.2 APPROACH

To assess the risks of emerging substances at the OBL the research was divided in four phases, of which an outline can be seen in Figure 1. In the first place the substances that might be present in the waste at the OBL are identified (phase 1 – waste characterisation). The fate, behaviour and effects of these substances are then described and ranked on their risk potential (phase 2 – substance properties). The spreading of the contaminants is determined by the use of a hydrological model (phase 3 – contaminant spreading). The outcomes of the model shows the exposure routes and these are linked to the potentially affected users in the surroundings of the OBL (phase 4 - exposure). To achieve this historical information on the OBL is combined with data on its surrounding and available scientific literature. As the phases have a strong temporal relation concluding remarks, including discussion points, are given at the end of each phase, serving as the input for the following phase.

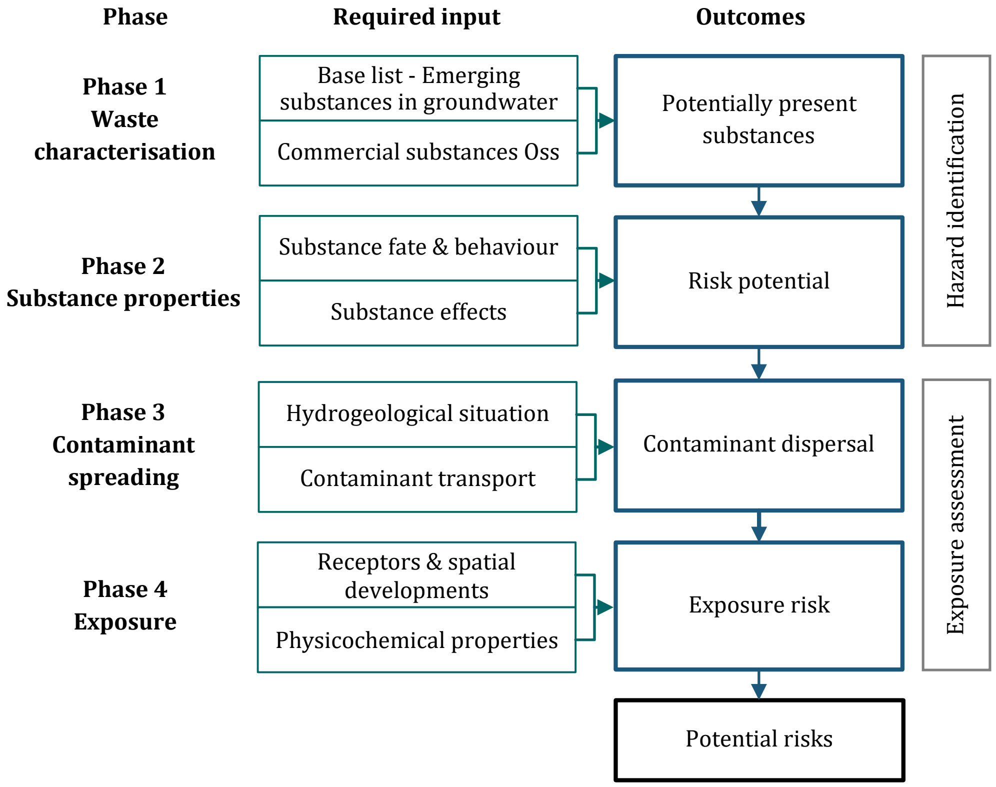


Figure 1 Research method phases, required input and outcomes.

The study into the risks at the OBL was carried out as a graduation internship at Tauw, a consultancy firm, commissioned by the municipality of Oss.

1.3 SITE DESCRIPTION

1.3.1 *Oijense Bovendijk* landfill

The *Oijense Bovendijk* landfill is located in the municipality of Oss in the Dutch province of *Noord-Brabant*, alongside the Meuse dike in a polder area. During the operation of the landfill it was owned by the municipality of Lith, but managed by a local business. The municipality of Lith merged with Oss in 2009, shifting the ownership of the landfill to the municipality of Oss along with the water board *Aa en Maas,* which owns a part of the terrain due to land reparcelling. On the southern part of the site a car business is run, which is left out of this study as an owner, but is taken into account as a possibly affected user.

The OBL was active from 1951 until 1961 and was used for both municipal and industrial waste. It is assumed in previous studies that about 30% of the waste came from industries (DHV, 1993).

The waste has been dumped in a low-lying swampy area (DHV, 1993). The landfill owners had a permit for filling up the marshes with waste (Gemeente Oss, 1951). The total landfill has an area of 2 hectares.

The packaging material of the waste is of importance for the availability of the substances to spread and pose risks. Iron drums can corrode under anaerobic conditions, but is relatively slow once the acidic phase is over (Kjeldsen et al., 2002). So, the amount of damage or decay of the packaging determines the spreading potential. The materials and packaging in the OBL have not been studied previously in the field.

1.3.2 Geology

Regional situation

The OBL is located on the *Peelhorst,* a block of geological sediments that has been lifted due to tectonical movements (Berendsen, 2004). The sediments in the *Peelhorst* are marine and river deposits that have eroded because of the upward movement. The top layer is made of young clays from river deposits. This covers the coarse sand deposits from the Pleistocene era which are on top of the fine loamy glauconite sands of marine deposits from the Tertiary era (Berendsen, 2004). This grouping, as can be seen in Table 1, has been used in previous studies by IWACO (2000).

Table 1 Regional geology as used in previous studies (IWACO, 2000).

Depth below NAP [m]	Geological formation	Lithological description
+5 - 0	*Betuwe*	(sandy) clay
0 - -50/-60	*Kreftenheije*	Course sands with gravel
	*Veghel**	Course sands with gravel
	*Tegelen**	Medium course to course sands, with loamy sands
	Oosterhout	Medium fine to medium course sands
> -50/-60	*Breda*	Fine loamy glauconite sands

*the formations of *Veghel* and *Tegelen* have been reallocated to the formations of respectively *Beegden* and *Waalre*

In the Regis II subsurface model, developed by the Dutch Organisation for Applied Scientific Research (TNO) thick clay layers are modelled in the sand pack. Near the OBL a first layer is found at 12 – 22 meters below NAP and ceases after about 2 km land inwards from the Meuse. A second layer is found between 37 and 42 meters below NAP, which ceases directly besides the landfill (Dinoloket, 2015).

The Meuse meandered through the area for centuries, leaving clay and gravel deposits behind. These deposits can be found in the aquifer. In Figure 2 it can be seen that the Meuse flowed in two channels close to the landfill about 4000 years ago. The stream belt Macharen consist of coarse silt and gravel containing sand and is found south of the landfill (Artesia, 2013). The more recent belt, which is currently fixed by dykes, also left behind deposits near the landfill (Cohen et al., 2009).

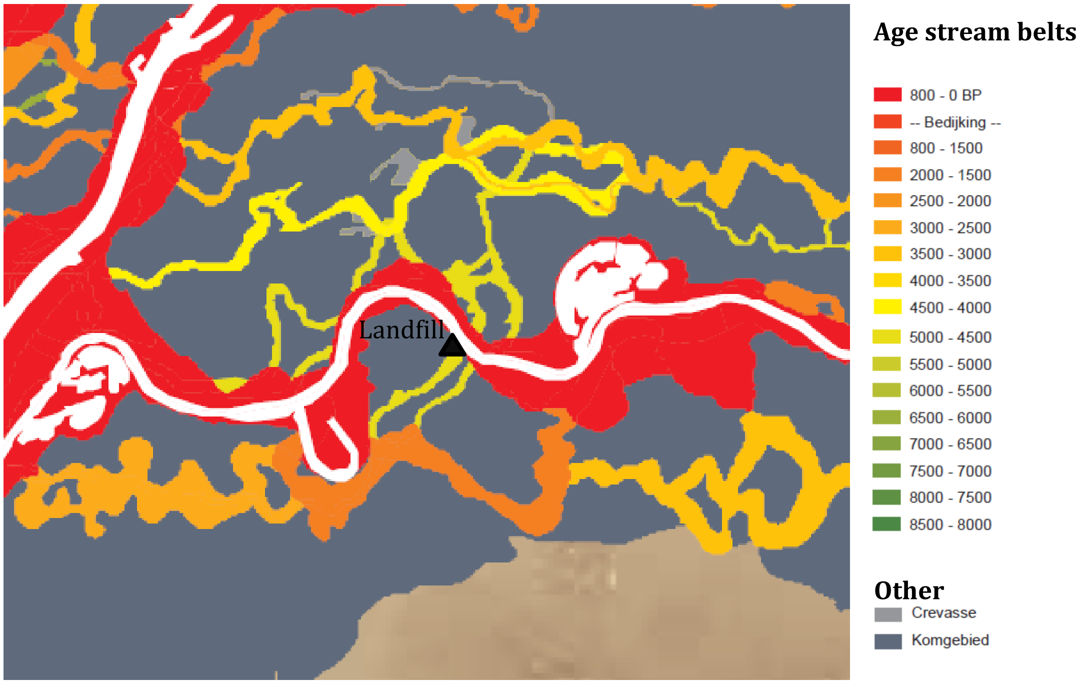


Figure 2 History of Meuse delta. Adapted from Cohen et al. (2009).

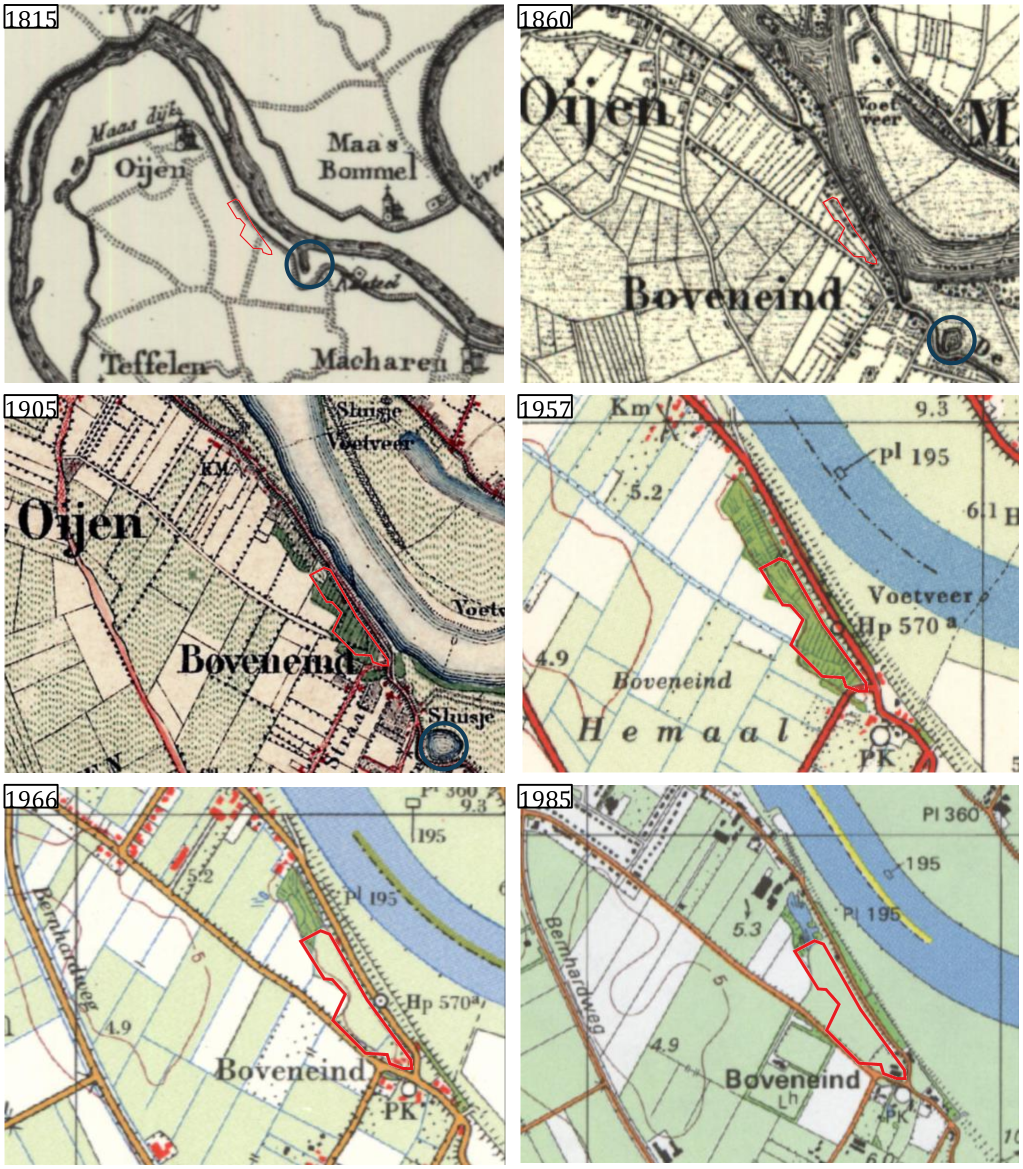
Local situation

The geological situation around the landfill has not been fully studied. Some drillings around the site have shown a clay layer of about 4 meter thick atop the first aquifer. The clay layer has been partly excavated and filled with waste materials (IWACO, 2000). Afterwards a covering layer of sandy clay has been applied, which is between 0.4 and 1.2 meters thick (NIPA, 2013). The ground surface is found at circa 5 meter above NAP (NIPA, 2013).

The bottom of the landfill is assumed to be at around 2 meters below surface level and located in the clay layer. In old studies it is assumed that an old pit from a dike breach is found on the site. Analysing old maps however shows the pit directly south of the landfill, see Figure 3. The site itself is mapped as a swampy area before the terrain was used as a landfill. The military map of 1830-1855 shows that the area was a swamp forest, a type of forest found in wet seepage areas. Besides, the environmental impact assessment done for the area around Lith indicates the area as an old pit where clay has been excavated to build and strengthen the dykes (Commissie MER, 2010).

The wetness of the terrain can be an indication of fully excavated clay, giving seepage the possibility to exfiltrate or can be explained by being a lower laying area on which runoff formed puddles before infiltration. When the clay would be fully excavated the waste is in direct contact with the aquifer. However, field experiments show (vertical) hydraulic conductivities in landfills to be in the 10^{-4} m/d range (Fleming, 2011). Vertical conductivities are about an order of magnitude less than horizontal conductivities (Fleming, 2011). These values are comparable with hydraulic conductivity values for clay, as shown in

Table 2, so for flow calculations no difference between those two scenarios will be made.



= location landfill

○ = location dike breach pit

Maps: 1815 Kraijenhoff, 1860 TMK 50D, 1905 Bonnenbladen. 1957-1985 TOP25

Figure 3 Historical development of the area at the OBL. Adapted from Kadaster (2015).

Table 2 Hydraulic conductivities (m/d) based on soil material (Dufour, 1998).

Material	Hydraulic conductivity [m/d]		
Coarse gravel	10^3	-	10^4
Fine gravel	10^2	-	10^3
Coarse sand	5	-	10^2
Fine sand	10^{-1}	-	5
Silt	10^{-2}	-	10^{-4}
Clay	10^{-4}	-	10^{-6}

In previous studies it has been shown that the water in the aquifer is contaminated (DHV, 1993). The depth and characteristics of the aquifer have not been studied.

Geochemistry

Clay layer

The properties of the clay layer in which the landfill is located have not been studied. However, the covering layer has been studied at 5 locations by NIPA (2013) and is probably made of local sediments, which makes the results comparable. The fraction organic matter is on average 3.2% of dry matter and the lutum fraction is 14% (NIPA, 2013).

Aquifer

The sediments of the aquifer have to been studied. The only available data is that the groundwater pH ranges from 6.2 till 7.3 around the landfill (NIPA, 2013).

1.3.3 Hydrology

The subsurface can be grouped in a top layer of poorly permeable clay on top of a sandy aquifer with a base layer of impermeable marine clays. The aquifer roughly ranges from NAP to 60 meters below and the general flow direction of the groundwater is in south-western direction (IWACO, 2000). There is no second aquifer, as the base clay formation has a thickness of about 200 meters (Berendsen, 2004). However, as in the surroundings of the OBL a clay layer is found in the aquifer at circa 12 m below NAP this separates the aquifer locally in an upper aquifer of about 14 meters thick and a thicker lower aquifer, as can be seen in Figure 4.

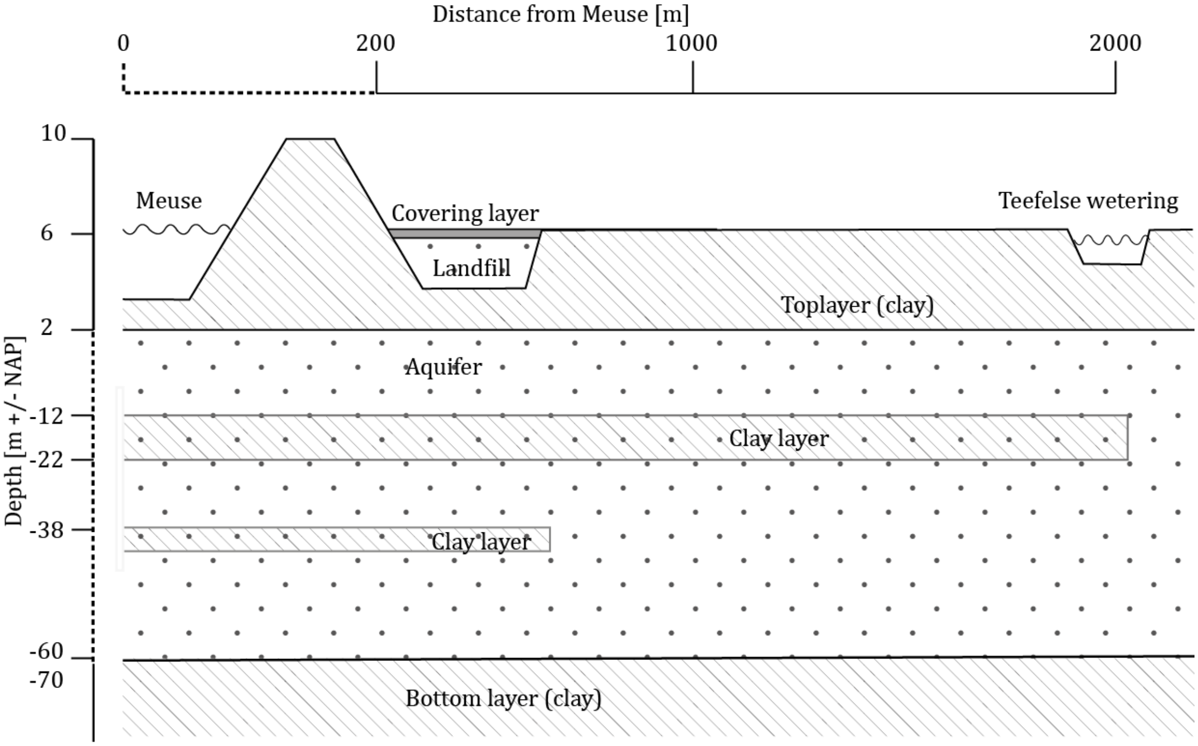


Figure 4 Schematic overview of the position of landfill in its hydrogeological surroundings.

The water level in the polder in which the landfill is located is controlled by the water board, see Figure 5. The levels differ in allocated compartments. Surplus water is drained by the *Teefelse Wetering,* which flows through the *Hertogswetering* back to the Meuse, see Figure 6.

The groundwater level strongly depends on the water level of the Meuse, which is located at a distance of circa 150 meters from the landfill. In a natural situation the groundwater would shortcut the meander of the river, due to the gradient in surface water. However, the Meuse has a weir downstream of the OBL since 1936, leading to a smaller gradient and a relatively constant water level.

Because the average Meuse level is above the average groundwater level inlands in the polder the flow is perpendicular to the Meuse, see Figure 7. The higher level also causes seepage in the polder area (IWACO, 2000).

Both the water levels of the Meuse and the polder are managed and have varied over time. Extremes in Meuse river runoff take place on a small timescale, causing a temporal change in groundwater flow, see Figure 8.

At 1-2 kilometre distance from the landfill the pumping field of drinking water station Macharen is located. Since the pumping started in 1950, it influenced the flow direction and velocity and the groundwater levels in the area (IWACO, 2000). The drinking water company calculated the capture zone with a numerical MODFLOW model (Artesia, 2013). The capture zone does not include the landfill as can be seen in Figure 9. In 2017 the pumping station will be terminated (Brabant Water, 2014). This leads to a smaller gradient between the water levels of the Meuse and the polder. At the OBL there will be no significant influence as the water board keeps the groundwater level in the polder as constant as possible.

At the northern side of the landfill a pond is present, which can cause contaminated groundwater to flow into the pond as this route might have a smaller resistance. From the pond the contaminants can infiltrate again or spread in the surface water.

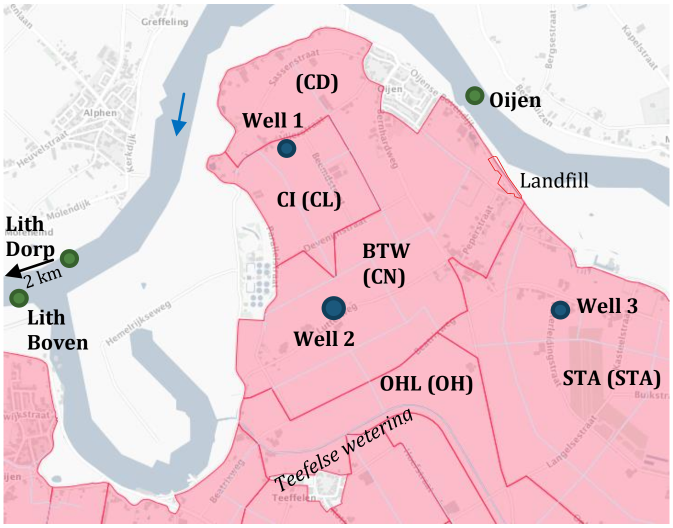


Figure 5 Monitoring and management of surface and groundwater in the surrounding of the OBL. Surface water management compartments in the OBL area since 1998. Including old compartment names (..). Well 1 = B39G0281, well 2 = B45E0433, well 3 = B45E0442. Source compartments: Waterschap Aa en Maas (2015), wells: Dinoloket (2016), Meuse: Rijkswaterstaat (2015).

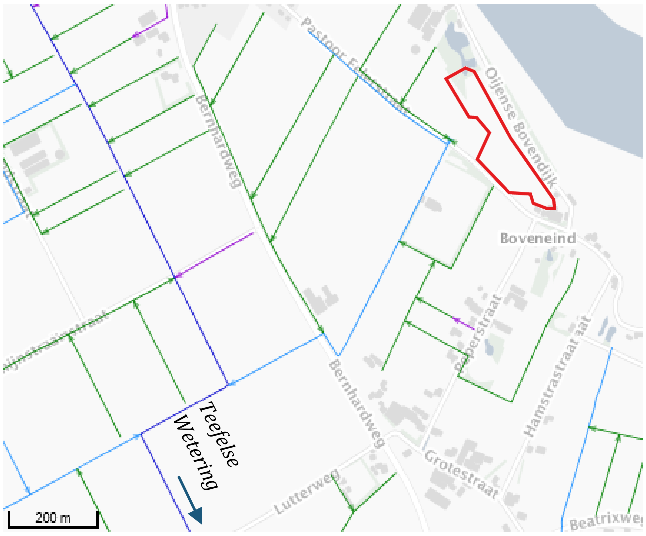


Figure 6 Waterways in the surroundings of the OBL (Waterschap Aa en Maas, 2016).

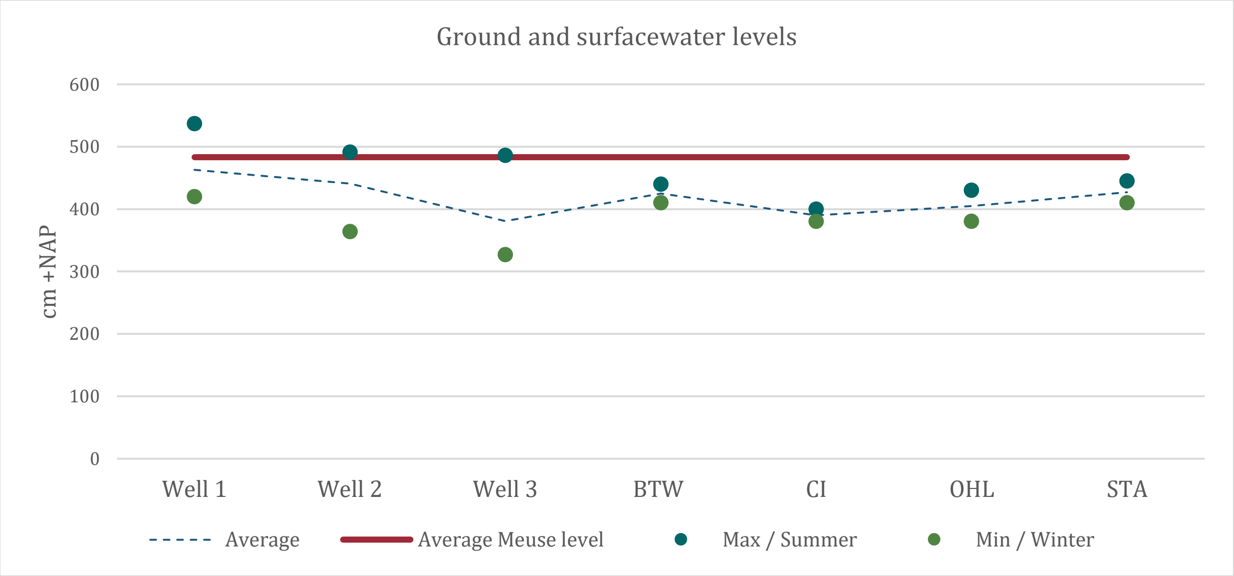


Figure 7 Average, maximum and minimum groundwater level 1989-2000 in monitoring wells and average summer, winter and annual surface water level 1950-1998 in management compartments around the OBL in cm above NAP. Data monitoring wells: Dinoloket (2016) and surface water compartments: IWACO (2000).

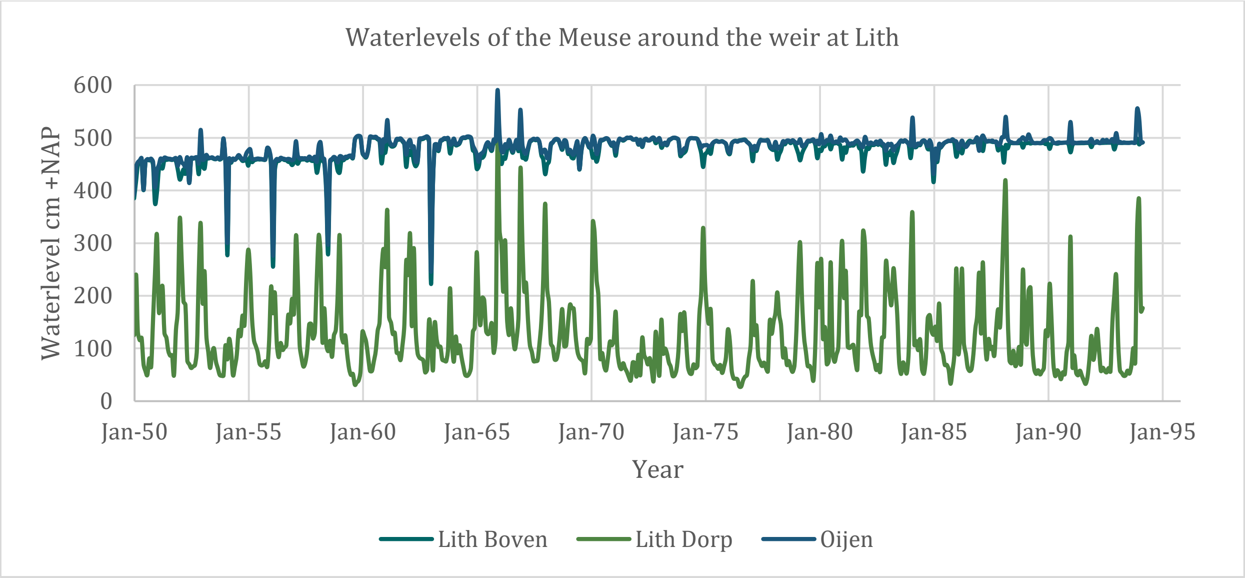


Figure 8 Water level (monthly averages) of the Meuse in the surroundings of Lith. Measurement points Lith Boven and Oijen are upstream of the weir, Lith Dorp downstream. Data: Rijkswaterstaat (2015).

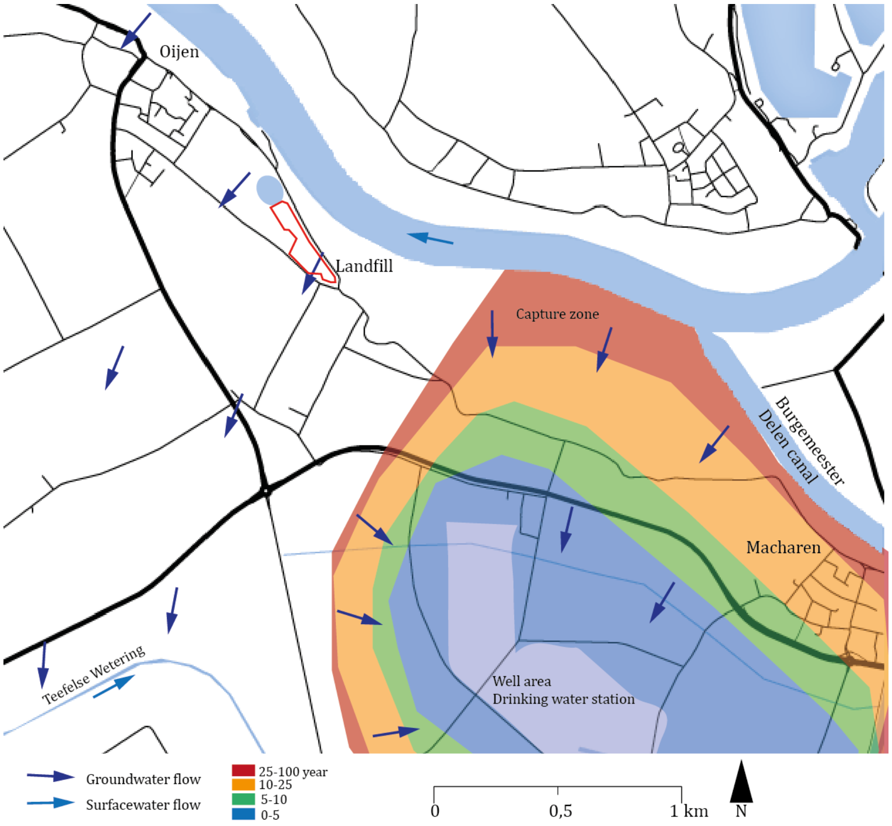


Figure 9 Area around the OBL with general flow directions and recharge area, divided into residence times, of the pumping station. Source recharge area: Artesia (2013).

2 Theoretical Background

2.1 EMERGING SUBSTANCES

As stated in the introduction, emerging substances are substances that have been detected in the environment, but are currently not included in routine monitoring programmes and their toxicological properties are not fully understood (NORMAN, 2015). Modern measurement and monitoring techniques are able to detect previously undetected contaminants, for example organic micro-contaminants (Stuart et al., 2012). The improving detection possibilities show that emerging substances are not necessarily new in the environment and that in the future, with improving analysis techniques even more emerging substances may be found. Substances that were already known but their presence and significance are being elucidated only recently, are also emerging substances (Stuart et al., 2012).

This research focusses on all substances that have not been measured before at the OBL, especially from commercial wastes. It looks into the risk potential of all these new substances, which might not all be considered to be emerging substances in the literature.

2.2 RISK ASSESSMENT

In 1992, an action plan for the management of chemicals was agreed upon at the United Nations Conference on Environment and Development (UNCED) in Rio de Janeiro. This plan focussed on the need to expand and accelerate the international assessment of chemical risks. Arising from this, the European Union (EU) introduced a program, called REACH, which concentrates on the improvement of the protection from risks posed by chemicals. This EU legislation makes companies responsible for the safety of chemicals they place on the market (Hansen, 2007). As a result, all new chemicals are tested before entering the environment. These test are based on OECD test guidelines and EU test methods and focus on four factors, namely; physical-chemical, human health, environmental fate and ecotoxicity. Both a hazard assessment and exposure assessment are done to define the risk characterisation.

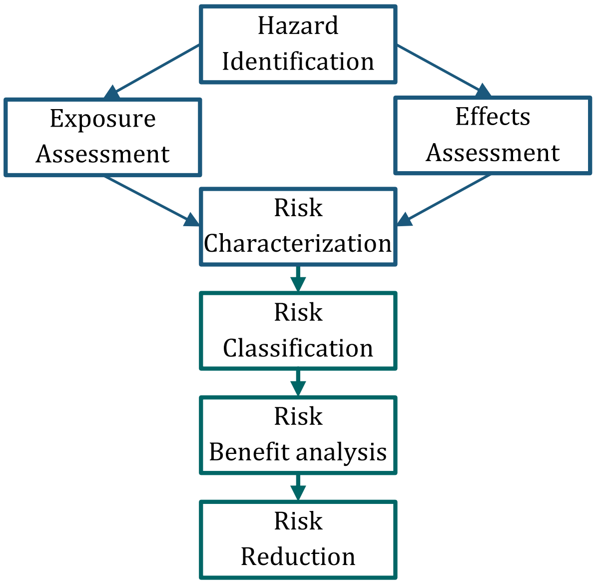


Figure 10 Risk Assessment. Adapted from van Leeuwen & Vermeire (2012).

The OECD and WHO also use risk assessment based on this principle and their process begins with problem formulation, followed by four steps: 1) hazard identification, 2) exposure assessment, 3) effects assessment and 4) risk characterization. Hazard identification focusses on the effects that a substance inherently has on human and environmental health. Exposure assessment looks at the actual or expected exposure to a certain substance. As substances might enter the environment through different pathways that are time and location depended, the exposure assessment is an uncertain part of the risk assessment process (van Leeuwen & Vermeire, 2012). The exposure assessment and effect assessment (step 2 and 3) generate the scientific baseline for a valid risk assessment (Hansen, 2007). The risk characterization may lead to risk classification, benefit analysis, and reduction steps (Figure 10). Risk assessments can take many different forms, depending mainly on the scope and purpose, the available data and resources (van Leeuwen & Vermeire, 2012).

As emerging substance intrinsically miss information on their risks, for every emerging substance that researchers come across a risk assessment should be done. In practice, this is a time consuming and costly operation.

2.3 CONTAMINANT FATE AND BEHAVIOUR

The fate and behaviour of substances in the environment are of utter importance to the toxicity of the substance to humans or to the ecology (Arnold et al., 2014). All physical, chemical and biological processes linked to movement and transformation of a chemical after entering the environment are part of the environmental fate of a substance (Gruiz et al., 2015). Industrial substances and pesticides are often neutral molecules, while pharmaceutical substances are large, complex molecules that are often charged. This difference strongly influences the fate and behaviour, as further explained below.

Contamination from landfill waste can end up in leachate through three mechanisms; hydrolysis and biological degradation of solid waste, dissolution of soluble salts and transport of particulate matter (Christensen et al., 1992). Processes important for both transport and exposure of contaminants are advection, adsorption and degradation. Other processes important for exposure are volatility, bioaccumulation and plant uptake (Halling-Sorensen et al., 1998). All processes are described below in more detail.

2.3.1 Advection

Advection is the transport of substances due to groundwater flow and is key to both entering the leachate from the landfill and to transport in the groundwater. Substances can either dissolve into the groundwater or be transported with the groundwater flow (Christensen et al., 1992).

Dissolution

Solubility depends on the chemical properties of the substance itself and environmental parameters, such as temperature and chemical composition of the water. Substances dissolve until fully dissolved or the solvent is saturated, after which precipitation will occur (Berkowitz et al., 2014). Ionic substances, like soluble salts, split into ions while dissolving. This is a reversible reaction called dissociating. When detecting dissociated compounds, free ions, in groundwater it is often impossible to trace these back to the parent material (Berkowitz et al., 2014).

Organic substances, like pesticides, dissolve depending on their polarity. In general polar (hydrophilic) substances dissolve in polar liquids like water and nonpolar (lipophilic) substances in nonpolar liquids like turpentine (Berkowitz et al., 2014). Substances are often considered soluble when their solubility exceeds 0.1 g/100ml (or >1000 mg/l) (Bradley et al., 2014). However in literature trace concentrations of micrograms per litre of pharmaceuticals are shown to have adverse effects (Kümmerer, 2008). Dutch drinking water standards are 1 μg/l for unknown substances, a factor 7 smaller than 'insoluble' substances (Schriks et al., 2010).

Pharmaceuticals are generally an acid or base, and only a small amount (5-10%) is neutral or hydrophobic (Brooks & Huggett, 2012). As many pharmaceuticals have several ionising groups the solubility strongly depends on the pH (Kümmerer, 2008). But as pharmaceuticals intend to have effect in the human body most are produced as salts to increase solubility. However, because of the complex structures the molecules can bind together in polymorphs or pseudopolymorphs, all having different solubility's. In an experiment with chemically identical, but different binding forms of the antibiotic rifampicin, solubility differed with a factor 8 in identical water samples (Kümmerer, 2008).

Non-dissolving substances

Substances in the subsurface can enter the groundwater without dissolving, whenever the particles can be transported by water flow. Due to weathering particles can develop, which can be transported by the groundwater flow (Christensen et al., 1992).

Non-dissolving liquid substances can easily pollute the soil, as they penetrate and disperse within the soil without difficulty, transporting themself to the groundwater. So-called Non-Aqueous Phase Liquids (NAPLs) do not mix with water, but can either float on top (Light NAPL) or sink below (Dense NAPL) groundwater (Fetter, 1999). NAPLs can be transported by the groundwater flow (Berkowitz et al., 2014).

2.3.2 Adsorption

Dissolved substances in the groundwater can adsorb to the sediment. Hydrophobicity is the main factor determining the adsorption of neutral substances to organic matter. The most important adsorption materials are clay minerals, oxides and humic acid. The partitioning between the water and adsorbed phase is measured by the K_{oc} value, which is in combination with the fraction of organic content of the sediment used in transport calculations (Berkowitz et al., 2014). Substances with a K_{oc} below 1000 are considered unlikely to adsorb, with increasing likelihood to adsorb with increasing K_{oc}. From K_{oc} above 10.000 a substance is likely to adsorb (Bradley et al., 2014).

Pharmaceutical substances are mostly non-hydrophobic. But, because they are ionising substances they can adsorb through cation exchange and the formation of surface complexes and hydrogen bonds (Kümmerer, 2008). This complicates adsorption models, as pH, fraction of metal oxides, ionic strength and cation exchange capacity of the soil are key factors. Although studies into sorption of pharmaceuticals are conducted and the first models are being developed, these are not yet broadly applicable (Brooks & Huggett, 2012). For reliable adsorption data a pharmaceutical substance has to be measured in the specific soil (Brooks & Huggett, 2012).

2.3.3 Degradation

The properties of both organic and inorganic substances are affected by biological activity or water properties. Degradation of substances is caused by biodegradation, chemical degradation and photo-degradation.

Biodegradation depends on the chemical structure and the molecular mass of the substance, as microorganisms mineralise the substances (Gruiz et al., 2015). The biological degradation process of landfill waste consists of both aerobic and anaerobic phases.

Firstly, during the operation time of the landfill aerobic decomposition takes place due to the presence of oxygen. Little leachate is produced at this time (Christensen et al., 1992). Secondly, with decreasing oxygen, methane production starts. However, aerobic process can continue as air diffuses into the landfill, creating aerobic zones.

Anaerobic degradation starts with acid-fermentation. Microorganisms hydrolyse cellulose, carbohydrates, fats and proteins into soluble organic compounds. These compounds are transformed to fatty acids and alcohols and later in acetic acids, carbon dioxide and hydrogen. The volatile fatty acids and high CO_2 concentrations cause the pH to drop between 5 and 6, which causes other waste materials to be mobilised, such as heavy metals. Simultaneously, easily soluble substances in the waste and those made available by degradation cause the leaching of anions and cations. Anaerobic bacteria continue the breakdown of materials and lower the redox potential, which is essential to methanogenic bacteria that need a high biochemical oxygen demand (BOD) and organic carbon concentrations (Christensen et al., 1992; Stuart & Klinck, 1998).

These methanogenic bacteria cause an increase in methane gas, lowering hydrogen, carbon dioxide and volatile fatty acid concentrations and an increase in pH. This causes a decrease in the solubility of heavy metals and other compounds, but the waste stabilisation and leachate production continues for decades (Christensen et al., 1992).

The solubility and susceptibility to redox reactions of acids and bases is influenced by the pH of the water, while simultaneously the pH is influenced by the substances that dissolve. Redox reactions take place in both aerobe and anaerobe circumstances. Oxygen plays a role in oxidation of substances that might enhance biotransformation (Berkowitz et al., 2014). Groundwater is generally anaerobic, while surface waters are aerobic. Besides, substances in groundwater are not exposed to light, which is necessary for photodegradation. This causes all forms of degradation to be generally slower or missing in groundwater, leading to more persistence and the ability to transport over larger distances.

Very little literature is available on the degradation and degradation products of pharmaceuticals in groundwater or sediments. Available literature is often based on lab-experiments and outcomes of these experiments are often contradictive between different studies (Fent et al., 2006). Walters et al. (2010) conducted field studies to establish the half-life of pharmaceuticals and personal care products in sediments, which resulted in a range between 180 and 3500 days. Substances with a half-life in soil above 30 days are considered moderately persistent, and above 100 days highly persistent. For water and air substances with a half-life above 40 days are considered highly persistent (Bradley et al., 2014).

Although persistence of substances in the environment is a key factor in the toxicity, its importance is only of relative interest when a substance has a constant input into the environment. For example, substances entering surface water from waste water have a quite constant input, so even if these degrade, the concentration in surface water does not decline and a substance is so-called pseudo-persistent (Kümmerer, 2008).

2.3.4 Volatility

The volatility of a chemical is the extent to which a chemical enters the air-phase through vaporisation. This is directly related to a chemical's vapour pressure, which is the maximum concentration that can be found in air, increasing with temperature. Substances with a vapour pressure above 1.3 Pascals are considered to be likely to volatilise (Bradley et al., 2014). The concentrations of a substance in the air and fluid-phase are considered to be in equilibrium, of which the ratio depends on the vapour pressure, the presence of other chemicals, temperature and gas pressure (Fetter, 1999).

Waste degradation in landfills causes the production of gases, containing mostly methane (±50-60%) and carbon dioxide (±40%). Other gases detected are traces of volatile organic compounds (VOCs), which can be either produced due to biological degradation or were present in the waste (Zou et al., 2003). Landfills are covered with a soil layer, preventing most exposure to VOCs. However, VOCs can diffuse into ambient air and travel in air and soil water due to advective transport (Hodgson et al., 1992). VOCs contribute for less than 1% of landfill gases and include hydrocarbons, organic alcohols, halogenated and sulphur compounds (Zou et al., 2003). Experimental studies show that landfill gases can migrate in the subsurface to basements of closely located houses and cause indoor exposure to elevated concentrations of contaminants (Hodgson et al., 1992). Humans can be exposed to toxic volatilising chemicals through inhalation.

2.3.5 Bioaccumulation

Bioaccumulation is the accumulation of a substance in an organism, caused by faster uptake of a substance than the reduction due to catabolism and excretion. Bioaccumulation and persistence are often used as inherent properties of a substances in hazard identification (van Leeuwen & Vermeire, 2012). The bioaccumulation potential is determined from the octanol-water distribution coefficient (D_{ow}). This coefficient is the ratio of the equilibrium concentration of a substance in octanol and water. As octanol is used as a surrogate for lipids, this ratio is used as an indicator for the partitioning into lipids or fats. The ratio is also used for sorption to sediments, biomass and sludge, leading to an indication for the distribution of a substance among several environmental compartments (Kümmerer, 2008). However, these relationship are not applicable to pharmaceutical compounds, whenever these are ionised in the aquatic environment, for which the octanol-water partitioning coefficient (K_{ow}) is used (Kümmerer, 2008). The bioaccumulation factor (BAF), which is the ratio between the concentration of a chemical in an organism and in its environment, is determined using regression equations. Substances with a K_{ow} above 1000 are considered likely to bioaccumulate (Bradley et al., 2014). However, the used regression equations are built up from limited data sets, so they are not applicable for all substances (Meylan et al., 1999).

2.3.6 Plant uptake

Uptake of substances by plants is a process possibly leading to bioaccumulation. Factors influencing bioaccumulation in plants are chemical, plant, soil and atmosphere properties (van Leeuwen & Vermeire, 2012). Plants can take up contaminants via (soil)water or air. Uptake into roots from the soil water happens through passive transport due to concentration gradients. The equilibrium ratio between roots and water ($K_{root\text{-}water}$) depends on the fraction of water, air and lipids in the root and the octanol-air and octanol-water partitioning coefficients of the chemical (van Leeuwen & Vermeire, 2012). An empirical relation is developed to predict the bioaccumulation factor in roots, based on the log K_{ow}, as chemicals with a higher log K_{ow} tend to sorb to the roots, leading to higher BAFs (Briggs et al., 1987). From the roots contaminants can be transported to the foliage, as a result of transpiration and accompanying pressure differences. Foliage can also take up contaminants from the atmosphere, through its stomata. This process is regulated by the equilibrium partitioning ratio between the foliage and the surrounding air ($K_{foliage-air}$) and is highly dependent on the air temperature, as stomata regulate gas exchange processes and transpiration, preventing a plant to dry out (Briggs et al., 1987).

2.4 TOXICITY

The toxicity of a substance depends largely on the physical and chemical behaviour. There are three types of toxicity, namely; chemical, biological and physical. Chemical toxicants are substances that react with living creatures. Biological toxicants are bacteria and viruses. And physical toxicants interfere with processes in living creatures by their presence, as for example asbestos particles (Casarett & Doull, 2013).

Toxic reactions can be either acute, normally caused by high exposure concentrations in a short time or chronic, caused by long-term exposure to lower concentration. An often used measure for toxicity are the predicted or derived no effect levels (PNELs or DNELs). The incidence and severity of the effects likely to occur due to the actual or predicted exposure are often expressed as the PEC/PNEC ratio (Predicted Environmental Concentration / Predicted No Effect Concentration). Also the LC50 or EC50, the test concentration at which fifty per cent of an organisms is expected to die (Lethal Concentration) or have a certain adverse effect (Effect Concentration).

Special attention is needed for mixture effect of toxicants, as in real-life humans are simultaneously exposed to several substances. Chemicals can both strengthen or weaken each other. Interaction effects can therefore provoke variation from the expected values in dose levels, routes, timing and duration of exposure. Three basic types of action for combinations of chemicals are defined (Risks et al., 2011):

- Similar action
- Dissimilar/independent action
- Interactions

For mixtures of substances with similar action the effects can be directly estimated from the sum of the doses or concentrations of the individual substances. The sum should be scaled for the relative toxicity of the substances. For mixtures of independently acting substances the effects can be estimated from the responses of the individual substances or the sum of the effects. For mixtures of substances that interact it is not possible to estimate the effects directly (Risks et al., 2011).

The Stockholm Convention on Persistent Organic Pollutants determines the hazard of a chemical based on its biodegradability, bioaccumulation potential and toxicity to aquatic organisms (van Leeuwen & Vermeire, 2012). The latter is assessed by testing the sensitivity of three trophic levels of aquatic organisms, namely fish, Daphnia (commonly called water fleas) and algae (van Leeuwen & Vermeire, 2012). On fish and Daphnia the acute toxicity are tested, determining the LC50 and EC50 relatively. On algae the growth inhibition is tested determining the IC50, the concentration expected to cause 50% inhibition of growth or growth rate. The LC/EC/IC50 value (in mg l^{-1}) for the most sensitive organisms is used to appoint a toxicity category. LC/EC/IC50 <1 is considered as very high toxicity; 1–10 as high toxicity; 10–100 as moderate toxicity; and >100 as low toxicity (van Leeuwen & Vermeire, 2012).

2.5 SOLUTE TRANSPORT

Solutes in porous media are transported by several processes, which are described below in their relation to the transport calculations. All information is retained from Fetter (1999) unless indicated otherwise.

2.5.1 Advection

Advection is the transport of dissolved substances by the movement of its host-fluid, in this case the groundwater. The velocity and direction of groundwater flow depend on the hydraulic gradient and transmissivity of the soil. Advection can be calculated with:

$$x = \frac{q}{n} t = vt \qquad [\ 1\ ]$$

Where x = distance from source, q = flow velocity, n = porosity, t = time and v = real velocity.

2.5.2 Hydrodynamic dispersion

Hydrodynamic dispersion causes a substance to spread within the medium and consists of two processes; diffusion and mechanical dispersion.

Diffusion is based on the gradient of concentration of dissolved substances and takes place regardless of flow. Mechanical dispersion is caused by variances in pore size and flow velocity on the micro scale. Aquifers are normally non-homogeneous, with layers of different grain sizes and small clay layers, causing also macro dispersion in the field. Mechanical dispersion causes the subtle transition of concentrations at the contamination front and is a three-dimensional process, even when the flow direction is only one-dimensional. From empirical studies by Neuman & Zhang (1990) it has been determined that longitudinal dispersion (α_L) per length (L) is approximately:

$$\alpha_L(L < 100m) = 0.0175L^{1.46} \qquad [\ 2\ ]$$

$$\alpha_L(L > 100m) = 0.32L^{0.83}$$

Transversal dispersion is about 5 till 10% of the longitudinal dispersion.

The hydrodynamic dispersion coefficient used in solute transport calculations is:

$$D_L = D_{eff} + \alpha_L v \qquad [\ 3\ ]$$

However, whenever flow is present, diffusion is often negligible, as is shown in below example using the Peclet number. This number compares the diffusion (or dispersion) and advection.

$$Pe = \frac{v x_R}{D} \qquad [\ 4\ ]$$

The diffusion coefficient (D_{eff}) in porous media is about 10^{-9} m^2/s (≈10^{-4} m^2/day). Groundwater velocity in aquifers varies between 1 cm and 1 m per day (v). Over a distance of 10 meters (x_R), the Peclet number would range between 10.000 and 100.000, as can be seen below. A large Peclet number means that advection is significantly more important than diffusion.

$$Pe = \frac{0.01.10}{10^{-4}} \approx 1000 \qquad Pe = \frac{1.10}{10^{-4}} \approx 100000$$

In clay layers where flow velocities are very low, diffusion is more important for solute transport.

So when flow is present the diffusion is negligible and the hydrodynamic dispersion coefficient is:

$$D_L \approx \alpha_L v \tag{5}$$

The peak of the substance travels with the average flow velocity, and therefore the formula [1] stays valid for the peak concentration.

2.5.3 Retardation

Retardation is the delay in the transport of substances in the groundwater. It is mainly caused by adsorption and leads to lower reactivity, bioavailability and decay.

The ratio of dissolved and adsorbed substances is expressed in the distribution coefficient K_d. At the front of the contamination plume dissolved substance adsorb until the ratio is met, while at the tail of the plume adsorbed material desorb. These processes cause the plume to move slower that the groundwater flow velocity, which is called retardation. The retardation coefficient is:

$$R = 1 + \frac{(1-n)\rho^s}{n} K_d \tag{6}$$

Hydrophobic organic solutes such as pesticides, dissolved petroleum products and cleaning agents are mainly prone to hydrophobic interaction with organic carbon in the soil. A larger organic carbon content of the soil therefore leads to a larger $K_{d,}$ which can in these cases be assumed to be proportional to the fraction of organic carbon (f_{oc}): $K_d = f_{oc}K_{oc}$. The value of $K_{oc,}$ the organic carbon partition coefficient, is considered to be a constant for a given substance. The formula is not valid for small values of f_{oc}, the critical level of f_{oc} differs for each sorption material. Values normally used for f_{oc} range from 0.001-0.0005 for sands (Weiner, 2012).

The rate of adsorption is not solely dependent on the concentration of the substance, as the adsorption capacity of the soil is limited. Hence, the linear approximation might overestimate the amount of adsorbed material and is not usable in all cases. Both Freundlich and Langmuir determined relations that take the limited adsorption capacity into account, changing equation [6].

2.5.4 Decay

Decay of substances can take place in both the dissolved and adsorbed state. However, decay of adsorbed substances is often neglected as rates are normally very low. Decay depends on the substance properties and factors in its environment, like temperature and pH and is indicated with the half-life value.

2.5.5 Overall transport equations

When all above processes take place the following governing equation is valid.

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - \frac{q}{n}\frac{\partial C}{\partial x} - \mu C \tag{7}$$

The case of the landfill can be described as a semi-infinite column of porous media with decay when x > 0, so after entering the groundwater from the landfill.

Initial conditions:
C = 0 x ≥ 0

Boundary conditions:
$C = C_0$ x = 0
C = 0 x = ∞

The analytical solution in this case is:

$$C(x,t) = \frac{1}{2} C_0 \exp\left(\frac{vx}{2D}\right)\left[\exp(-x\beta)\ \mathrm{erfc}\left(\frac{x-(v^2+4\mu D)^{\frac{1}{2}}\,{}^{t}/_{R}}{\sqrt{4Dt/R}}\right)+\ \exp(x\beta)\ \mathrm{erfc}\left(\frac{x+(v^2+4\mu D)^{\frac{1}{2}}\,{}^{t}/_{R}}{\sqrt{4Dt/R}}\right)\right] \quad [\,8\,]$$

Where $\beta = \sqrt{(v/2D)^2 + \mu/D}$

This formula is used to determine the concentration (C) on a certain distance from the source (x), after a certain time (t) considering advection (v), dispersion (D), retardation (R) and decay (μ).

3 Methods

3.1 PHASE 1 WASTE CHARACTERISATION

In phase 1 an answer is given to the first sub question:

Which not previously analysed substances are potentially present at the Oijense Bovendijk landfill?

The aim of this sub question is to identify substances likely to have been disposed that have not been measured at the OBL before.

Firstly, a literature study was conducted into the emerging substances commonly found in groundwater in Europe. These substances were checked to fit the time and location of the OBL. Secondly, research was conducted into the companies and industries that were active in Oss in the 1950s and their products and production processes. Lastly, the substances already analysed by NIPA (2013) were excluded, as this is the most recent and broadest study conducted at the OBL.

This led to a list of substances that were not only available during the operation time of the site, but also used in closely located facilities. The process can be seen in Figure 11.

Detailed methods and used data per step are described below.

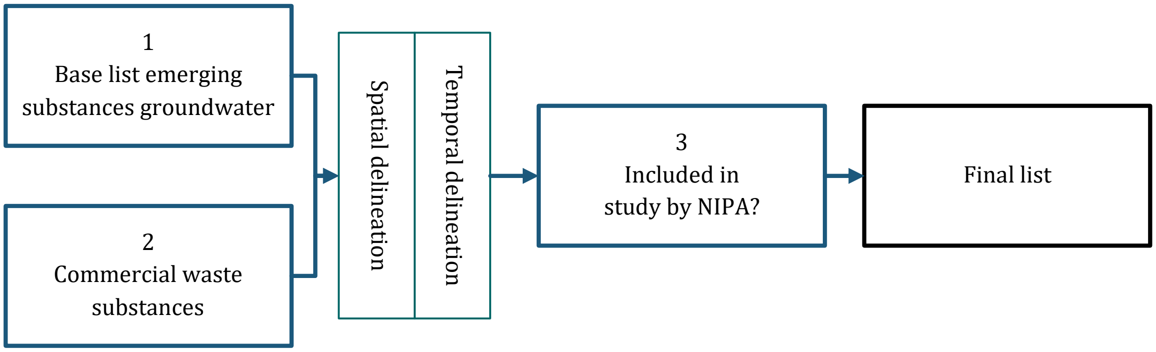


Figure 11 Outline of methods used in phase 1.

3.1.1 Step 1 Base list of emerging substances in groundwater

In the first step a base list of emerging substances that have been found in groundwater was prepared that was used to give guidance in the next steps and to ensure no substances were overlooked. The list was delineated for the time and area relevant for the OBL case.

Klein & Duijnhoven (2013) conducted a substance screening of 80 substances in the groundwater in the Meuse river basin in the Netherlands and came across 14 emerging substances above the reporting limit. Although this study spatially corresponds with the OBL case, a more extensive list was considered necessary, as using the list of Klein & Duijnhoven (2013) might cause the unintended exclusion of substances. In France a broad screening of emerging substances in the groundwater was conducted at 494 sites by Lopez et al. (2015) and of the 411 substances in the screening 180 substances are detected (above the detection limit). Even though the French situation might differ from the Dutch, this study is expected to cover the most important substances found in Dutch groundwater because of the broad spectrum of substances considered. Moreover, the French study used varying limits of quantification between 0.005 and 10000 ng/l and not the reporting limit as in the Klein & Duijnhoven study, which they set to be 0.1 μg/l for all substances except 9, for which an intervention value was available, ranging between 0.00025 and 440 μg/l (Klein & Duijnhoven, 2013; Lopez et al., 2015).

Temporal delineation

The list of substances found in groundwater was cross-referenced with the time of operation of the OBL. The landfill was in operation from 1951-1961 (Bodemloket, 2015). To identify the time in which the substances were developed and used, the medical publications database of PubMed was used. This database is managed by the *US National Center for Biotechnology Information* from the *National Institute of Health* and contains a large number of both medical and chemical literature (NCBI, 2015). The oldest records about a substance in the database indicates the time it was firstly produced or researched. This method leaves room for errors as not all substances are studied scientifically at the moment they are developed and the development may last several years. To cope with this only substances with a first record after 1970 were excluded from further research.

Spatial delineation

It is checked whether the substances found in groundwater were used or produced around Oss. Information on the companies and industries which were active in Oss is found in historical information and information from nuisance acts permits issued by the municipality. All industries in Oss were taken into consideration, as commercial waste was collected throughout the city and disposed of at the OBL.

3.1.2 Step 2 Commercial waste

In this step, research into the industries and associated companies that were active around Oss in the 1950s was conducted. Consequently, no further spatial and temporal delineation was necessary in this step. The products and processes of the industries were analysed for products and used substances. Nieuwkoop (1993) performed a study into the contaminations that can be expected from the industries in the 19th and 20th century in the province of *Noord Brabant*, based on their production processes. The study of Nieuwkoop (1993) formed the basis of the analysis, and was expanded and checked with literature and archive information, as emerging contaminants might be excluded due to the age of the research. The British Department of Environment (DoE) (1995a, 1995b, 1995c) composed lists of contaminants detected from different industries, which were used to check the study by Nieuwkoop (1993).

3.1.3 Step 3 Previous studies

In this step the substances that were measured in previous studies at the OBL were excluded. The few detected substances in these measurements were not considered to pose risks at the found concentrations. This research therefore focusses on the substances that were not measured before. It has to be noted that the substances detected in previous studies might be harmful considering mix-effects, but this is beyond the scope of this research.

In 2013 NIPA measured field samples at the OBL based on the TerrAttesT Spectrum Sheet 7.22. The study included metals, radiation, halogenated hydrocarbons, phthalates and a large variety of pesticides.

3.2 PHASE 2 SUBSTANCE PROPERTIES

In phase 2 an answer is given to the second sub question:

What are the behaviour, fate and effects of these substances?

The aim of this sub question is to determine the risk potential of the substances that are potentially present at the OBL. The risk potential is seen as the extent to which a substance could cause adverse effects as a contaminant. This was done by looking into the fate, behaviour and effects on humans and the environment.

Because little data was available on the properties of emerging substances the substances were divided into groups, based on their purpose. The purpose of substances, especially pharmaceutical substances, is an important parameter for the mode of action of their effects in the environment and human body.

A literature study was carried out to identify the fate, behaviour and effects of the substances. Factors describing the fate and behaviour of contaminants are adsorption and degradation, as described in the theoretical background. However, leachate composition is influenced by the waste composition, pH, redox potential and the landfill age (Christensen et al., 1992). As these factors are unknown, except from the age, it was not possible to give informed conclusions on the leachate production in this research. Therefore, all substances were considered to be possibly found in the leachate. Adsorption is mostly important for substance transport, but not for the risk potential in itself. Information on adsorption is therefore used later in the exposure parts of this research.

The risk potential was assessed using a matrix including the qualitative data availability, persistence and possible effects. Data availability was classified based on recent literature reviews on the current state of knowledge to gain the best available knowledge and findings. The work of Fent et al. (2006), who analysed the current knowledge on the ecotoxicology of pharmaceuticals and Kümmerer (2008) who edited a book on pharmaceuticals in the environment were used as resources on the data availability. The persistence of substances was based on literature research. Whenever no data of persistence in groundwater from field studies was available, laboratory and surface water studies were used to give some insight in the persistence. The toxicity of substances was also taken from literature whenever available, looking into the harmfulness of the substances to humans and the environment. A large variety of effects within a substance group was qualified as uncertain.

Because of the limited data on substance effects only three scores were given (+, +/- and -).

+	= Affirmative	(data available / persistent / harmful),
+/-	= Uncertain	(some data / some degradation / some substances harmful),
-	= Negative	(no data / full degradation / not harmful).

Substances with mostly ‘+’ have the highest risk potential, while ‘+/-’ and ‘-’ respectively have little or no risk potential.

3.3 PHASE 3 CONTAMINANT SPREADING

In phase 3 an answer is given to the third sub question:

How has the contamination spread and how was this influenced by varying water levels in the nearby Meuse River and the surrounding polder area?

The aim of this sub question is to get insight into the spreading of the contamination in the surroundings of the OBL. This was achieved by using a hydrological model to determine the groundwater flow and using this in combination with substance transport calculations to determine the contaminant spreading.

3.3.1 Groundwater flow model

The Meuse River is the driving factor of the local groundwater flow, as surface water levels in the polder are artificially kept below Meuse water levels, as described in 1.3. The importance of extremes runoff events and the variety in management levels was researched with the help of the hydrological model.

Model description

The FLOWNET model software was used to calculate and visualise the groundwater flow. This model calculates flow patterns and groundwater travel times based on static hydraulic heads. The hydraulic head distribution in the polder was calculated with the Dupuit approximation with 2 constant heads and a leaky aquifer (Hendriks, 2010). A representation of the hydrogeological situation is shown in Figure 12.

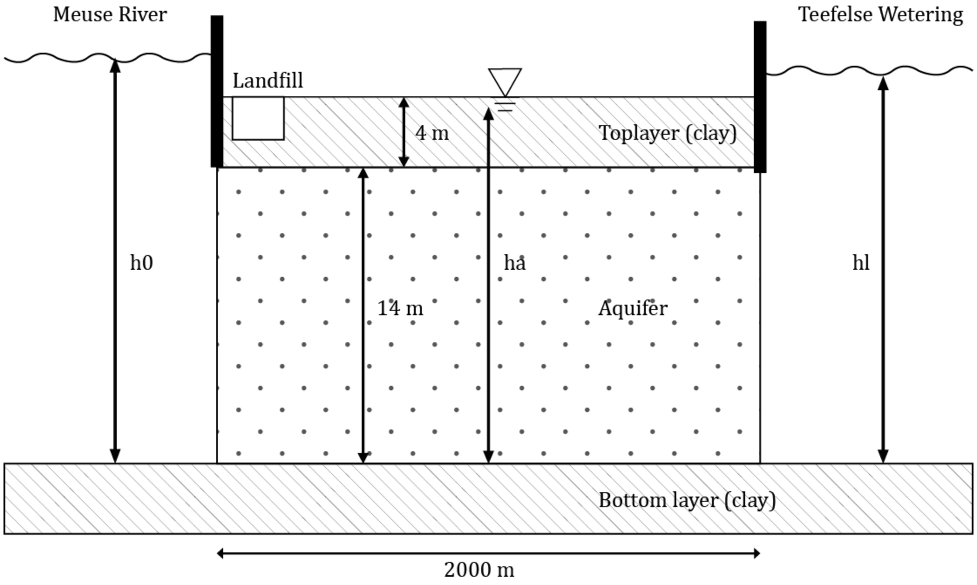


Figure 12 Conceptual representation of the hydrological situation in the surroundings of the OBL.

The general solution to the Dupuit approximation is:

$$h = h_a + C1e^{x/\lambda} + C2e^{-x/\lambda} \qquad [\,9\,]$$

where:
h = hydraulic head [m]
h_a = head artificial in polder [m]
x = distance from river [m]
λ = leakage factor [m] = $\sqrt{KD(c)}$

As the boundary conditions are:
at x = 0, h = h_0
and at x = L, h = h_l

$$C_1 = \frac{(h_l - h_a) - (h_0 - h_a)e^{-L/\lambda}}{e^{L/\lambda} - e^{-L/\lambda}} \qquad [\ 10\ ]$$

$$C_2 = h_0 - h_a - C_1 \qquad [\ 11\ ]$$

where:
h_0 = water level Meuse river [m]
h_l = water level *Teefelse wetering* [m]
L = distance Meuse - *Teefelse wetering* [m]

The extreme runoff periods were identified by taking the surface water levels deviating at least the standard deviation from the average value. The hydraulic head distribution of the extreme events was calculated in the same manner using data for polder and surface water levels from the corresponding time period. The model input can be found in Annex 4 – Groundwater flow model input.

Streamlines for normal circumstances and extreme events were calculated with the FLOWNET model, with time steps visualised every 10 years using isochrones. The program uses static heads, so the temporal variances are described by interpreting the outcomes of the model.

Input data

The general groundwater flow is influenced by the river and surface water levels. The period from 1951, when the landfill opened, until present was divided in 6 time periods, based on differences in the management of water levels and seasons. The Meuse level was altered in 1959, but kept constant throughout the year. The surface water levels in the polder have seasonal variances, and in 1998 the levels were changed (IWACO, 2000; Waterschap Aa en Maas, 2014). The water board uses summer water levels from May-September (5 months) and winter levels from October-April (7 months). This lead to six time periods, namely:

1.	Summer	1951-1959
2.	Winter	1951-1959
3.	Summer	1960-1998
4.	Winter	1960-1998
5.	Summer	1998-present
6.	Winter	1998-present

Hydraulic head distribution

For the Meuse water level data was available for the period 1950-1994, while monitoring data from the surface water levels were not available. For the Meuse the actual seasonal averages were used up to 1994, after which the management level was used (Rijkswaterstaat, 2015). For the surfaces water levels the management levels were used (Waterschap Aa en Maas, 2014). The surface water compartments used were from the BTW compartment for the artificial polder-levels and the OHL compartment for the *Teefelse Wetering*, see Figure 5.

The water level data for the different time periods is summarised in Table 3.

Table 3 Water level data (m) of the different time periods used for calculation of the hydraulic head distribution. Levels are relative to the bottom of the model at NAP -12m.

	Summer 1951-1959	**Winter 1951-1959**	**Summer 1960-1998**	**Winter 1960-1998**	**Summer 1998-present**	**Winter 1998-present**
h_0	16.56	16.6	16.92	16.89	16.9	16.9
h_a	16.4	16.1	16.4	16.1	16.2	15.8
h_l	16.3	15.8	16.3	15.8	16.3	15.8

The average Meuse water level between 1980 and 1994 is NAP +4.65m, with a standard deviation of 0.23m. Extreme water events were considered for all periods in which 4.65 ± 0.23m was exceeded and lasted for at least 2 days. The events are shown in Annex 3 – Extreme surface water level events, and 38 events of low water and 7 high water events took place. Of these, 5 events were chosen to represent a range of the most extreme and moderate events considering the number of days and water levels, which are shown in Table 4.

Table 4 Extreme low and high surface water events in the Meuse at Oijen between 1980 and 1995. Data: Rijkswaterstaat, 2015.

Low water	**Month**	**Nr. of days**	**Extreme level (NAP +cm)**	**Average level (NAP +cm)**	**Time period**
7	Aug-'82	4	445	448	Summer 1960-1998
15	Jan-'85	7	47	191	Winter 1960-1998
16	Mar-'85	29	445	450	Winter 1960-1998
38	Feb-'95	10	377	411	Winter 1960-1998
High water					
7	Jan-'95	12	679	613	Winter 1960-1998

As K = 36 m/d (Artesia, 2013), D = 13 m (Dinoloket, 2015) and c = 90d (Artesia, 2013), the leakage factor (λ) = 205m. Full input data for the time periods and extreme events, including the hydraulic head distribution is found in Annex 4 – Groundwater flow model input.

Groundwater flow model

The general model input is summarised in Table 5.

Table 5 Constant groundwater flow model input.

Parameter	Input	Unit	Source
Number of columns	20		
Number of rows	10		
Model length (m)	2000	m	
Model height (m)	20	m	
upper side	open		
left side	closed		
right side	closed		
bottom side	closed		
Clay layers			
Horizontal hydraulic conductivity	0.1	m/d	*
Vertical hydraulic conductivity	0.04	m/d	Artesia, 2013
Porosity	0.6		Artesia, 2013
Sand layer			
Horizontal hydraulic conductivity	36	m/d	Artesia, 2013
Vertical hydraulic conductivity	33	m/d	*
Porosity	0.3		Artesia, 2013

*Hydraulic conductivities were calculated using the block scale anisotropy (E[Kvh]/E[Kvv]) for fluvial medium to coarse sand of 1.1 and for fine and loamy sand of 3.7 (Bierkens, 1994).

3.3.2 Solute transport

To get insight in the spreading and therefore risks of the contaminants in groundwater, the groundwater flow modelling was combined with substance transport calculations. Transport in porous media is prone to the several processes described in 2.5. Formula [8] was used to perform all calculations.

The velocity of groundwater flow depends on the transmissivity of the aquifer, which is a combination of the Hydraulic Conductivity k [m/d] and the thickness of the aquifer D [m]. The hydraulic conductivity depends on the porosity and sediment type and is therefore vertically heterogeneous. The hydraulic conductivity was taken as 36 m/d (Artesia, 2013). The flow velocity (v) was calculated with the hydraulic conductivity (k), gradient (i) and porosity (n).

$$v = k \, . \, i \, / \, n \qquad [\,12\,]$$

The weighted average of the local gradient is 0.43 m/km and the porosity of the sediment is about 30% (IWACO, 2000). This leads to a velocity of 18.7 m/year. With this formula the hydraulic head distribution is considered to be linear, deviating from the Dupuit approximation used in the groundwater flow model.

The input concentration of substances is unknown, but considered to be a constant input as contaminants have to dissolve and move through the clay layer and was taken as the relative value of 100%.

As no exact data for substance specific adsorption or decay rates was available calculations were made for different scenarios.

1 – Advection and dispersion: Base

Contamination poses the biggest risk when no adsorption or decay takes place. This causes substances to travel with the groundwater velocity and disperse. The dispersion factor was calculated with Formula [2] and changes over distance.

2 – Base + Retardation

Retardation is strongly substance dependent and is taken into account as retardation factors of 2; medium retardation, 5; strong retardation and 10; very strong retardation.

3 – Base + Decay

The landfill was opened 63 years ago, so decay can have severe effects. Although half-lives of days or weeks are quite common, on this long term these are negligible. Half-lives of 100, 50, 10, 1 and 0.5 years were used. It has to be noted that calculating decay with this formula considers substances to decay only once entered into the aquifer. This is a reasonable condition as substances might still be in solid state when in the landfill. However, the circumstances in the landfill and the groundwater are both anaerobic and without sunlight, so this might cause underestimation of the decay for substances in solution.

4 – Base + Retardation and Decay

Decay is influenced by retardation as adsorbed substances do not decay as dissolved substances. Retardation factors of 1, 2 and 10 were used in combination with half-life values of 100 and 10 years and no decay.

Performed calculations are a simplification of the real situation. The used parameters were kept constant, while in the field parameters vary. Variety in soil parameters might alter retardation (fraction organic content) and flow velocity (porosity). Moreover, half-life values of organic substances depend strongly on environmental parameters such as pH, which vary with the soil, but are also influenced by the present contaminants. Infiltration of precipitation causes variations in flow velocity and therewith alter dispersion (Serrano, 1992). However, the calculation used for dispersion is based on field studies, so the approximation includes some variety in flow velocities.

In the clay layers the vertical conductivity is very low ($0,001 \leq k < 1$), which prevents groundwater and contamination to flow through. Contamination was therefore not expected to be found in the clay layers, but to be transported primarily horizontally within the aquifer.

3.4 PHASE 4 EXPOSURE

In phase 4 an answer is given to the fourth sub question:

Which contaminants can be present in contaminated media, corresponding to the exposure routes present in the area of the *Oijense Bovendijk* landfill?

The aim of this sub question is to assess the risks of the contaminants leaching from the landfill, based on the exposure routes.

3.4.1 Exposure routes

The outcomes of the substance transport calculations and the hydrological model were used as the basis to identify the exposure routes. The soil and groundwater-contamination model Csoil, developed and used by the RIVM (National Institute for Public Health and the Environment) recognises the following exposure routes:

- ingestion of contaminated soil particles
- dermal contact with contaminated soil particles
- inhalation of contaminated soil particles
- inhalation of contaminated vapours
- consumption of contaminated crops
- contact via contaminated drinking water (Brand et al., 2007)

The above exposure routes are based on human exposure, excluding ecosystem health. Therefore contamination of surface water and plant uptake were also included into this phase of the research. Potentially contaminated media from the Csoil model are soil particles, vapour, crops and drinking water. To broaden this for non-human exposure also surface water, bed sediments and water side mowings were included. After dredging and mowing the sludge and mowings are left behind at the local farmers (Personal Communication D. van Son, *Waterschap Aa en Maas*).

All routes and media were evaluated on their relevance in the OBL situation, based on the current and future land use. Information on the receptors and spatial developments was taken from maps, a field visit and personal communication with the municipality of Oss (N. van der Pluijm and J. Janssen).

3.4.2 Substances in contaminated media

For the relevant routes the presence of substances in the corresponding contamination media was evaluated using physiochemical substance parameters important to enter the medium. As no field measurements into the present concentrations were conducted a qualitative description of the probable environmental distribution of the contaminants is given. The extent to which contaminants can be present in the media depends on the substance properties, which are described in chapter 4.2.

Limited anaerobic degradation is a prerequisite for presence in groundwater and therefore drinking water. Presence in seepage and therewith surface water depends on the retardation of a contaminant in the subsurface and on aerobic degradation in the surface water. Although adsorption is generally considered with K_{oc} values below 10.000, which corresponds to K_D values below 5 when assuming f_{oc} to be 0.0005, the travel distance and time are of utter importance for the travelled distance of adsorbing solutes. These factors were determined in phase 3. Presence on soil particles and sludge is determined by the adsorption capacity and in crops or mowings by the uptake by plants. Sludge and mowings were only considered as a potential contaminated media when a contaminant was present in the seepage. The threshold values for the physiochemical factors were taken from phase 2 Substance Properties and were considered per substance group, as specific data was mostly lacking. In Table 6 the used driving factors for presence in a medium and the corresponding physiochemical factors are summarised.

Table 6 Determining factors for presence in contaminated media. Thresholds from Bradley et al. (2014)

Contaminated medium	Driving factor for presence	Physiochemical factor threshold
Ground and drinking water	High persistence	Half-life in soil >100 days
Seepage	Limited adsorption	K_{oc} < 10.000 (≈K_D <5)
Surface water	High persistence	Half-life in water >40 days
(Bed) sediment	Adsorption	K_{oc} > 10.000
Crops / mowings	Bioaccumulation	K_{ow} > 1000

4 Results

4.1 PHASE 1 WASTE CHARACTERISATION

4.1.1 Step 1 Base list of emerging substances in groundwater

The list of emerging substances found in groundwater by Klein & Duijnhoven (2013) and Lopez et al. (2015) consist of 187 substances, with on overlap of seven substances. The study of the time of development of these substances through the PubMed database ruled out 90 substances, because they were developed after 1970. Another 7 substances, like certain life style products, were not expected to have been used in the industries around Oss (see the substances from the base list in Annex 1 and 2; sources: 1 Lopez et al, 2015; 2 Klein & Duijnhoven, 2013)

None of the pharmaceutical substances and pesticides on the base list were excluded from further study, as the research- and development-labs of Organon might have been developing or testing these substances. The research facilities of Organon both improved the existing manufacturing procedures and developed new preparations. Furthermore, the development of analytical and pharmacological methods was part of the research activities. In the late 1940s Organon started to produce synthetic substances that were not found in nature and tested hundreds of substances for positive effects on known diseases, as tuberculosis (Tausk, 1978). These forms of research create a lot of miscreations and metabolies, which are possibly disposed of. Although, the expected amounts per substances are relatively small, while the variety of substances from research is large, the substances were not excluded from this research as more concrete information on the research practises is missing.

4.1.2 Step 2 Commercial waste

The contents of the waste depends strongly on the industry type and products that are produced. Historical research identified eight commercial industries in Oss in the 1950s that might have disposed of their waste at the OBL. These industries are the pharmaceutical, electrical lightning, abattoirs and meat production, textile finishing, margarine, asphalt, soap, and cleaning agents industry. Below the industries and associated products and substances are described in more detail.

Pharmaceutical Industry

The pharmaceutical industry in Oss was embodied by Organon, part of the current MSD. In the NIPA report (2013) it is assumed that Organon disposed of waste of unknown nature at the OBL. Injection needles and ampules have been found at the site.

Organon was founded in 1923 and started with the extraction of insulin out of animal glands from the Zwanenburg abattoirs. The pancreases were macerated and extracted with a slightly acidic alcohol of 95%. Then the moist from the filtrate was evaporated in warm air leaving a dry residue that was emulsified in a Ringer's solution. A Ringer's solution is a mixture that resembles body fluids and typically contains sodium chloride, potassium chloride, calcium chloride and sodium carbonate (Rosenfeld, 2002).

Besides insulin Organon developed hormones and steroids as lynestrenol and launched its first anticonception pill in 1962. Hormonal preparation were firstly extracted from animal products and urine. As with most extracts from natural substances the development stages include extraction and isolation, preferably in crystalline form, discovering the chemical structure and then the syntheses of the substance (Tausk, 1978).

Other preparations at that time mainly included vitamins (Organon, 1957). All substances listed in the compositions of the preparations were taken into account in this research. Extracts from animal organs were severely contaminated with other organic substances, even up to 99.9% (Tausk, 1978). Therefore, the active ingredients of these preparations are only found in fractional quantities and excluded from further research. The crystalline and synthetical forms of these substances were, as far as they were available at the time, included.

Electrical lighting industry

In Oss one of the Philips factories was active since the 1940s. Philips was the first company to produce light bulbs and developed sodium-, mercury-, trifluorescent and blended lamps. However, as in Oss mainly the lighting fittings were produced and not the bulbs itself the Philips factory is more similar to a metal processor and painting factory. Processes in the metal processing are cleaning, pickling, processing, joining, and surface treatments. The cleaning process can be both mechanically and chemically with alkanes, detergents, solvents or corrosive substances. For pickling, joining, treatments and painting chemicals and paints are also used (Department of Environment, 1995a, 1995b; Nieuwkoop, 1993). Philips also developed its own material, called Philite, which was produced in Oss from urea, phenol and formaldehyde. The factory also produced fluorescent powders for the lamps, using antimony and manganese from 1948 on (Gemeente Oss, 1949).

Abattoirs and meat products industry

In Oss meat production has played a major role since the end of the 19th century with companies as Hartog Factories and Zwanenberg Abattoirs and Factories. Both companies merged into Unilever (Unox) and Zwanenberg formed the basis for Organon.

The nuisance act permit for the OBL prohibited the dumping of cadavers and offal (Gemeente Oss, 1951). But the OBL was closed down because of mismanagement (DHV, 1993). Therefore the possibility that waste from the meat production industries is disposed of at the OBL cannot be excluded, so the industry was not excluded from this research.

The main processes in the industry are the slaughtering of animals, especially pigs, and the processing of the carcasses. Slaughtering includes anesthetizing, bleeding, scalding, depilation and skinning. As anesthetizing was electrical at the time no special substances are expected. During the processing phase, traditionally salt or smoke and later preservatives, emulsifiers, and antioxidants are used for preservation and tin for the packaging (Nieuwkoop, 1993). In the 20th century, refrigerant gases like ammonia, carbon dioxide, sulphur dioxide, and freon were used for the cooling of the meat, however, these substances are gasses and therefore unlikely to be found at the OBL. The chemical preservatives that have been used for meat in the 1950s include butylhydroxyanisole (BHA), sodium nitrite, sodium lactate, and sodium tripolyphosphate (Cassens, 2008).

Textile finishing

The textile finishing in Oss is mainly associated with the carpet production at Bergoss and Desseau. Bergoss later merged into its competitor, which is now known as Desso. During the 1950s wool and cotton were the main raw materials and they were treated, dyed or printed and finished. A broad variety of chemicals were used during the processes which are not all described in literature. In the treatment the fabrics were bleached with lime and chlorine, and later with sodium peroxide or hydrogen peroxide. For wool besides the peroxides also hydrosulphite preparations and permanganate were used. Both natural and chemical dyes were used with the addition of acids, salts and alkalis (Nieuwkoop, 1993). For the backside of the carpets PVC, polyurethane and styrene-butadiene were used (Department of Environment, 1995c). Fire retardants, like chlorinated substances and ammonium phosphate, and plasticizers were also in use and are now considered emerging substances (Department of Environment, 1995c).

Margarine production

Margarine as a butter from plant or animals fats was produced by Hartog Factories among others. During the production oils are merged with other ingredients and crystallized and kneaded. Emulsifying agents as lecithin or glycerides, preservatives as salt or benzoic acid and colorants as carotene or annatto were added.

Asphalt manufacturing

In Oss the factories of Elsbach Chemical Industry produces asphalt. During the production petroleum or bitumen were mixed with gravel leading to oil and tar by-products. Zinc chloride and sulphuric acid are used as supplementary substances.

Soap industry

The abattoirs of Zwanenberg also founded a soap factory where soap from fats from slaughterhouses was produced. In the production bleaching agents and bases used.

Cleaning agents industry

Elsbach's Chemical Industry also produced cleaning agents and polishes from oils and fats. These were made into soap with foliage and purified with sulphuric acid.

4.1.3 Concluding remarks and discussion

The base list of emerging substances detected in groundwater contains 187 substances. The industrial analysis brought in total 155 new substances to the light, containing 82 substances from Organon. Of the base-list substances, 90 were developed after 1970, and 7 not expected to be used in Oss and were therefore excluded from further research. Another 27 substances were already studied by NIPA and were not-detected or are not considered harmful, resulting in the potential presence of 218 substances (see Annex 1 - List of substances likely disposed of at the OBL). The construction of this list is illustrated in Figure 13. In Annex 2 – List of excluded substances the substances that have been excluded due to spatial or temporal delineation or previous studies are given.

It has to be kept in mind that it is uncertain whether the substances on this list have been disposed of at the OBL and in which quantities. Besides, the risks that are associated with the substances properties are not yet taken into account, let alone in the concentrations present. The list only shows the substances that can normally be expected to be disposed of at the landfill. Although the list has been produced carefully the historical character of the research and the lack of reporting at the time of operation can have caused substances to be overlooked.

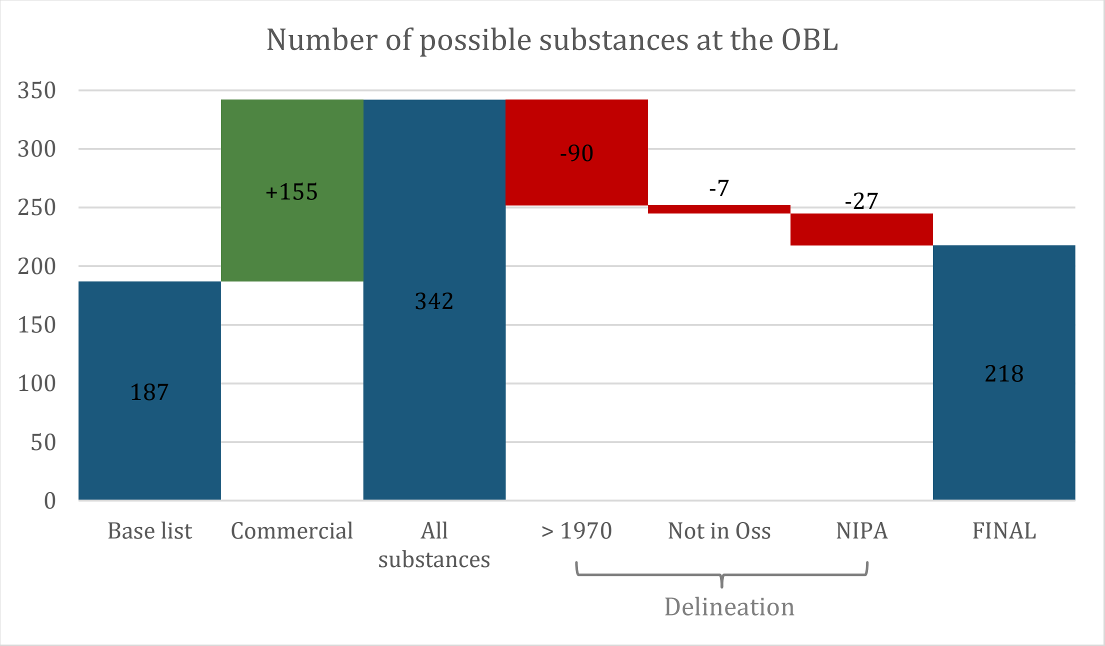


Figure 13 Waterfall graph of the number of possible emerging substances at the *Oijense Bovendijk* landfill.

4.2 PHASE 2 SUBSTANCE PROPERTIES

The substances identified in phase 1 are grouped according to their use and below the results of the literature review of the properties are described, to determine the risk potential of these groups.

4.2.1 Pharmaceutical substances

Antibiotics

Antibiotics are one of the most used medications for both prevention and curing of micro-organism induced illnesses. Antibiotics are also used in livestock, agriculture and aquaculture. Antibiotics from the agricultural sector mainly enter the environment directly via manure, while human antibiotics enter the environment via waste water (Kümmerer, 2008). However, antibiotics are not effectively removed in waste water treatment and are found in surface waters and rivers at various places ranging from ng/l to µg/l (W. Li et al., 2015).

Although studies regarding antibiotics in the environment severely increased in numbers in current years, their fate is still largely unknown (Kümmerer, 2008).

Biodegradation plays a role in the degradation of antibiotics in surface water and half-lives are found ranging from 4 up to 104 days (Ingerslev et al., 2001). However, contamination is demonstrated to be strongly related to human activities, as concentration spikes in densely populated urban areas, leading to a relatively constant input and, therefore, pseudo persistence (W. Li et al., 2015).

Antibiotics adsorb to soil, but this is a highly pH depended process. In the soil antibiotics may be biotransformed leading to degradation and inactivation, but might also produce metabolites. Both antibiotics and metabolites can be persistent in soils (Kümmerer, 2008).

Most antibiotics entering the environment directly are veterinary antibiotics. Most research on plant uptake is therefore done on veterinary antibiotics (Kumar et al., 2005). These studies show that most antibiotics are taken up by plants in the µg/g range, but others were not taken up. The differences could not be explained by adsorption coefficients, but simply the size of the molecule seems to be of influence (Kumar et al., 2005). Herklotz et al. (2010) studied the plant uptake of human pharmaceuticals and the antibiotic showed a BAF of 0.08 for the stem and leaf, and 10.9 for the roots (both dry weight).

The effects antibiotics have on microorganisms in the environment are comparable to their therapeutic anti-bacterial effects. Besides, low concentrations of antibiotics can cause microorganisms to become resistant to antibiotics. These resistant organisms can spread over large distances through water, air or by humans or animals (Berkowitz et al., 2014). As resistant organisms pose risks to human and environmental health, some scientists consider them to be pollutants themselves (Zhang et al., 2009).

Organon produced Isoniazide (Isonicotinic acid hydrazide) to cure tuberculosis and three varieties of sulphonamide antibiotics. Sulphonamide antibiotics are used in both human and veterinarian medicine. Holm et al. (1995) detected sulphonamide antibiotics in a contamination plume downgradient of a landfill. Sulphonamides are photodegradable, like many organic substances. Baran et al. (2006) studied several sulphonamides in sediments and found that they are not biodegradable and can accumulate in the food chain. The anti-bacterial therapeutic effects of sulphonamides can have a severe effect on micro-organisms in the soil. On higher organisms the acute toxicological effects are limited (Baran et al., 2011).

Hormones

Steroidal hormones are used as contraceptives and growth promoters. The latter are also often used in agriculture. Oestrogens, progesterone and testosterones are found with other steroidal hormones in the environment (Kümmerer, 2008). Both natural and synthetic oestrogens are found in surface water and counteract with the endocrine system of water organisms. The discovery of the sexual disruption in fish caused by hormones in the surface water, marked the start of studies concerning emerging pharmaceutical substances (Jobling et al., 1998; Ying et al., 2002).

The degradation rates of hormones in surface waters are disputed, as some oestrogens are biodegraded under aerobic conditions in both water and sediments, but a continues input of wastewater in surface waters causes pseudo-persistence (Holthaus et al., 2002; Kümmerer, 2008).

Hormones are mostly hydrophobic organic compounds and can therefore adsorb to organic matter (Dussault et al., 2009). In rivers in the United Kingdom oestrogens showed to have distribution coefficients (K_d) ranging from 4 to 121 l/kg, while sorption to suspended sediment in the water had K_d values up to 260 l/kg (Holthaus et al., 2002).

Oestrogens can be taken up by plants and their log K_{ow} is determined to be around 4.0. Bioavailability is considerably lower when adsorbed to soils than dissolved in water (Dussault et al., 2009). Artificially created wetlands are used as wastewater treatment in the removal of oestrogens. Removal efficiencies up to 84% have been found by Song et al. (2009), mainly influenced by the degree of aerobic circumstances and root density of the wetland.

Organon developed and produced the contraceptive pill, consisting of several oestrogens. Studies show a relatively fast decay of oestrogens in surface water, with half-life values ranging from several days to several months (Ying et al., 2003). However, the effects are not reduced, as the input of oestrogens in the environment is relatively constant. In anaerobic circumstances, such as groundwater, half-lives are considerably longer and in several experiments no decay has been detected (Ying et al., 2003). PNECs for steroid oestrogens in surface waters are suggested in the ng/l range (Caldwell et al., 2012).

Non-steroidal anti-inflammatory drugs (NSAID)

NSAIDs are pain-killing drugs and have anti-inflammatory effects at higher doses. NSAIDs are one of the most widely used drugs and many forms are available as over-the-counter drugs for which no doctor prescription is needed. Because of their wide use NSAIDs are often found at municipal landfills (Eggen et al., 2010).

Most NSAIDs are susceptible to biodegradation under aerobic conditions, leading to half-lives ranging from 4.8 to 69.3 days (Lin & Gan, 2011). NSAIDs have a moderate adsorption onto sediments from surface water and adsorbed NSAIDs also show to be susceptible for biodegradation (Antonić & Heath, 2007).

Non-steroid anti-inflammatory drugs have been tested in laboratory experiments in order to determine the uptake by plants. After application of high concentrations of NSAIDs most were detected in plants, but ibuprofen was not detected in soil nor plants after three months. A study with environmental concentrations of 4 NSAIDs was done by Cortés et al. (2013), who did not detect any of the NSAIDs in plants. Ibuprofen and diclofenac were detected in the sludge in the ng/g range.

Acute toxicity concentrations differ between substances and studies on water organisms show broad ranges of effect concentrations. Diclofenac has effect concentrations for all studied water organisms below 100 mg/l (Kümmerer, 2008). Experiments with a mixture of NSAIDS shows acute toxicity on algae species at concentration ranging from 72 until 626 mg/l (Cleuvers, 2004). The EU appointed diclofenac and aspirin as possible harmful to water organisms. For ibuprofen PNEC values between 5-10 µg/l are proposed (Stuer-Lauridsen et al., 2000).

Organon produced the first fever-reducing pain-killer, called fenacetine. Fenacetine turned out to cause kidney infections and some indications of carcinogenetic effects were found, leading to a ban on the product. Salicylamide, a pain-killer nowadays still available in tablets with aspirin and caffeine, was also produced by Organon.

Beta-blockers

Beta-blockers are used to reduce the heart speed and lower the blood pressure. Several beta-blockers are found in waste, surface and groundwater. Experiments with 3 beta-blockers showed the substances to be hydrophobic, leading to adsorption to sediment (Kibbey et al., 2007). One of the most used beta-blockers, propranolol, shows to be persistent and bioaccumulative (Maszkowska et al., 2014).

Acute toxicity of beta-blockers has been barely studied, so no clear effects can be proven (Fent et al., 2006). Moreover, the chronic toxicity is also unknown. Organon did not produce beta-blockers in the 1950s, so only limited concentrations, from research, might be present.

Lipid regulators

Lipid regulators are used to lower the amount of lipids in the blood to prevent heart attacks. Some lipid regulators also lower the triglyceride concentration in the blood plasma (Kümmerer, 2008). The fate and behaviour of lipid regulators is largely unknown, but clofibric acid, a metabolite of several lipid regulators has regularly been detected in surface waters, indicating the relative persistence (Halling-Sorensen et al., 1998).

Very limited studies regarding acute toxicity of lipid regulators are conducted. Only for clofibric acid a LC50 value in the range of 7.7-39.7 mg/l has been found and is therefore harmful to water organisms (Fent et al., 2006). Some effects are detected on animals, insects and plants (Kümmerer, 2008). Organon did not produce lipid regulators in the 1950s, so only possible research products can be expected at the OBL.

Neuro-active medication (anti-epileptics and antidepressants)

Medication to relieve epileptic effects has been in use since 1850 and consists of a large variety of substances with different action mechanisms. Carbamazepine is one of the most used neuro-active drugs and is detected in waste and surface waters. As it does not adsorb to sediments it can transfer to groundwater and enter the drinking water. Two commonly used neuro-active drugs, carbamazepine and diazepam, are classified as potentially harmful to water organisms with an acute toxicity below 100 mg/l (Fent et al., 2006).

Organon produced reserpine, a medicine base on a plant extract, for which no information about the fate and behaviour is available. The plant is found in eastern Asia, where it has been in use as traditional medicine for a long time. Nowadays substances with less side effects are used as alternatives for reserpine.

Vitamins

Vitamins are natural substances, but are also available in synthetic forms. Vitamins stimulate a specific function, but are excluded as pollutants by the EU. No literature on the fate of vitamins in the environment is available. However there are studies that show the toxicity of certain vitamins. Vitamin A can have negative effects on the nutrient uptake at concentration of at least twice the recommended daily amount for a longer period (Penniston & Tanumihardjo, 2006). Organon produced a broad assortment of vitamins, of which vitamin B and C are water soluble, while the others are fat-soluble.

Life-style products and Medication additives

Caffeine and nicotine are substances used for their stimulating effects. Besides the use as life-style products they may also be used as additives in drugs, as did Organon with caffeine in pain-killers (Organon, 1957). Both caffeine and cotinine, a degradation product of nicotine, are found in waste and surface waters. In the United States an extensive study showed caffeine concentration in creeks up to 6 µg/l. Caffeine does not degrade in surface or groundwater and can therefore be used to trace the anthropogenic influence on water systems (Fent et al., 2006). Caffeine shows to be toxic to surface water microbial communities (Lawrence et al., 2011).

Most other substances used as additives are natural salts and sugars. These substances are used in several products and production processes. Natural sugars and extracts like glucose or typhoid powder are mostly used as additives and are therefore considered to be found is quantities that have no negative effects on its surroundings and are therefore excluded.

Solvents were used in production processes and can spread over large distances in groundwater as they do not adsorb. Solvents can be used as tracers for the extent of contamination from pharmaceutical industries (Grodowska & Parczewski, 2010).

4.2.2 Industrial substances

Flame retardants and plasticizers

Organic phosphates are used in flame retardants (brominated or chlorinated) and plasticisers (non-halogenated) (Andresen et al., 2004). Flame retardants are used to create saver materials and nowadays about a quarter of the products contains bromide. Brominated flame retardants are mostly Polybrominated diphenyl ethers (PBDEs) and have be studied for a long time (Darnerud, 2003). PBDEs are persistent in surface water and the log BAF ratio ($C_{\text{fish tissue}} / C_{\text{dissolved water}}$) is found to be 5.5 to 8.5 for whole lake trout (Streets et al., 2006).

PBDEs are mostly hydrophobic and adsorb to sediments and solid matter in water (Gallen et al., 2016). Log K_{oc} was found between 6.2 and 6.5 (Streets et al., 2006). Severe soil contaminations of PBDEs have been detected in e-waste recycling sites in China with concentration in the surface soils of 2720–4250 ng/g dry weight (Leung et al., 2007). With soils from these e-waste sites plant-uptake experiments have been done by Huang et al. (2011), who showed that especially lower brominated PBDEs are detected in plants. Soil concentrations of 4.8 to 533 ng/g dry weight resulted in vegetation concentration from 2.1 to 217 ng/g dry weight for a sum of PBDEs (Wang et al., 2011). Distribution between roots and above ground tissues ranges between 0.18-0.31 ($C_{\text{aboveground tissue}}/C_{\text{root}}$) for BDEs. Higher brominated PBDEs decompose into low ones within a retention period of 4-350 days (Kim et al., 2006).

PBDEs are bioaccumulative and the highest concentrations are detected in fish and other water organisms. They are also detected in human tissue and breast milk. The effects of PBDEs remain largely unknown. Animal test show some PBDEs to have possible carcinogenic effects and have adverse effects on reproduction. Direct effect on humans have only be reported in cases in which employees were daily exposed to high concentrations of PBDEs during their work in factories. They showed severe neurotoxic effects (Darnerud, 2003).

Plasticisers are often used in the production of plastics and rubbers and are detected in the environment. With increasing length of the alkyl chain both sorption and persistence increase (Cousins et al., 2003). Plasticisers are, just like PBDEs neurotoxic and possibly carcinogenic (Andresen et al., 2004). Most plasticisers are phthalates, which have been measured but not detected by NIPA.

Pesticides

Pesticides are all substances used to prevent, destroy, repel or mitigate a pest. A large variety of pesticides is used and several are banned because of their side effects (Arias-Estévez et al., 2008). Many pesticides are persistent and pesticides are an important group within the Persistent Organic Pollutants (POPs), including DDT (Secretariat of the Stockholm Convention, 2015). They can also bioaccumulate in the food chain and because their organic and often neutral chemistry then sorb to soil particles (Arias-Estévez et al., 2008).

Pesticides are made to avert or kill certain organisms, which gives them an intrinsic toxicity. Several pesticides are banned because of adverse effects on human and environmental health. Because of the large variety of pesticides the effects can be very divers. Neurotoxicity, endocrine disruption and irritation are possible effects. Beside the target species, also non-target species can be adversely affected by the pesticides (Berkowitz et al., 2014).

Lindane and DDT were produced by Organon and are appointed as emerging contaminants by the EU. In a broad screening of 63 pesticides, including Lindane and DDT, NIPA (2013) did not detect any of the substances in the groundwater. Therefore, the chance that other pesticides, that are not known to have been produced in Oss, are present at the OBL is small.

Chemical cleaning agents

Chemical cleaning in metal works is often done with use of volatile organic compounds (VOCs), which are detected in groundwater. Some VOCs are persistent, while other undergo (bio)degradation. Sorption depends on the solubility of the substances (Zogorski et al., 2006). VOCs and other chemical cleaning substances are not considered as emerging substances, as many of these substances are included in routine monitoring (Nieuwkoop, 1993).

Dyes, paints and fixing agents

Dyes used in food production in the 1950s were natural substances, extracted from plants (Nieuwkoop, 1993). Nowadays all food additives are tested by the EU, labelling them with E-numbers.

Some paints or pigments are seen as emerging substances, like sulphate paints, mordents and pigments which can have toxic effects. These substances often contain metals as sink, chromium or cobalt, which causes these metals to enter the environment (Barceló & Petrovic, 2008). Toluidine-substances (methylaniline) are used to produce paint and are toxic, but no data about their fate and behaviour is known (Barceló & Petrovic, 2008). It is unknown which pigments have been used in the past.

Fixing agents bind substances that intrinsically do not bind. These substances are often used in the printing industry to improve the attachment of the ink to the paper. No literature on adverse effects of these substances is available and they are not considered emerging substances (NIPA, 2013; NORMAN, 2015).

Surfactants

In the textile and metal processing industries surfactants, such as Perfluorinated compounds (PFCs) are used as water and oil repellents. PFOS (perfluorooctane sulfonate) and PFOA (perfluorooctanoic acid) are the most extensively studied among the PFCs are detected in drinking water and the tissues of fish, birds, marine mammals and human blood. The routes of spreading are not fully understood, but their persistence has been clearly demonstrated (Eggen et al., 2010). PFCs are also persistent in surface waters and a PNEC of 36 µg/l has been estimated by the Stockholm Convention for PFOS (Brooke et al., 2004).

PFCs can adsorb to sediments. For PFOS K_d values have been found between 7 and 35 l/kg in different soils (Brooke et al., 2004). Zareitalabad et al. (2013) compared laboratory studies of sorption coefficients with the distribution detected in surface waters and sediments. They concluded that field situations may overestimate PFC concentrations in water, as K_{oc} values are usually higher in the field studies. Kwadijk et al. (2010) concluded after calculating sediment-water (K_d) distribution ratio's that the values correlate with the chain length of the compounds. This relation was also found by others in laboratory experiments (e.g. Higgins & Luthy (2006); Li et al. (2009)). Log K_{oc} values were found in a range from 2.2 – 3.7 (Kwadijk et al., 2010).

PFCs can be taken up by plants, with concentrations in plant tissues are normally around 1-2 times the concentration in the soil. Of which the higher ratios are related to lower concentrations in the soils. Concentrations in fruits were maximum 10% of soil levels (Brooke et al., 2004).

PFOS and PFOA are potentially toxic and bioaccumulative (Eggen et al., 2010). PFCs may have endocrine disrupting effects, however most studies focused on acute toxicology and only of PFOS and PFOA, while several other PFCs are used (Suja et al., 2009).

4.2.3 Concluding remarks and discussion

In Table 7 the above information is summarised and qualitatively ranked to provide insight into the risk potential of a substance group. It has to be noted that the risk potential +/- ? for substance groups with unknowns is unequal to +/- for substance groups without unknowns. However, in both cases negative effects cannot be excluded.

Information concerning pharmaceutical substances is often still lacking, leaving room for unforeseen effects. The mixture of substances is of great importance for the behaviour of the substances, as they strongly depend on pH. Mix-effects of pharmaceuticals can be quite severe as comparable action mechanisms of substances strengthen the effects, but are impossible to identify theoretically with current knowledge. Pharmaceuticals that are probably disposed of in larger quantities and can give adverse effects in are antibiotics, hormones, and NSAIDs.

Several industrial substances are not considered emerging substances or ions of the substances have been measured before. The most important industrial substances that were probably disposed of and potentially harmful are the brominated flame retardants and perfluorinated compounds.

Table 7 Summarising information on available data, persistence and harmfulness, leading to the risk potential. + = affirmative, +/- (?)= uncertain/neutral, - = negative, ? = unknown.

	Level of knowledge	Persistence	Harmful to humans	Harmful to environment	Risk potential
Pharmaceuticals					
Antibiotics	+/-	+	+/-	+	**+**
Hormones	+	+/-	+/-	+	**+**
NSAID	+/-	+	+/-	+	**+**
Beta-Blockers	-	?	?	?	+/- ?
Lipid regulators	-	?	?	?	+/- ?
Neuro-active drugs	-	+/-	?	?	+/- ?
Vitamins	+/-	?	-	-	**-**
Life Style Products & additives	+/-	+	+/-	+/-	**-**
Industrial substances					
Flame retardants	+	+	+	+	**+**
Surfactants	+/-	+	+	+	**+**
Pesticides	+	+	+	+	+/-*
Dyes, paints & fixing agents	+/-	+/-	-	-	+/-**
Chemical cleaning & solvents	+/-	+/-	+/-	+/-	**-***

* NIPA measured over 60 pesticides and none were detected around the OBL (NIPA, 2013)

** Except for some pigments, but no information on the used pigments is available

*** Many of the compounds on the list dissociate in water and are therefore untraceable

4.3 PHASE 3 CONTAMINANT SPREADING

4.3.1 Groundwater flow model

Making use of the Dupuit approximation, the hydraulic head distribution of the different time periods and the extreme surface water level events were calculated and are shown in Figure 14.

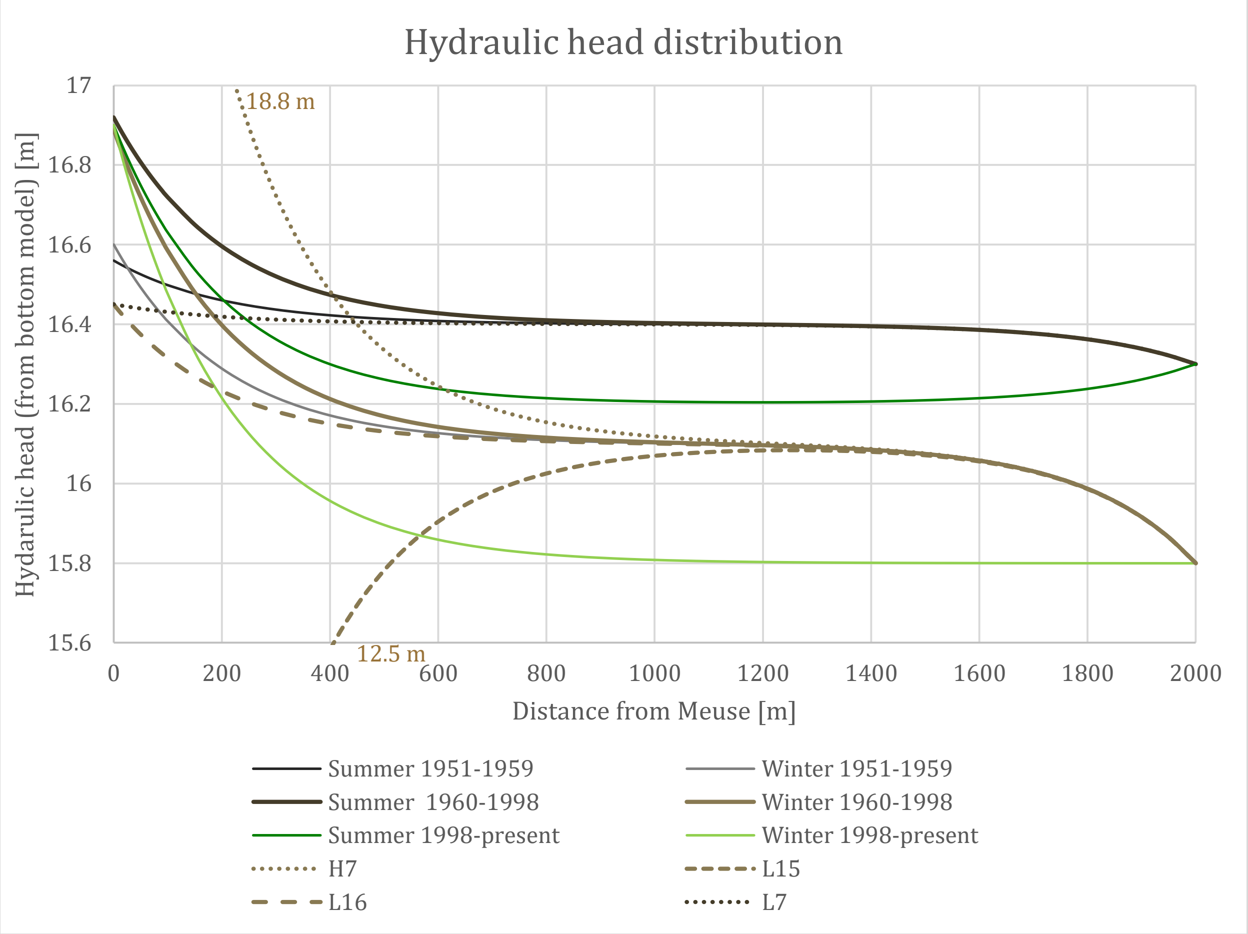


Figure 14 Hydraulic head distribution in the polder between the Meuse and the *Teefelse Wetering* for standard time periods and extreme water events in winter and summer (H = high water event, L = low water event).

As can be seen in the model outcomes (Figure 15), in which the isochrones have an interval of 10 years, groundwater flow velocities increased over time. Besides, a stagnation point settles in, which can also be explained by the hydraulic head distribution, as seen in Figure 14, that flattens or does even show an increase near the *Teefelse Wetering*, leading to no flow, or flow in the opposite direction.

A stagnation point or water divide withholds water to flow to the other site and causes seepage. Water will exfiltrate before the stagnation point, depending on the place of infiltration. As the landfill is located at 100-200 meters from the Meuse, the water will be transported more locally, see Figure 15.4. Residence times of potentially contaminated groundwater is at least 30 years, varying in the different time periods. It can be expected that at this time contaminated groundwater seeps up to the surface at approximately 100-300 meters downgradient from the landfill. As in most separate time period scenarios contaminated groundwater currently exfiltrates in approximately the same area, no time varying scenario is explored.

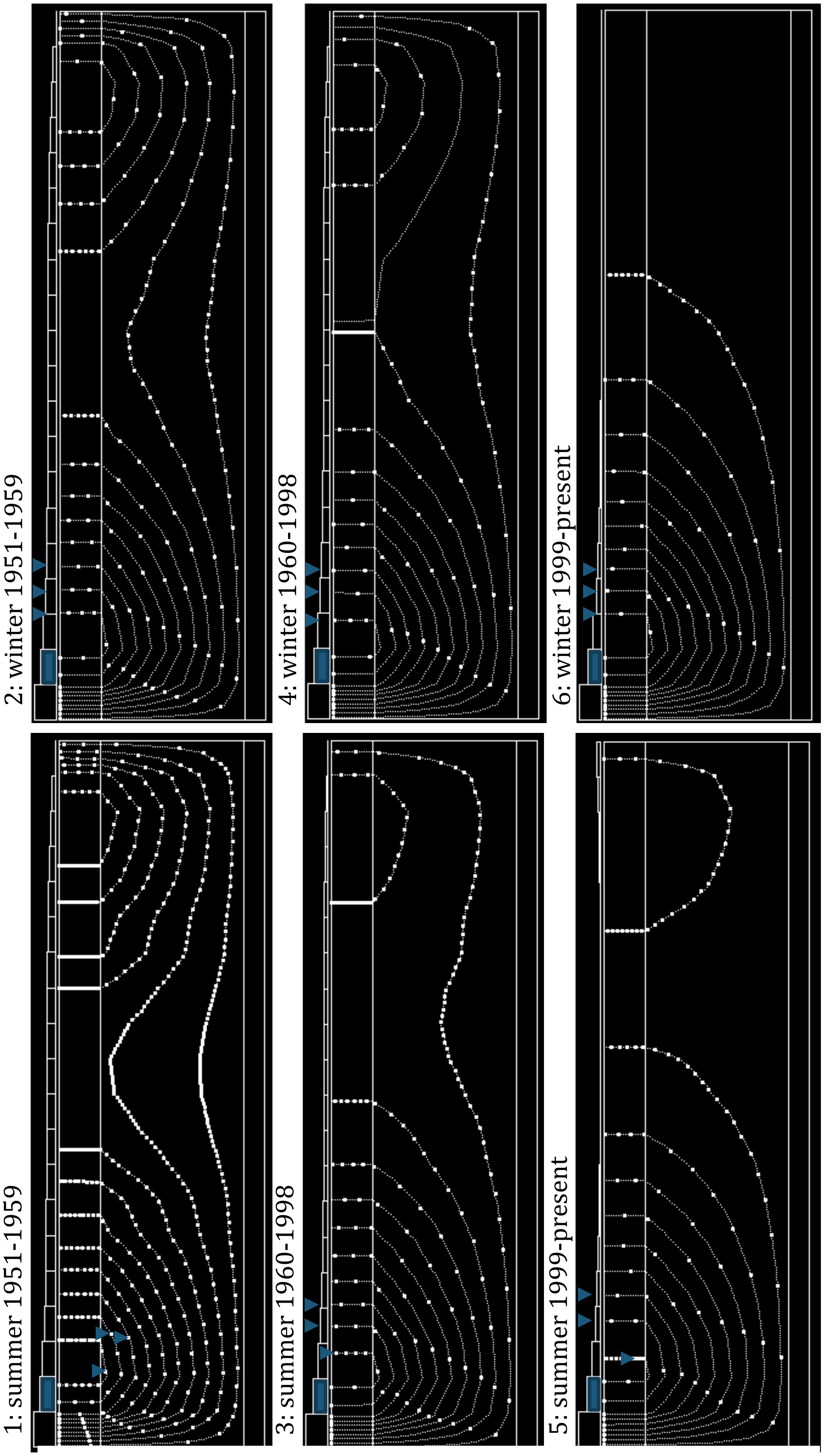


Figure 15 Flownet-model outcomes for normal circumstances for the different time periods, highlighting the landfill and location of the groundwater after 60 years. Isochrones highlight 10 year intervals along the streamlines.

High water events cause an increase in the hydraulic gradient and, therefore, an increase in groundwater flow velocities. Almost all high water events take place in winter, with the most extreme in January 1995. In Figure 16 the streamlines of this event are visualised and it shows that the residence time of groundwater near the landfill reduced to below 10 years, a decrease of a factor 4. It can also be seen that no stagnation point is present during the high water event, which theoretically means that water can be pushed behind the water divide during an event like this. However, this water is over a 100 years old Meuse water, so this is not an important process for contaminated groundwater.

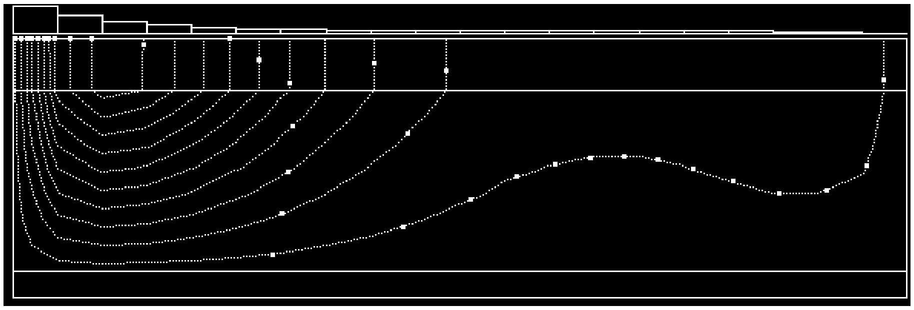


Figure 16 Streamlines during the high water event in January 1995.

Low water events also mostly take place in winter, but some summer events are present. An extreme low was detected in January 1985 with a lowest level of only 47 cm above NAP, lasting for 7 days. Because of this low water level and supposed constant water levels in the polder area and *Teefelse Wetering* the groundwater flow will reduce to standstill and start flow in the opposite direction, see Figure 17.

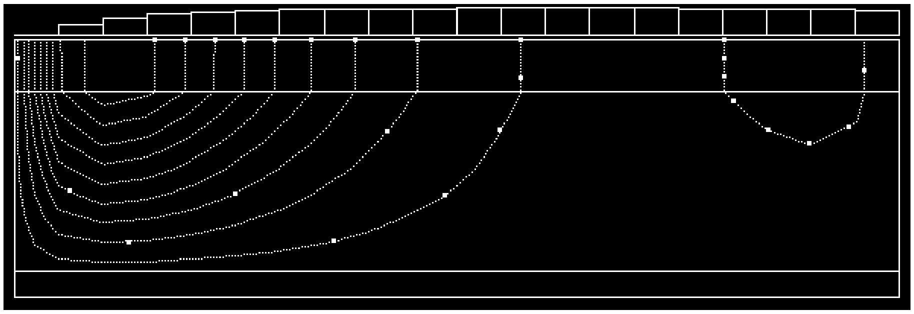


Figure 17 Streamlines during the low water event in January 1985.

As it is uncertain how the water levels in the polder and *Teefelse Wetering* reacted in this event, it cannot be stated that a change in flow direction actually took place, but a decrease in flow velocity will definitely occur. For a low water event in February 1995 some groundwater data is available, see Figure 18 and Figure 19. As can be seen in the graph, the groundwater in the polder does show variation with the extreme water events, especially the high water event. However, the variances are minor compared to the variances in Meuse water level. The streamlines modelled for the high and low water events with constant polder levels will therefore be adequate.

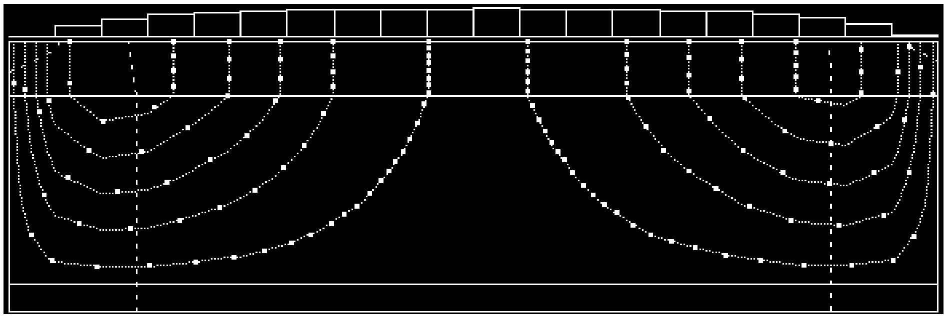


Figure 18 Streamlines of low water event February 1995.

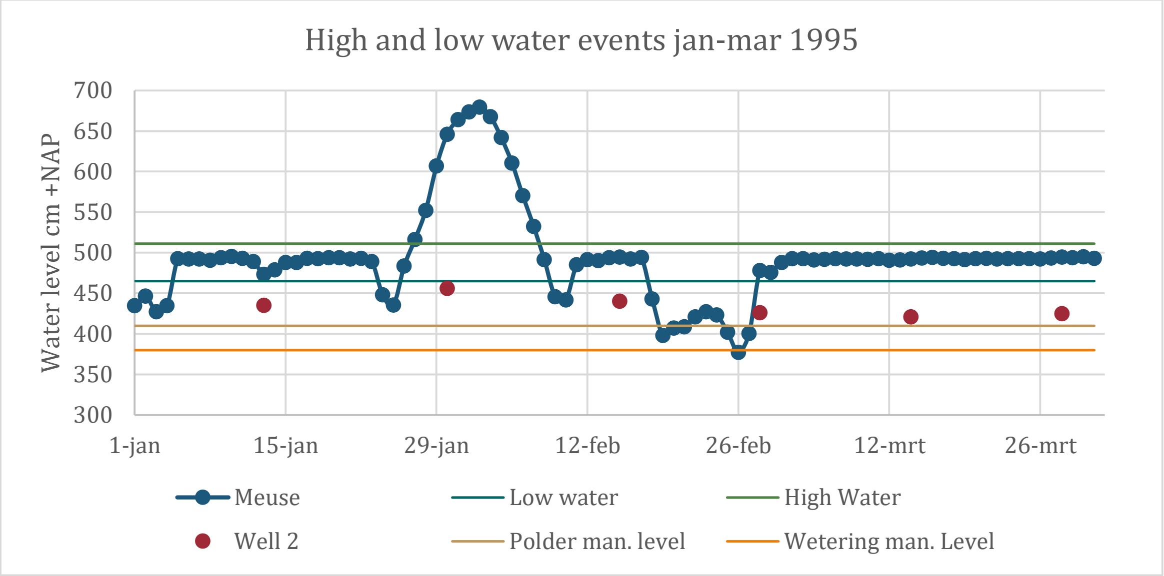


Figure 19 Water levels in the Meuse and monitoring well 2 in January - March 1995, with thresholds for high and low water events. Management levels of the polder and *Teefelse Wetering* included for reference. Source Meuse levels: Rijkswaterstaat (2015) and groundwater monitoring well: Dinoloket (2016).

A summer event with low water in August 1982 shows a severe slowdown of groundwater velocities and the occurrence of local flow systems and the relocation of the water divide, see Figure 20. The decreased flow velocities can also be expected by the limited hydraulic head difference, as can been seen in Figure 14.

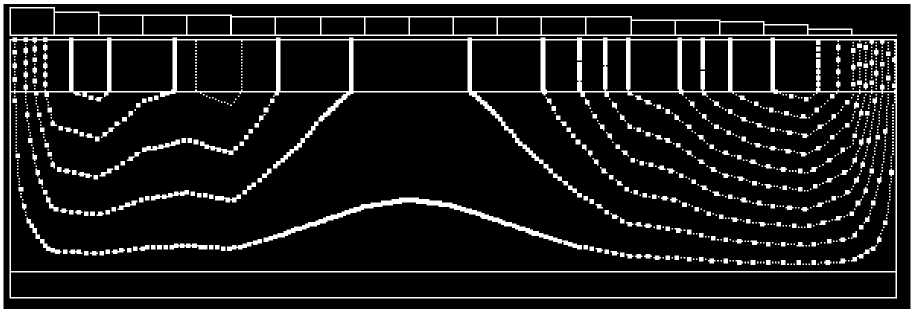


Figure 20 Streamlines during the low water event in August 1982.

4.3.2 Solute transport

1 – Advection and dispersion: Base

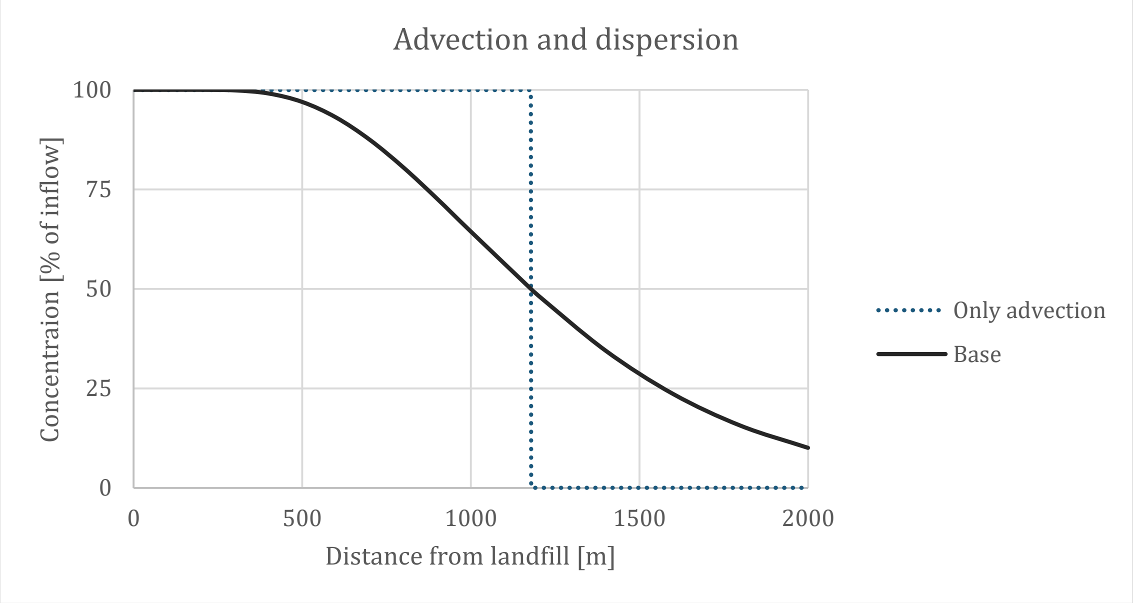


Figure 21 Relative concentration of substances at distance from the landfill due to advection and dispersion.

As can be seen in Figure 21 advection caused the substances to travel approximately 1 km, but dispersion caused the spreading of the front about twice the distance.

2 – Base + Retardation

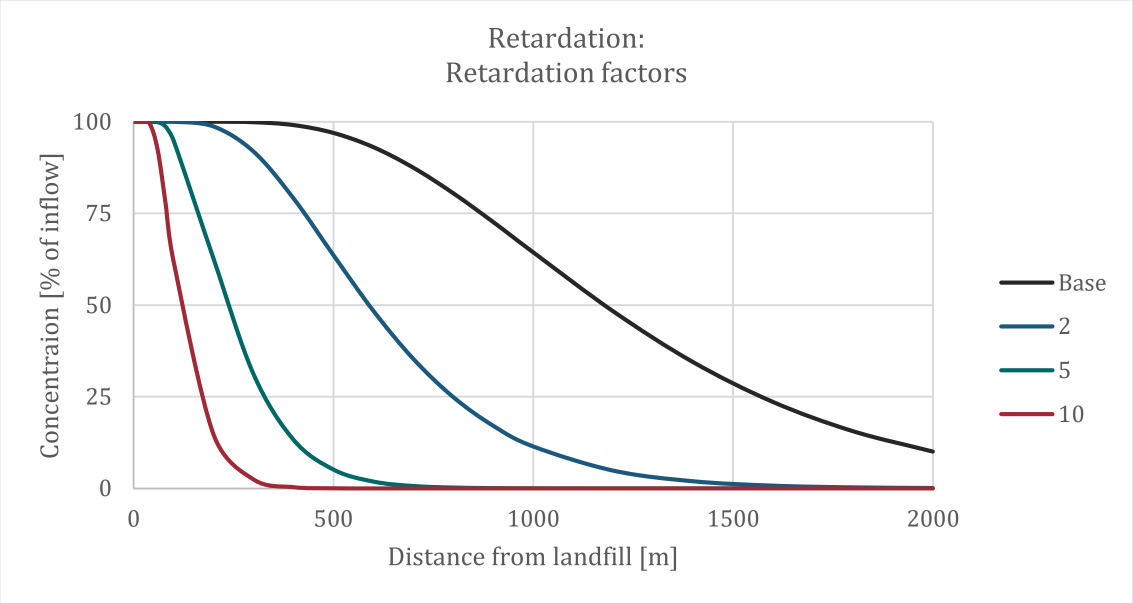


Figure 22 Relative concentration of substances at distance from the landfill due to advection, dispersion and retardation.

Figure 22 shows that medium retardation causes concentrations to drop, while stronger retardation severely shortens the travelled distance.

3 – Base + Decay

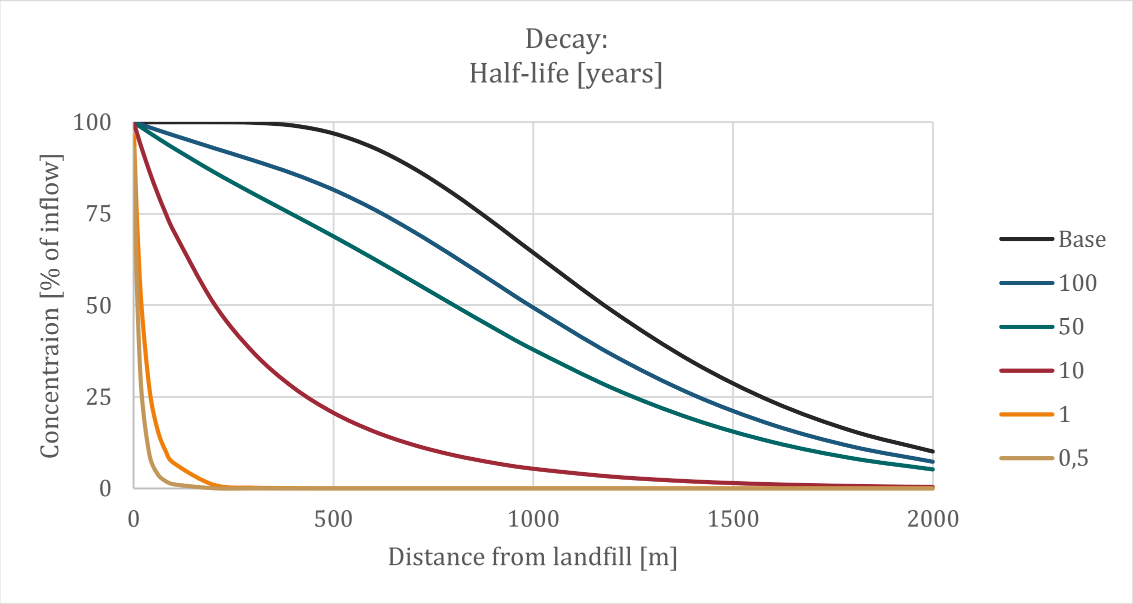


Figure 23 Relative concentration of substances at distance from the landfill due to advection, dispersion and decay.

Half-life values of 1 year and lower cause the substances to have fully decayed after 200 meters. A 10 year half-life value considerable lowers the expected concentrations, while values of 50 and 100 years only reduce the concentrations slightly.

4 – Base + Retardation and Decay

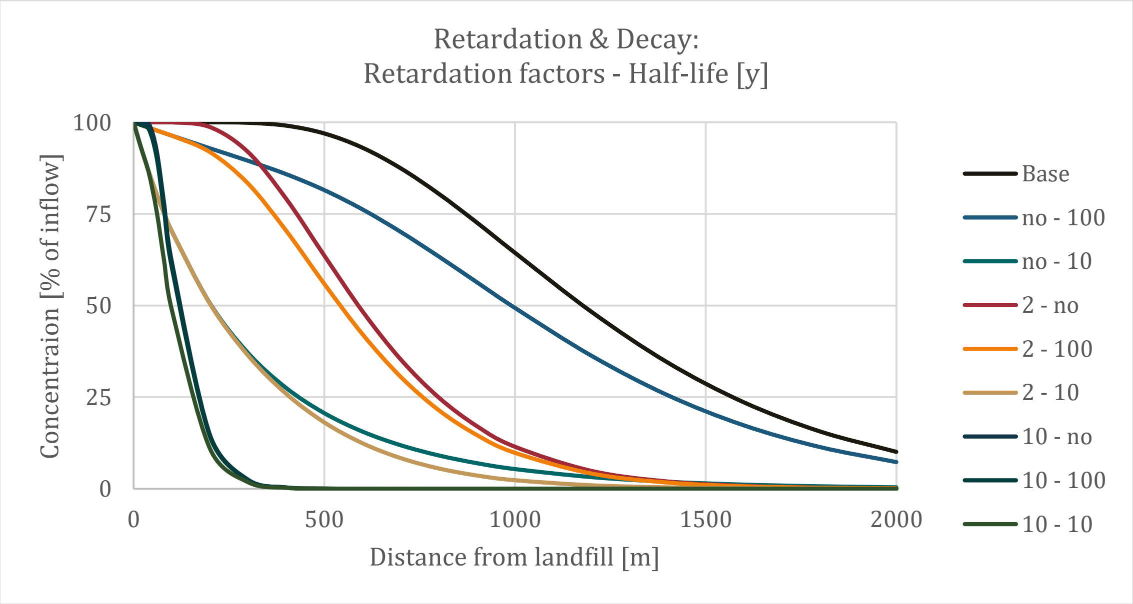


Figure 24 Relative concentration of substances at distance from the landfill due to advection, dispersion, retardation and decay.

Including all transport processes results in a wide variety of possible outcomes, as can be seen in Figure 24. Stronger retardation causes a decrease in the importance of decay, as adsorbed substances are calculated not to decay. It has to be noted that the concentrations are relative to the input concentration.

At a distance of 300 meters from the landfill groundwater exfiltrates, as shown in the hydrological model runs. When using an average groundwater flow of 18.7 m/y substances with no or low retardation and a medium or high persistence are expected to be currently (after 63 year) present in concentrations above 75% of the inflow concentration at this distance, see Figure 25. Substances with a relatively low persistence (10 year half-life) are expected in concentrations around 35%. Substances with high retardation (factor 10) are not yet expected. The geographical location of waterways around the OBL can be seen in Figure 6.

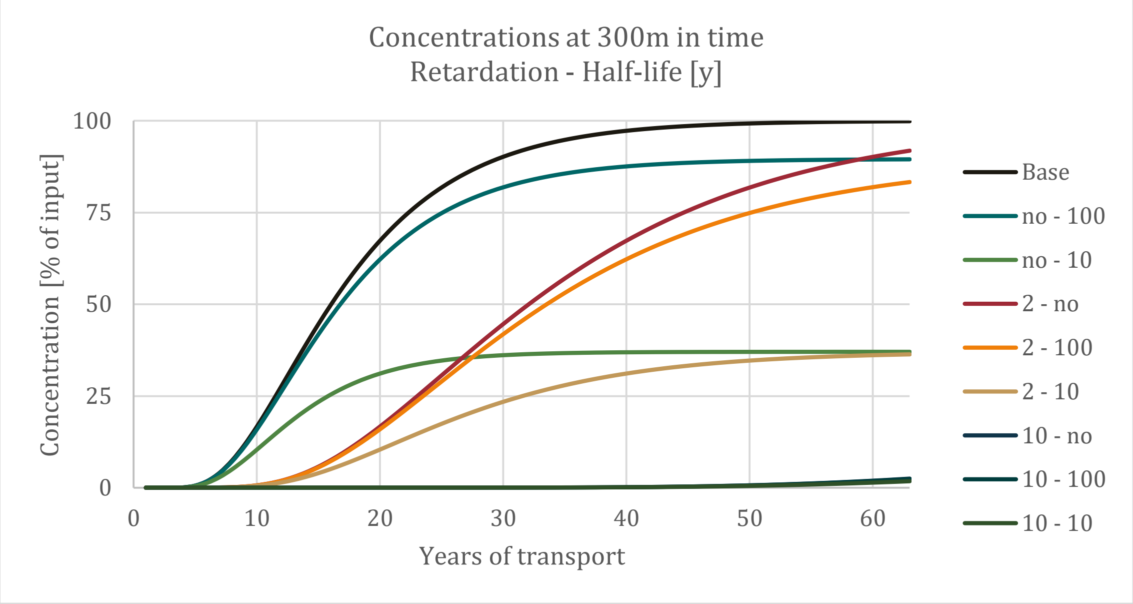


Figure 25 Break through curves of substances with different properties at a distance of 300 meter from the landfill, where exfiltration is expected.

4.3.3 Concluding remarks and discussion

Substance transport is expected to be in south-eastern direction, as groundwater flows from the higher Meuse level to the lower groundwater level in the polder area until it is drained at the *Teefelse Wetering*. The residence time of groundwater near the landfill ranges from 30-80 years, so contaminated water can currently exfiltrate in the polder. This also holds for substances with moderate adsorption capacities. The occurrence of extreme water events in the Meuse has a temporal influence on the groundwater flow. High water levels cause an increase in flow velocity and might push water to the other side of the water divide. This is however not the case for contaminated water, as this will exfiltrate spatially before the divide. Low water events slow the flow velocities and might even cause a change in flow direction, whenever the polder and *Teefelse Wetering* water levels are higher than the Meuse level.

Although these events influence the groundwater flow and therewith the substance transport it is due to the simplicity of the used model not possible to give an informed conclusion on specific substance transport routes and residence times. The hydrological model shows large varieties in groundwater flow velocities (30-80 year for 200-500 meters) and therefore the used fixed velocity in the substance transport equations could be an overestimation. This limitation is enhanced by the insufficiency of available data on the substance properties to develop a transport model or make substance specific calculations.

The identification of uncertainties is however an important step in the further research. The most important uncertainties are (1) which substances are found in the landfill and in which form and (2) how the contamination spreads, depending on the hydrogeological situation, whether a clay layer is present below the landfill and the substance properties.

4.4 PHASE 4 EXPOSURE

4.4.1 Exposure routes

Identification of receptors

Ingestion, dermal contact and inhalation of contaminated soil particles is largely prevented by the covering layer at the landfill terrain, although the thickness does not fully comply with the 0.5 meters required by law (NIPA, 2013). On the southern end of the landfill a car business and a residential building are located, on a partly hardened surface. The covering layer on this terrain has not been studied and the users might be exposed to the waste materials or contaminated soil particles. Another house is located beside the landfill, but has a garden overlapping the terrain of the landfill.

Inhalation of contaminated vapours is partly prevented by the covering layer and the fact that the terrain is not directly accessible. Besides, in the anaerobic decomposition phase mainly methane and carbon dioxide are produced (Kjeldsen et al., 2002).

Consumption of crops due to direct uptake of contaminants from soil is an unlikely exposure route, as no crops are grown on the landfill terrain and in the surroundings of the OBL most agricultural land is used for livestock. However, the few maize plots or other crops might be irrigated with contaminated surface water or groundwater, especially since the upper clay layer is prone to become dry in dry periods.

The drinking water company is in all probability not affected by the contamination of the groundwater, and the groundwater would be treated, lowering the possible risks. However, if a drinking water pipeline is found in or close to the landfill contaminants can permeate the pipe (Brand et al., 2007). Groundwater used by others as irrigation or drinking water is not treated and may be contaminated. Users with a capacity below 10 m^3/day do not need a permit and are therefore unknown.

Surface water can receive contaminated groundwater through seepage and is used by animals and nature. In the polder area no special nature areas are found, only along the Meuse a nature area from the Ecological Main structure is planned. Contaminants found in surface waters can have various sources in addition to the landfill and may enter aquatic organisms or bed sediments. The surface waters are managed by the water board, which leaves both sludge and mowed vegetation behind at the local farmers. Whenever surface waters are contaminated the sludge and mowings may also contain contaminants.

All possible relevant sources, routes and receptors in the OBL situation are shown in Figure 26. The geographical location of the objects is shown in Figure 27. The groundwater flow direction excludes the nature area and pumping station as users of contaminated water.

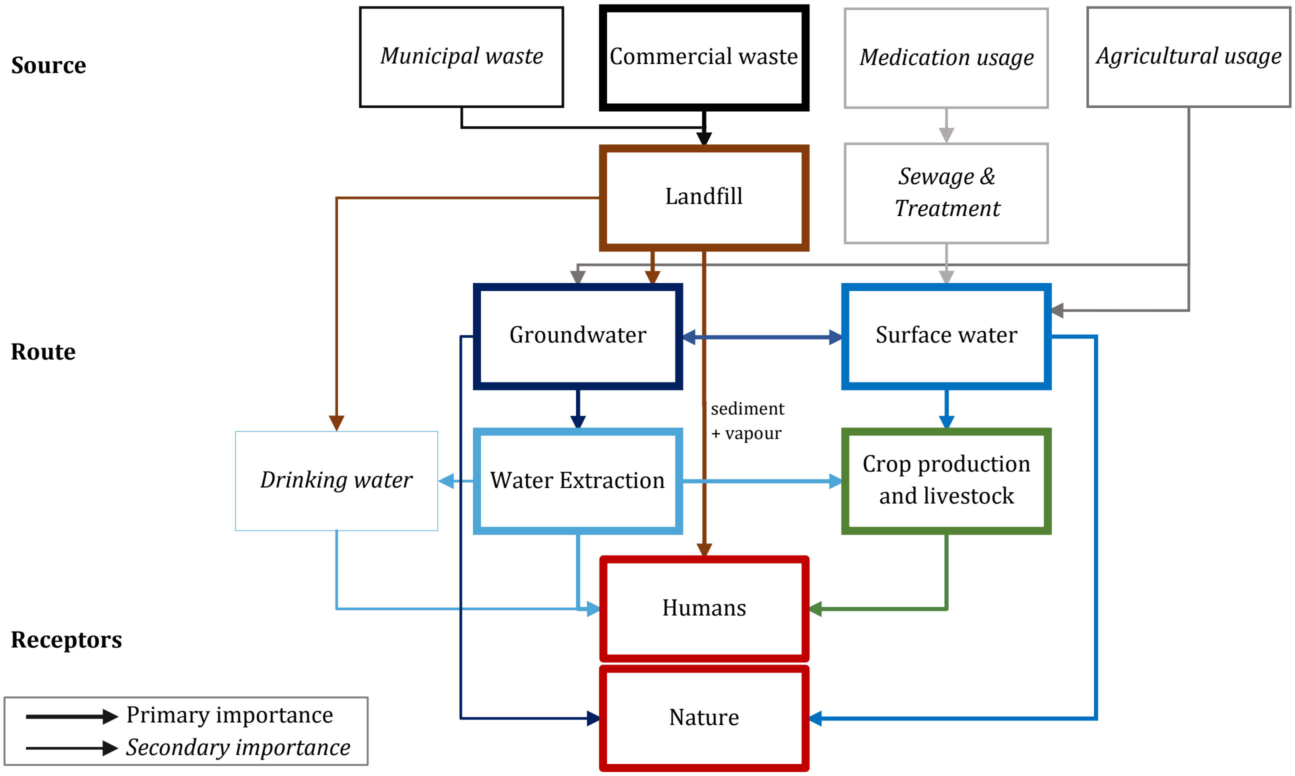


Figure 26 Sources, routes and receptors of contamination at the OBL. Importance of exposure routes is determined from qualitative data described in section 4.4.1.

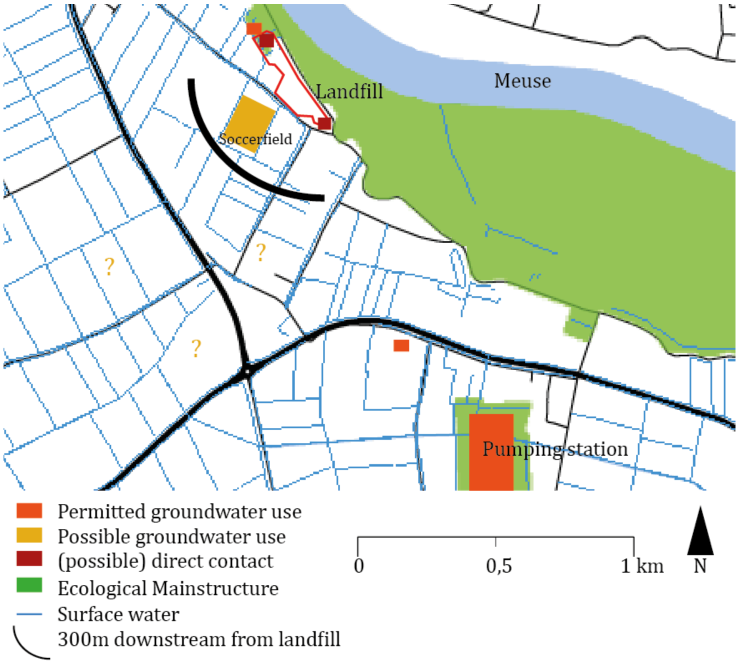


Figure 27 Geographical location of possible affected users.

Spatial Developments

No concrete development plans are available for the landfill site. The municipality of Oss intends to create a nature area closed for public or a second soccer field. For both plans no extensive excavation is necessary.

4.4.2 Substances in contaminated media

Based on above evaluation of the exposure routes, groundwater and surfaces water, including mowings and sludge, are identified as the most important media potentially containing contamination and facilitating exposure to its users.

The presence of contaminants in groundwater depends on the persistence under anaerobic circumstances and adsorption to sediments. While for surface water aerobic biodegradation and photo-degradation are important parameters determining the presence of contaminants. The presence in sludge depends on the adsorption potential of the substances and in mowing on the extent of plant uptake, and especially into the stems and leafs.

As antibiotics are prone to biodegradation and adsorption in soils, transport may inhibit the seepage into surface waters in the surrounding of the OBL. Biodegradation under aerobic circumstances varies from low to high persistence (half-lives 4-104 days) (Ingerslev et al., 2001). Chances of antibiotics to be present in high concentrations in the surface water are relatively small, but depend on the adsorption and persistence per substance. Exposure through mowings is not expected as uptake by plant stems and leaf is low (BAF 0.08) (Kumar et al., 2005). However, some antibiotics are persistent in sediments and low concentrations of antibiotics can cause resistance (Kümmerer, 2008; Zhang et al., 2009), so hazards from antibiotics cannot be excluded entirely.

Hormones are not expected to have reached the surface water in the surroundings of the OBL in 63 years as they adsorb to sediments (K_d 4-121 l/kg) (Dussault et al., 2009). However, as degradation does not take place in groundwater (Ying et al., 2003), the hormones can enter the surface water in the future. Although, degradation of oestrogen hormones in surface waters is relatively fast (half-lives < 100 days) (Ying et al., 2003), the proposed PNECs for steroid oestrogens in surface waters is low and therefore possibly reached in the future (ng/l range) (Caldwell et al., 2012). Although plant uptake of hormones is relatively high from the dissolved phase, it is considerably lower from the adsorbed phase, so currently no exposure through mowings is expected (Dussault et al., 2009).

Non-steroidal anti-inflammatory drugs are persistent in groundwater and have limited adsorption capacity (Lin & Gan, 2011), so they may be expected to travel over larger distances and be present in seepage in the surroundings of the OBL. However, as NSAIDs are prone to biodegradation under aerobic circumstances (half-lives 5-70 days) (Lin & Gan, 2011), the accumulation of the concentration in the surface water is tempered. However, no data on the effects of chronic exposure to low concentrations of NSAIDs on aquatic organisms is available, so risks cannot be excluded (Kümmerer, 2008). As NSAIDs potentially adsorbed to bed sediments also biodegrade and no plant uptake at environmental concentrations takes place (Antonić & Heath, 2007; Cortés et al., 2013), no risks for sludge of mowings is expected.

Polybrominated diphenyl ethers are persistent in both ground and surface water, but are not yet expected in the seepage in the polders around the OBL as strong adsorption of PBDEs takes place due to hydrophobicity (Log K_{oc} 6.2 to 6.5) (Streets et al., 2006). As transport is limited, uptake of PBDEs from soils by plants can only take place very close to the landfill. Leaves and stems may contain concentrations circa 10-15% of soil concentrations, as research by Wang et al. (2011) showed root concentrations to be about half of the soil concentrations and above ground tissues circa 20-30% of root concentrations. However, the root depth of the grass on top of the landfill prohibits it from taking up contaminants from below the covering layer.

Perfluorinated compounds are highly persistent in both ground and surface waters. However, adsorption depends on the chain length of the compound, and therefore the presence of PFCs in the seepage in the polders near the OBL will be substance depended and limited (Log K_{oc} 2.2-3.7) (Kwadijk et al., 2010). It is uncertain whether the PNEC of 36 µg/l (Brooke et al., 2004) will be reached in the local surface waters, as this depends on the initial concentration in the landfill leachate. Where PFCs are found in soil water, mainly close to the landfill, plants can take up the contaminants with concentrations up to twice the soil concentration (Brooke et al., 2004).

4.4.3 Concluding remarks and discussion

In the surrounding of the OBL the major exposure routes are through contact with groundwater and surface water, including bed sediments and waterside plants.

In Table 8 the probabilities of substances to be found in an exposure medium based on the substance properties are summarised. Substances are assumed to be present in seepage when they are persistent in groundwater and have limited or no adsorption capacity. The presence in the seepage and the persistence in surface water determine the presence in the surface water. When a substance is present in the seepage it can adsorb to bed sediments or mowings, depending on the substance properties.

Table 8 Summarising the properties and presence of a substance group in an exposure medium in the OBL case. + = affirmative, +/- = to some extent, - = negative.

	Groundwater		**Seepage**	**Surface water**		**Sludge**	**Mowing**
	Persistent	Adsorbing	Present	Persistent	Present	Present	Present
Antibiotics	+/-	+	-	-	-	-	-
Hormones	+	+	-	-	-	-	-
NSAIDS	+	+/-	+/-	-	+/-	-	-
PBDEs	+	+	-	+	-	-	-
PFCs	+	+/-	+/-	+	+/-	+/-	+/-

Antibiotics, hormones and NSAID adsorb to some extent to the soil and therefore spread more slowly in groundwater, and reduce the input in surface water. Besides, persistence of pharmaceuticals in surface water is lower due to biodegradation. As concentrations in surface water are currently limited in the OBL case, no risks for the sludge and mowings are expected at this moment. Large differences in fate and effects between different pharmaceuticals are present, so specific pharmaceuticals may show other behaviour than stated above.

PBDEs adsorb to the soil and are therefore not yet expected in the surface water. As uptake into stems and leaves is relatively low, mowing is not expected to be harmful.

PFCs are persistent and mostly non-adsorbing compounds, so they are expected to be present in the surface water. Compared to contaminant levels in ground and surface water the concentrations of PFCs that can be expected in sediments or plants are considerably lower, as adsorption and plant uptake are limited.

The concentration of the contaminants in the contaminated media are unknown, but determine the actual possible exposure to the users of the surroundings of the OBL.

5 Discussion

This research focusses on the risks posed by emerging substances in the leachate of the *Oijense Bovendijk* landfill. Limitations in data availability and methodology have been described within the relevant chapters. Moreover, the following points are relevant for discussion.

Potentially present substances

Historical research showed substances, such as pharmaceuticals, brominated flame retardants, pesticides, paints and surfactants, to be potentially present in the commercial waste. As this list is partly based on historical research and partly on emerging substances detected in European groundwater, and has not been checked with field sampling, a comparison with other studies can only partly be made. Leachates with toxic substances have been detected at unlined landfill in several studies (e.g. Barnes et al., 2004; Baun et al., 2000; Eggen et al., 2010; Holm et al., 1995). Eggen et al. (2010) performed a broad screening of emerging substances in the leachate of two municipal landfills and compared the outcomes to 4 other studies. Chlorinated alkyl phosphates (flame retardants), DEET, PFCs and NSAIDs were detected at nanogram or microgram per litre levels. Holm et al. (1995) studied a municipal and industrial landfill with pharmaceutical waste and detected several sulphonamide antibiotics and seven other pharmaceutical substances. Except DEET, which was already measured by NIPA (2013) these substances are all considered to be potentially present at the OBL.

Risk potential

Literature review into the substance properties shows above all the lack of data on these substances. No or little available data on a substance (group) does however not imply that a substance cannot cause harm, but in this literature based research no possibilities for toxicity tests were present and all relevant studies have been used in the analysis. Although the lack of data the groups of antibiotics, hormones, NSAIDs, flame retardants and surfactants have been identified as the groups with the highest risk potential. These substances have relatively high persistence and show some adverse effects. These substances all have been detected in landfill leachate by the studies of Eggen et al. (2010) and/or Holm et al. (1995). Toxicity of the contamination plume of the latter study was measured by Baun et al. (2000) using bioassays and showed toxicity up to 80 meter downgradient of the landfill.

Spreading and exposure

The spreading of the contaminated groundwater has been modelled and calculated and shows that contaminated groundwater will cause seepage at the surface and surface water within several hundred meters of the landfill. This is caused by the management of the surface water and polder levels. As extreme water events in the Meuse are only slightly mimicked by the groundwater levels, the events have a strong effect on a temporal scale. However, as residence times range from 30-80 years under normal circumstances and extreme events are up to 1 month, the overall effect is small. As there are more low water event than high water events the groundwater velocities based on the management levels can be an overestimation of the average velocity.

Foppen & Griffioen (1995) modelled exfiltrating groundwater in Dutch polders and concluded that this is an important contributor to surface water runoff in these areas. Their model showed a 30-90%

discharge via drains, transporting Fe and PO_4. This shows that substances can be transported to the surface water in polder areas. However, their study also shows that the upper groundwater and surface water dilutes with precipitation, considerably lowering the concentrations. This process is not considered in this research.

In a study by Tauw into a landfill in a polder area near Dordrecht in the Netherlands a comparable hydrological situation was modelled and the contaminant spreading was confirmed by field sampling. A drain was constructed to enhance the seepage-effect in the polder and to control the exposure of the contaminants (Verschoor, 2015).

The spreading of contaminants at the OBL has been calculated with substance transport calculations. These calculations are a simplification and are based on a linear head distribution, which is not the case in a polder system. However, combining the hydrological modelling and the calculations gives insight into the possible exposure routes. As the contaminated groundwater will exfiltrate within circa 300 meters from the landfill it can be seen in the substance transport calculations that substances with medium to high persistence (half-life > 10 year) and low or medium retardation (retardation factor ≤2) can be expected at relatively high concentrations.

The pharmaceutical substances are all susceptible for adsorption to sediments, causing retardation. But under anaerobic circumstances they are prone to (limited) degradation. So they are considered present in groundwater, but for a limited extend in the seepage and surface water. The persistent PBDEs also adsorb to sediments, limiting their presence in seepage, while PFCs show high persistence and only limited adsorption and retardation. PFCs therefore have the highest potential to travel to the surface water and adsorb to bed sediments or be present in mowings. The actual concentrations in the media and the amount of contact determine the risks for the users of the surroundings of the OBL.

The properties important for contaminant spreading are however dependent on the environmental conditions and can vary within the substance groups, which are unknown in this case. Besides, as no measurements of the actually present contaminants and their spreading have been conducted in this research, the risk assessment is based on the assumption that all contaminants are present in the leachate. Consequently, comparison to other studies is only partly possible and relevant.

Potential risks

The risk assessment of leachate from the landfill in this research is based on the spreading, exposure and effects of the separate detected contaminants. As mixture effects may take place and other contaminants might be present this can underestimate the total toxicity. Pieterse et al. (2015) used CALUX bioassay tests to determine the toxicity of leachate from a pesticide landfill in Tajikistan. The test showed endocrine disrupting effects which could not all be explained by the substances identified in the performed chemical analysis. This shows that chemical analysis might underestimate the toxicity of landfill leachates and a bioassay test would be essential to give an informed risk assessment at the OBL.

Other landfills with comparable waste, like other landfills in the municipality of Oss, might have groundwater contamination with emerging substances, but exposure routes and media can differ, changing the risks.

Management implications

In the Netherlands the contaminations from landfills have been explored and whenever deemed necessary further studied. As closed landfills fall under the jurisdiction of municipalities and provinces, but the national research program has terminated, the municipalities have limited incentive and budget, so often no clear management or research objectives are set. However, as other studies (eg Eggen et al., 2010) and this research emphasise, previously unmonitored groundwater contamination can pose risks to the surroundings of closed landfills. One of the most important stakeholders in groundwater contamination are the drinking water companies. In an interview with researchers from Vitens, the largest Dutch drinking water company, it was said that the gap between the authority of the municipality and their own interest is a big hindrance and Vitens is planning to stimulate further research on the national or regional level and simultaneously do their own research on emerging substances from old landfills to secure their water sources (Personal Communication Rob Klijn, Martin de Jonge en Rob Breedveld from Vitens).

6 Conclusion

Historical research and monitoring outcomes from other studies show a large amount of substances and substance groups to be potentially present at the OBL, which were not yet measured in previous studies. Important substance groups include pharmaceuticals, some pesticides, surfactants, pigments and cleaning agents. Considering the persistence, harmfulness and availability of these substance groups the risk potential of antibiotics, hormones, NSAIDs, PFCs and PBDEs show to be the highest. These substances have also been detected in landfill leachate in other studies.

The hydrology in the surroundings of the OBL is driven by the managed water levels of the Meuse and the polder surface waters. Although levels are artificially kept constant, the Meuse level can vary due to extreme runoff events. The changes in hydraulic head distribution can have considerable temporal effects on the groundwater flow velocity and in some cases even the flow direction. However, as residence times are in the order of tens of years the events have limited influence. As more low water events occur then high water events, the average groundwater flow velocity might be overestimated.

Groundwater seeps up at a distance of circa 300 meters from the landfill, where it enters the surfaces water and could adsorb to bed sediments and be taken up by waterside plants. Under aerobic circumstances the pharmaceuticals show biodegradation and all groups can be expected to adsorb and be taken up by plants to some extent.

Groundwater contamination of emerging substances in the leachate from the OBL can be expected. However, the risks of the contamination cannot be determined as leachate concentrations are unknown. Contaminated groundwater is not used in large quantities, but does seep up to the surface water in the polder area. However, due to adsorption most substance groups will probably not yet have reached the surface water. Acute toxicity to humans or the environment is not expected as due to retardation, spreading and precipitation the contaminants dilute. However, low concentrations of contaminants can have adverse effects on aquatic organisms. It is therefore recommended to conduct field sampling in the leachate and the surrounding surface waters. Detection of toxic amounts of substances requires further research into the exposure and possible solution at the OBL. This also urges for more research and possibly revaluating the risk perception of closed landfills.

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Annexes

ANNEX 1 - LIST OF SUBSTANCES LIKELY DISPOSED OF AT THE OBL

Table 9 List of substances likely disposed of at the OBL by industry and substance type.

Sources: 1 Lopez et al, 2015; 2 Klein & Duijnhoven, 2013; 3 Organon, 1957; 4 Nieuwkoop, 1993; 5 DOE, 1995a, 1995b & 1996

Industry	Substance type	Substance	Source
Asphalt	Other Industrial Compounds	Tar acids	4
Margarine	Other Industrial Compounds	Annatto	4
Margarine	Other Industrial Compounds	Benzoic acid	4
Margarine	Other Industrial Compounds	Carotene	4
Margarine	Other Industrial Compounds	Lecithin	4
Meat	Other Industrial Compounds	Butylated hydroxyanisole (BHA)	4
Meat	Other Industrial Compounds	Sodium lactate	4
Meat	Other Industrial Compounds	Sodium tripolyphosphate	4
Metal	Chemical cleaning	Ammonium fluoride	4
Metal	Chemical cleaning	Hydrochloric acid	5
Metal	Chemical cleaning	Hydrofluoric acid	4, 5
Metal	Chemical cleaning	Massicot (Lead(II) oxide)	4
Metal	Chemical cleaning	Methyl chloride	4
Metal	Chemical cleaning	Nitric acid	5
Metal	Chemical cleaning	Sodium carbonate	4
Metal	Chemical cleaning	Sodium dodecyl sulfate	4
Metal	Chemical cleaning	Sulfuric acid	4, 5
Metal	Chemical cleaning	Trisodium phosphate	4
Metal	Dyes and paints	Phenol formaldehyde	5
Metal	Dyes and paints	Tripheyl methane	5
Metal	Other Industrial Compounds	Benzotriazole	2
Metal	Other Industrial Compounds	Manganese	6
Metal	Other Industrial Compounds	Urea	4
Metal	Solvent	1-nitropropane	5
Metal	Solvent	Acetone	4, 5
Metal	Solvent	Aniline	5
Metal	Solvent	Cyclohexanone	5
Metal	Solvent	Dimethylformamide	5
Metal	Solvent	Methyl ethyl ketone	5
Metal	Solvent	Methyl isobutyl ketone	5
Metal	Solvent	N-methyl-2-pyrrolidone	5
Metal	Solvent	Toluene	4, 5
Metal	Solvent	Xylene	4, 5
Pharmaceutical	Life Style Products	Caffeine	1, 2, 3
Pharmaceutical	Medicine	Carbarsone	3
Pharmaceutical	Medicine	Codeine	1, 3
Pharmaceutical	Medicine	Egraine	3
Pharmaceutical	Medicine	Ephedrine hydrochloride	3
Pharmaceutical	Medicine	Ergometrine	3
Pharmaceutical	Medicine	Ferrous tartrate	3
Pharmaceutical	Medicine	Heparin	3
Pharmaceutical	Medicine	Hydrocortisone acetate	3
Pharmaceutical	Medicine	Insulin	3
Pharmaceutical	Medicine	IP-6	3
Pharmaceutical	Medicine	Isonicotinic acid hydrazide	3
Pharmaceutical	Medicine	Magnesium trisilicate	3
Pharmaceutical	Medicine	N1-sulfanilyl-N2-n-butylcarbamide	3
Pharmaceutical	Medicine	Nitroglycerine	3
Pharmaceutical	Medicine	Norethisteron	6
Pharmaceutical	Medicine	Papain	3
Pharmaceutical	Medicine	Papeverini	3
Pharmaceutical	Medicine	Phenacetine	3
Pharmaceutical	Medicine	Prednisolone	3
Pharmaceutical	Medicine	Prednisone	3
Pharmaceutical	Medicine	Quinine	3
Pharmaceutical	Medicine	Regonol	3
Pharmaceutical	Medicine	Reserpine	3
Pharmaceutical	Medicine	Rutin	3
Pharmaceutical	Medicine	Salicylamide	3

Pharmaceutical	Medicine	Salicylic acid	1, 3
Pharmaceutical	Medicine	Saponins	3
Pharmaceutical	Medicine	Sodium sulfacetamide	3
Pharmaceutical	Medicine	Sulphanilamide	3
Pharmaceutical	Medicine	Tripelennamine	3
Pharmaceutical	Medicine additive	Aluminum hydroxide	3
Pharmaceutical	Medicine additive	Ammonium chloride	3
Pharmaceutical	Medicine additive	Boric acid	3
Pharmaceutical	Medicine additive	Calcium carbonate	3
Pharmaceutical	Medicine additive	Calcium gluconate	3
Pharmaceutical	Medicine additive	Dicalcium phosphate	3
Pharmaceutical	Medicine additive	Magnesium Carbonate	3
Pharmaceutical	Medicine additive	Maleic acid	3
Pharmaceutical	Medicine additive	Potassium chloride	3
Pharmaceutical	Medicine additive	Protein hydrolysate	3
Pharmaceutical	Medicine additive	Fructose	3
Pharmaceutical	Medicine additive	Glucose	3
Pharmaceutical	Medicine additive	Honey	3
Pharmaceutical	Medicine additive	Lactose	3
Pharmaceutical	Medicine additive	Yeast	3
Pharmaceutical	Medicine extracts	Extractum Thymi	3
Pharmaceutical	Medicine extracts	Hyaluronidase-Preparation	3
Pharmaceutical	Medicine extracts	Intrinsic Factor Castle	3
Pharmaceutical	Medicine extracts	Liver Extract	3
Pharmaceutical	Medicine extracts	Liver Preparation	3
Pharmaceutical	Medicine extracts	Ovarian Extract Powder	3
Pharmaceutical	Medicine extracts	Pancreas Preparation	3
Pharmaceutical	Medicine extracts	Pituitary Lobe Extract	3
Pharmaceutical	Medicine extracts	Pituitary Pituitary Extract	3
Pharmaceutical	Medicine extracts	Stomach Preparation	3
Pharmaceutical	Medicine extracts	Thyroid Powder	3
Pharmaceutical	Medicine extracts	Ergometrini Maleas	3
Pharmaceutical	Steroids and Hormones	Cortisone acetate	3
Pharmaceutical	Steroids and Hormones	Deoxycorticosterone	3
Pharmaceutical	Steroids and Hormones	Estradiol	3
Pharmaceutical	Steroids and Hormones	Estradiol benzoate	3
Pharmaceutical	Steroids and Hormones	Estrone	1, 3
Pharmaceutical	Steroids and Hormones	Ethinyl estradiol	3
Pharmaceutical	Steroids and Hormones	Lindane	3
Pharmaceutical	Steroids and Hormones	Methylandrostenediol	3
Pharmaceutical	Steroids and Hormones	Methylestrenolone	3
Pharmaceutical	Steroids and Hormones	Methyl-testosterone	3
Pharmaceutical	Steroids and Hormones	Nandrolone	3
Pharmaceutical	Steroids and Hormones	Pregnenolone	3
Pharmaceutical	Steroids and Hormones	Progesterone	1, 3
Pharmaceutical	Steroids and Hormones	Testosterone	3
Pharmaceutical	Steroids and Hormones	Testosterone isocapronaat	3
Pharmaceutical	Steroids and Hormones	Testosterone phenylpropionate	3
Pharmaceutical	Vitamins	Vitamin A	3
Pharmaceutical	Vitamins	Vitamin B1	3
Pharmaceutical	Vitamins	Vitamin B12	3
Pharmaceutical	Vitamins	Vitamin B2	3
Pharmaceutical	Vitamins	Vitamin B3	3
Pharmaceutical	Vitamins	Vitamin B5	3
Pharmaceutical	Vitamins	Vitamin B6	3
Pharmaceutical	Vitamins	Vitamin B9	3
Pharmaceutical	Vitamins	Vitamin C	3
Pharmaceutical	Vitamins	Vitamin D2	3
Pharmaceutical	Vitamins	Vitamin E	3
Pharmaceutical	Vitamins	Vitamin K	3
Pharmaceutical research	Antibiotics	Ampicillin	1
Pharmaceutical research	Antibiotics	Chlortetracyclin	1
Pharmaceutical research	Antibiotics	Doxycycline	1
Pharmaceutical research	Antibiotics	Erythromycin	1
Pharmaceutical research	Antibiotics	Metronidazole	1
Pharmaceutical research	Antibiotics	Oxolinic Acid	1
Pharmaceutical research	Antibiotics	Sulfadiazine	1
Pharmaceutical research	Antibiotics	Sulfamethoxazole	1
Pharmaceutical research	Antibiotics	Sulfaquinoxaline	1

Pharmaceutical research	Antibiotics	Trimethoprim	1
Pharmaceutical research	Medicine	1-Hydroxy Ibuprofen	1
Pharmaceutical research	Medicine	Acetaminophen	1
Pharmaceutical research	Medicine	Amiodarone	1
Pharmaceutical research	Medicine	Carbamazepine	1
Pharmaceutical research	Medicine	Carboxyibuprofen	1
Pharmaceutical research	Medicine	Clofibric acid	1
Pharmaceutical research	Medicine	Cytarabine	1
Pharmaceutical research	Medicine	Dihydrocodeine	1
Pharmaceutical research	Medicine	DTPA	2
Pharmaceutical research	Medicine	Furosemide	1
Pharmaceutical research	Medicine	Hydrochlorothiazide	1
Pharmaceutical research	Medicine	Ibuprofen	1, 2
Pharmaceutical research	Medicine	Metformin	1
Pharmaceutical research	Medicine	Morphine	1
Pharmaceutical research	Medicine	Oxazepam	1
Pharmaceutical research	Medicine	Phenazone	1
Pharmaceutical research	Medicine	Propranolol	1
Pharmaceutical research	Medicine	Sotalol	1
Pharmaceutical research	Medicine	Trimetazidine	1
Pharmaceutical research	Pesticides	2,6-Dichlorobenzamide	1
Pharmaceutical research	Pesticides	2H-Atr	1
Pharmaceutical research	Pesticides	3,4-dichloroaniline	1
Pharmaceutical research	Pesticides	Amprolium	1
Pharmaceutical research	Pesticides	Chloropicrin	1
Pharmaceutical research	Pesticides	DEA	1
Pharmaceutical research	Pesticides	DEDIA	1
Pharmaceutical research	Pesticides	DIA	1
Pharmaceutical research	Pesticides	Dichlorophen	1
Pharmaceutical research	Pesticides	Diuron	1
Pharmaceutical research	Pesticides	Ethylenethiourea	1
Pharmaceutical research	Pesticides	N,N-Diethyl-m-toluamide	1
Pharmaceutical research	Steroids and Hormones	17alpha-Estradiol	1
Pharmaceutical research	Steroids and Hormones	Androstenedione	1
Soap	Other Industrial Compounds	Caustic potash	4
Soap	Other Industrial Compounds	Sodium hydrogen sulfite	4
Textile	Chemical cleaning	Disodium phosphate	5
Textile	Chemical cleaning	Sodium dichromate	5
Textile	Chemical cleaning	Sodium hydroxide	4, 5
Textile	Chemical cleaning	Sodium hypochlorite	5
Textile	Dyes and paints	1-Naphthylamine	4
Textile	Dyes and paints	Ferric ferrocyanide	4
Textile	Dyes and paints	O-Methylaniline	1
Textile	Dyes and paints	Orthophenylphenol	4
Textile	Dyes and paints	P-Methylaniline	1
Textile	Dyes and paints	Sodium chlorate	4
Textile	Dyes and paints	Sodium chloride	3, 4
Textile	Dyes and paints	Sodium nitrite	4
Textile	Flame retardants	Ammonium bromide	4
Textile	Flame retardants	Bromates	1, 4
Textile	Other Industrial Compounds	Acetaldehyde	1
Textile	Other Industrial Compounds	Acrylonitrile	1
Textile	Other Industrial Compounds	Chloroacetic acid	1
Textile	Other Industrial Compounds	Chloronitrobenzene-1,2	1
Textile	Other Industrial Compounds	Chloronitrobenzene-1,3	1
Textile	Other Industrial Compounds	Chloronitrobenzene-1,4	1
Textile	Other Industrial Compounds	Dichloronitrobenzene-2,3	1
Textile	Other Industrial Compounds	Dichloronitrobenzene-2,4	1
Textile	Other Industrial Compounds	Dichloronitrobenzene-2,5	1
Textile	Other Industrial Compounds	Dichloronitrobenzene-3,4	1
Textile	Other Industrial Compounds	EDTA	2
Textile	Other Industrial Compounds	Formaldehyde	1
Textile	Other Industrial Compounds	Free cyanide	1
Textile	Other Industrial Compounds	Glycerine	4, 5
Textile	Other Industrial Compounds	Isoquinoline	1
Textile	Other Industrial Compounds	Nitrilotriacetic acid	1
Textile	Other Industrial Compounds	Nitrobenzene	1
Textile	Plasticizers	Bisphenol A	1, 2
Textile	Plasticizers	Triphenyl phosphate	2

Textile	Solvent	1,4-Dioxane	2
Textile	Textile finishing	Dibutyltin (cation)	1
Textile	Textile finishing	Lead nitrate	4
Textile	Textile finishing	Magnesium chloride	4
Textile	Textile finishing	Methylene diphenyl diisocyanate	4
Textile	Textile finishing	Polyurethane	5
Textile	Textile finishing	Potassium hypochlorite	4
Textile	Textile finishing	PVC	4, 5
Textile	Textile finishing	Styrene-butadiene	5
Textile	Textile finishing	Toluene diisocyanate	4
Textile	Textile finishing repellent	PFDA	1
Textile	Textile finishing repellent	PFDoA	1
Textile	Textile finishing repellent	PFDS	1
Textile	Textile finishing repellent	PFHpA	1
Textile	Textile finishing repellent	PFHS	1
Textile	Textile finishing repellent	PFHxA	1
Textile	Textile finishing repellent	PFNA	1
Textile	Textile finishing repellent	PFOA	1
Textile	Textile finishing repellent	PFOS	1, 2
Textile	Textile finishing repellent	PFOsA	1
Textile	Textile finishing repellent	PFUn	1

ANNEX 2 – LIST OF EXCLUDED SUBSTANCES

Table 10 List of substances excluded from further research by product type, including reason of exclusion.

Sources: 1 Lopez et al, 2015; 2 Klein & Duijnhoven, 2013; 3 Organon, 1957; 4 Nieuwkoop, 1993; 5 DOE, 1995a, 1995b & 1996

Product type	Name	Reason Exclusion	Source
Industrial Compounds	Antimony	Researched by NIPA	5
Industrial Compounds	Chloronitrobenzene-1,2	Researched by NIPA	1
Industrial Compounds	Chloronitrobenzene-1,3	Researched by NIPA	1
Industrial Compounds	Chloronitrobenzene-1,4	Researched by NIPA	1
Industrial Compounds	Dichloronitrobenzene-2,3	Researched by NIPA	1
Industrial Compounds	Dichloronitrobenzene-2,4	Researched by NIPA	1
Industrial Compounds	Dichloronitrobenzene-2,5	Researched by NIPA	1
Industrial Compounds	Dichloronitrobenzene-3,4	Researched by NIPA	1
Industrial Compounds	Dinitrotoluene-2,4	Spatial exclusion	1
Industrial Compounds	Dinitrotoluene-2,6	Spatial exclusion	1
Industrial Compounds	Nitrobenzene	Researched by NIPA	1
Industrial Compounds	Trinitrotoluene	Spatial exclusion	1
Industrial Compounds	1,1,1-Trichloroethane	Researched by NIPA	5
Industrial Compounds	Naphthalene	Researched by NIPA	5
Industrial Compounds	PCE / TCE	Researched by NIPA	5
Industrial Compounds	Toluene	Researched by NIPA	4, 5
Industrial Compounds	Trichloroethene	Researched by NIPA	5
Industrial Compounds	Xylene	Researched by NIPA	4, 5
Industrial Compounds	1,2,3,4,6,7,8,9-OCDD	>1970	1
Industrial Compounds	1,2,3,4,6,7,8,9-OCDF	>1970	1
Industrial Compounds	1,2,3,4,6,7,8-HpCDD	>1970	1
Industrial Compounds	1,2,3,4,6,7,8-HpCDF	>1970	1
Industrial Compounds	1,2,3,4,7,8,9-HpCDF	>1970	1
Industrial Compounds	1,2,3,4,7,8-HxCDD	>1970	1
Industrial Compounds	1,2,3,4,7,8-HxCDF	>1970	1
Industrial Compounds	1,2,3,6,7,8-HxCDD	>1970	1
Industrial Compounds	1,2,3,7,8,9-HxCDD	>1970	1
Industrial Compounds	1,2,3,7,8,9-HxCDF	>1970	1
Industrial Compounds	1,2,3,7,8-PeCDF	>1970	1
Industrial Compounds	2,3,4,6,7,8-HxCDF	>1970	1
Industrial Compounds	2,3,7,8-TCDD	>1970	1
Industrial Compounds	2,3,7,8-TCDF	>1970	1
Industrial Compounds	2,4,5-Trichloroaniline	>1970	1
Industrial Compounds	2-nitrotoluene	>1970	1
Industrial Compounds	Anthracene	Researched by NIPA	1
Industrial Compounds	BDE-99	>1970	1
Industrial Compounds	Benzene	Researched by NIPA	4, 5
Industrial Compounds	DBP	Researched by NIPA	1
Industrial Compounds	DEHP	>1970	1, 2
Industrial Compounds	DEP	Researched by NIPA	1, 2
Industrial Compounds	Dibromochloromethane	>1970	1
Industrial Compounds	Dichlorophenol-2,3	>1970	1, 2
Industrial Compounds	Dichlorophenol-3,5	>1970	1, 2
Industrial Compounds	hxCDD	>1970	1
Industrial Compounds	NBBS	>1970	1
Industrial Compounds	PCB 31	>1970	1, 4
Industrial Compounds	Phenol	Researched by NIPA	4
Industrial Compounds	TCPP	>1970	2, 4, 5
Industrial Compounds	Tetrachlorobenzene	Researched by NIPA	1, 4
Industrial Compounds	Tin	Researched by NIPA	4
Industrial Compounds	Tolyltriazole	>1970	1
Industrial Compounds	Trichloroaniline-2,4,6	>1970	1
Industrial Compounds	Trichlorobenzene-1,2,4	Researched by NIPA	1
Organon products	Copper	Researched by NIPA	3
Organon products	DDT	Researched by NIPA	3
Life Style Products	Cocaine	Spatial exclusion	1
Life Style Products	Cotinine	Spatial exclusion	1
Life Style Products	Propylparaben	Spatial exclusion	1
Other Emerging Contaminants	Musk ketone	Spatial exclusion	1
Other Emerging Contaminants	1,7-Dimethylxanthine	>1970	1
Other Emerging Contaminants	Galaxolide	>1970	1
Other Emerging Contaminants	Ioxitalamic acid	>1970	2

Other Emerging Contaminants	Monobutyltin (cation)	>1970	1
Other Emerging Contaminants	Musk ambrette	>1970	1
Other Emerging Contaminants	Musk xylene	>1970	1
Other Emerging Contaminants	Octocrylene	>1970	1
Other Emerging Contaminants	Sucralose	>1970	1
Pesticides	2,4-D-ester	>1970	1
Pesticides	2-Chlorobenzoic acid	>1970	1
Pesticides	3,4-DCPMU	>1970	1
Pesticides	Acetamiprid	>1970	1
Pesticides	Atrazine	Researched by NIPA	1
Pesticides	Bentazone	>1970	1
Pesticides	Boscalid	>1970	1
Pesticides	Carbosulfan	>1970	1
Pesticides	Chlortoluron	>1970	1
Pesticides	Coumafene	>1970	1
Pesticides	Desmethylisoproturon	>1970	1
Pesticides	Didemethylisoproturon	>1970	1
Pesticides	Dimethenamid	>1970	1
Pesticides	Dinocap	>1970	1
Pesticides	Florasulam	>1970	1
Pesticides	Fluazinam	>1970	1
Pesticides	Flupyrsulfuron methyl	>1970	1
Pesticides	Imazalil	>1970	1
Pesticides	Imazamox	>1970	1
Pesticides	Imazapyr	>1970	1
Pesticides	Isoproturon	>1970	1
Pesticides	Isoxadifen-ethyl	>1970	1
Pesticides	mepiquat	>1970	1
Pesticides	Mesosulfuron methyle	>1970	1
Pesticides	Metolachlor	>1970	1
Pesticides	Metsulfuron-methyl	>1970	1
Pesticides	Oxadixyl	>1970	1
Pesticides	Permethrine cis	>1970	1
Pesticides	Propiconazole	>1970	1
Pesticides	Propyzamide	>1970	1
Pesticides	Prosulfuron	>1970	1
Pesticides	Simazine	Researched by NIPA	1
Pharmaceutical Products	4-Chlorobenzoic acid	>1970	1
Pharmaceutical Products	Amlodipine	>1970	1
Pharmaceutical Products	Atenolol	>1970	1
Pharmaceutical Products	Buflomedil	>1970	1
Pharmaceutical Products	Carbamazepine epoxide	>1970	1
Pharmaceutical Products	Ciprofloxacin	>1970	1
Pharmaceutical Products	Clarithromycin	>1970	1
Pharmaceutical Products	Desvenlafaxine	>1970	1
Pharmaceutical Products	Diclofenac	>1970	1
Pharmaceutical Products	Enrofloxacin	>1970	1
Pharmaceutical Products	Fenbendazole	>1970	1
Pharmaceutical Products	Fenofibric acid	>1970	1
Pharmaceutical Products	Flumequine	>1970	1
Pharmaceutical Products	Fluoxetine	>1970	1
Pharmaceutical Products	Gemfibrozil	>1970	1
Pharmaceutical Products	Gestodene	>1970	1
Pharmaceutical Products	Imatinib	>1970	1
Pharmaceutical Products	Ivermectin	>1970	1
Pharmaceutical Products	Ketoprofen	>1970	1
Pharmaceutical Products	Losartan	>1970	1
Pharmaceutical Products	Metoprolol	>1970	1
Pharmaceutical Products	O-Desmethyltramadol	>1970	1
Pharmaceutical Products	Ofloxacin	>1970	1
Pharmaceutical Products	Simvastatin	>1970	1
Pharmaceutical Products	Tramadol	>1970	1

ANNEX 3 – EXTREME SURFACE WATER LEVEL EVENTS

Table 11 Low (<) and high (>) surface water level events of the Meuse between 1980 and 1995 at monitoring point Oijen. Data from (Rijkswaterstaat, 2015).

	Month	**Nr. of days**	**Extreme level**	**Average level**
Low water events				
1	feb-80	6	398	436
2	jan-81	2	380	387
3	jan-81	7	375	418
4	mrt-81	5	410	431
5	dec-81	16	360	400
6	jan-82	8	386	436
7	aug-82	4	445	448
8	dec-82	10	359	386
9	feb-83	4	376	391
10	apr-83	6	352	367
11	mei-83	6	383	404
12	**jan-84**	**12**	**352**	**412**
13	feb-84	2	382	408
14	nov-84	4	381	424
15	**jan-85**	**7**	**47**	**191**
16	**mrt-85**	**29**	**445**	**450**
17	mei-85	3	450	452
18	jan-86	10	395	417
19	apr-86	4	386	414
20	jan-87	8	192	340
21	mrt-87	6	413	443
22	mrt-87	5	393	423
23	**feb-88**	**23**	**408**	**430**
24	mrt-88	2	423	437
25	mrt-88	9	397	426
26	dec-88	3	409	414
27	feb-90	4	409	419
28	jan-91	6	362	395
29	jan-91	2	411	429
30	dec-91	4	398	431
31	dec-92	4	383	392
32	jan-93	4	427	438
33	dec-93	8	388	410
34	jan-94	5	391	418
35	dec-94	6	411	431
36	jan-95	2	435	442
37	feb-95	2	442	444
38	feb-95	10	377	411
High water events				
1	Jul-80	2	534	526
2	Feb-84	7	624	578
3	Mar-88	5	573	553
4	Jan-91	4	537	527
5	Jan-93	3	541	528
6	Dec-93	14	**650**	**566**
7	**Jan-95**	**12**	**679**	**613**

ANNEX 4 – GROUNDWATER FLOW MODEL INPUT

Table 12 Input data varying per time periods

	1	**2**	**3**	**4**	**5**	**6**	**H7**	**L12**	**L15**	**L16**	**H1**	**L7**	**L11**
Season	Summer	Winter	Summer	Winter	Summer	Winter	Winter	Winter	Winter	Winter	Summer	Summer	Summer
Year span	1951-1959	1951-1959	1960-1998	1960-1998	1998-present	1998-present	1960-1998	1960-1998	1960-1998	1960-1998	1960-1998	1960-1998	1960-1998
Meuse	4.56	4.6	4.92	4.89	4.9	4.9	6.79	3.52	0.47	4.45	5.34	4.45	3.83
H0	16.56	16.6	16.92	16.89	16.9	16.9	18.79	15.52	12.47	16.45	17.34	16.45	15.83
Polder	4.4	4.1	4.4	4.1	4.2	3.8	4.1	4.1	4.1	4.1	4.4	4.4	4.4
Ha	16.4	16.1	16.4	16.1	16.2	15.8	16.1	16.1	16.1	16.1	16.4	16.4	16.4
Wetering	4.3	3.8	4.3	3.8	4.3	3.8	3.8	3.8	3.8	3.8	4.3	4.3	4.3
Hl	16.3	15.8	16.3	15.8	16.3	15.8	15.8	15.8	15.8	15.8	16.3	16.3	16.3
C1	-5.9E-06	-18E-05	-5.9E-06	-1.8E-05	5.9E-06	-3.8E-09	-1.8E-05	-1.8E-05	-1.8E-05	-1.8E-05	-5.9E-06	-5.9E-06	-5.9E-06
C2	0.16	0.50	0.52	0.79	0.70	1.1	2.69	-0.58	-3.63	0.35	0.94	0.05	-0.57

Distance	**Heads [m]**												
50	16.53	16.49	16.81	16.72	16.75	16.66	18.21	15.65	13.25	16.37	17.14	16.44	15.95
150	16.48	16.34	16.65	16.48	16.54	16.33	17.40	15.82	14.35	16.27	16.85	16.42	16.13
250	16.45	16.25	16.55	16.33	16.41	16.13	16.90	15.93	15.03	16.20	16.68	16.41	16.23
350	16.43	16.19	16.49	16.24	16.33	16.00	16.59	15.99	15.44	16.16	16.57	16.41	16.30
450	16.42	16.16	16.46	16.19	16.28	15.92	16.40	16.04	15.69	16.14	16.50	16.41	16.34
550	16.41	16.13	16.44	16.15	16.25	15.88	16.28	16.06	15.85	16.12	16.46	16.40	16.36
650	16.41	16.12	16.42	16.13	16.23	15.85	16.21	16.08	15.95	16.11	16.44	16.40	16.38
750	16.40	16.11	16.41	16.12	16.22	15.83	16.17	16.08	16.01	16.11	16.42	16.40	16.39
850	16.40	16.11	16.41	16.11	16.21	15.82	16.14	16.09	16.04	16.10	16.41	16.40	16.39
950	16.40	16.10	16.40	16.11	16.21	15.81	16.12	16.09	16.06	16.10	16.41	16.40	16.39
1050	16.40	16.10	16.40	16.10	16.21	15.81	16.11	16.09	16.08	16.10	16.40	16.40	16.40
1150	16.40	16.10	16.40	16.10	16.20	15.80	16.11	16.09	16.08	16.10	16.40	16.40	16.40
1250	16.40	16.09	16.40	16.09	16.20	15.80	16.10	16.09	16.08	16.09	16.40	16.40	16.40
1350	16.40	16.09	16.40	16.09	16.21	15.80	16.09	16.09	16.08	16.09	16.40	16.40	16.39
1450	16.39	16.08	16.39	16.08	16.21	15.80	16.08	16.08	16.08	16.08	16.39	16.39	16.39
1550	16.39	16.07	16.39	16.07	16.21	15.80	16.07	16.07	16.06	16.07	16.39	16.39	16.39
1650	16.38	16.05	16.38	16.05	16.22	15.80	16.05	16.05	16.04	16.05	16.38	16.38	16.38
1750	16.37	16.01	16.37	16.01	16.23	15.80	16.01	16.01	16.01	16.01	16.37	16.37	16.37
1850	16.35	15.96	16.35	15.96	16.25	15.80	15.96	15.96	15.96	15.96	16.35	16.35	16.35
1950	16.32	15.86	16.32	15.86	16.28	15.80	15.86	15.86	15.86	15.86	16.32	16.32	16.32