Universiteit Utrecht

Biofiltrating the air, one breath at a time

Substantiating the optimal biofiltration system to remove pollutants from the ventilated air of buildings

Writing assignment

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Abstract

Due to worldwide industrialization, air in the atmosphere became contaminated with pollutants. Hazardous for nature and humans, the WHO has determined the most prevalent and significant to be VOCs, methane, CO_2, NO_2 and SO_2. Indoor air pollution has traditionally received less attention than outdoor air pollution. However, indoor air pollution may be more relevant, as people on average spend approximately 90% of their time indoors. Due to accumulation and long-time exposure, indoor air pollution can lead to health implications such as respiratory- and neurodegenerative diseases. This research focuses on improving indoor air quality by purifying air that enters a building using biofiltration. The current application of biofiltration is to remove pollutants from exhaust gas of polluting instances. In this study, this process was reversed so that incoming air in buildings is purified from pollutants. Using literature, optimal microbial consortia, design of biofilter and packing material were substantiated. The final concept is a biotrickling filter with two consecutive chambers filled with ceramic beads. The first chamber breaks down VOCs and methane, while the second chamber removes NO_2, SO_2 and CO_2. The operating settings can be optimized for local concentrations of pollutants. This was the first study exploring the utilization of biofilters for purifying air entering buildings, and hopefully future research makes the system more efficient and affordable for the general public. When applied on a large scale, every single building would serve as an atmospheric cleaning station, simultaneously improving health for residents and preventing further climate change.

Layman summary

For the past centuries, exhaust gasses from factories, vehicles, and power plants have polluted the air. This affects nature in the form of climate change and humans because we breathe foul air. We mostly breathe this air inside buildings, where we spend on average 90% of our time. Because many buildings are poorly ventilated, the polluted air stays inside and people inhale a big part of it. Inhaling foul air for a long time eventually leads to diseases in the brain or lungs. A solution would be to remove the pollutants from the air entering buildings. This study focuses on doing this in a sustainable way. To filter the air sustainably, nature is studied and copied. Some bacteria and fungi break down pollutants from the air to get energy. These bacteria and fungi have been placed in air filters that remove pollutants from exhaust gasses of factories, called biofilters. For the goal of this study, this principle is reversed. When a biofilter with bacteria and fungi is installed at buildings, they clean the air that ventilates into the building. Using literature from previous research, all aspects of a biofilter were looked at so that an optimal version could be constructed. The type of filter that was chosen for the design is a biotrickling filter. In this filter, water and nutrients are continuously trickled down on bacteria and fungi so that they can degrade pollutants. The bacteria and fungi are surrounded by layers of sugars. The organisms together with these layers are called a biofilm. In a biofilm, the pollutants are degraded into simpler and less harmful molecules. The biofilm is attached to a material that fills the chamber and serves as a frame through which air flows. In previous research comparing different materials, it was found that ceramic beads support the biofilm and its functioning best. However, when the biofilm becomes too thick, the air is unable to pass through the filter. To prevent the biofilm from growing too much, a natural predator is added to the filter. A species of mosquitoes, *Bradysia odoriphaga*, is added to the biotrickling filter. By moving through the biofilm, it loosens and falls down. The biofilter also has many settings that can be changed to have the best configuration for a specific place. As the amount of pollutants in the air differs around the world, the biofilter has to be able to remove different mixtures of gas. By changing settings, the biofilter can operate at a high level at different places in the world. Examples of these settings are the time a gas stays in the biofilter and the temperature of the biofilter. Using previous research, settings for the startup of the biotrickling filter are made. In the process of creating the final design, the ability to clean the air was seen as more important than the cost of the biofilter. Therefore, the current design is most interesting for companies with a large budget to make their business more sustainable. By using this biofilter, the air quality inside a building improves, and pollutants are removed from the air outside. When the biofilter is distributed on a big scale, every building contributes to cleaning the air.

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Introduction

Starting in the late 18th century, industrialization in Europe led to heavily increased combustion of coal, producing high emissions of smoke and pollutants (Fowler et al., 2020). Until the 1950s, the consequences of these emissions were accepted by everyone. These included reduced visibility, erosion, and damage to natural ecosystems. Halfway through the 20th century, scientists found more consequences of air pollution; forest decline and eutrophication being the most significant. Research in the decades after found that increased levels of greenhouse gasses (GHGs) induced climate change, threatening to cause irreversible changes in ecosystems around the world (Abas et al., 2017).

However, not only nature is affected by pollution of the atmosphere. As early as 400 BC, people knew about the health implications of breathing smoke, as the ancient Greek doctor Hippocrates wrote about places where the atmosphere was “impure and unhealthy” (Jones et al., 1923). In 1273, the first legislation in England came about in the Smoke Abatement Act, in which the use of coal was prohibited because it was ‘prejudicial to health’ (Heidorn, 1978). However, for a long time, an exact causal relationship between air pollution and illness could not be found. This changed in 1952 during the Great London Smog, in which approximately 12,000 people died (Polivka, 2018). Research in the decades after substantiated the causal relationship between air pollution and illness, as air pollution affects our health in two ways. First, combustion produces particles that lead to respiratory diseases (WHO, 2021), diabetes (Thiering & Heinrich., 2015) and neurodegenerative diseases (Xu et al., 2016). Second, pollutants can add to climate change which in turn leads to more extreme drought and food scarcity, for example. Thus, air pollution can damage us directly by inducing sickness and indirectly by accelerating climate change. This brings us to the point at which we are now, with up to 7 million premature annual deaths due to air pollution (Orru et al., 2017).

The biggest pollutants for climate change are CO_2, methane and nitrous oxide (AK-BHD, 2021), and the major pollutants directly for our health are particulate matter, sulfur dioxide, ozone, nitrogen dioxide and carbon monoxide (World Health Organization, 2021). Another group of gasses that affects climate change, as well as our health, are volatile organic compounds (VOCs) (Montero-Montoya et al., 2018). These gasses are released in the atmosphere by biogenic or anthropogenic activities, have a long mean lifetime and can cause symptoms that may induce pathologies including neurologic diseases, asthma, and cancer (Montero-Montoya et al., 2018). Besides this, it contributes to the formation of particulate matter and ozone, making it a dual threat.

Knowing all the aspects of the problem and what triggers it, scientists came up with two solutions: reducing emission of pollutants and removing pollutants from the atmosphere. It seems only logical to first stop adding more pollutants to the atmosphere before removing them. However, with the industrialization and urbanization of development countries, it remains a struggle to lower emissions. Still, this is a problem we have to fix ourselves, as we have created it ourselves. The second solution is to remove pollutants from the atmosphere and contrary to the first one, nature already does this (Devinny et al., 2017). Learning from nature, this inspired the creation of biofilters. These devices remediate soil, waste water or air by relying on the metabolism of organisms like bacteria and fungi.

These microorganisms can break down, take in or modify the hazardous compounds into less toxic substances. At first, compost was used for biofiltration in which the mixture of microorganisms was not known. As more research was done on microorganisms, it became clear what kinds of toxins they can degrade. This led to the development of biofilters with mixtures of microorganisms specific to its application, for instance for the exhaust gas from paint factories, landfill sites and pharmaceutical industry (Ferdowsi et al., 2022). By doing so, the emissions of pollutants decrease, but it does not lower the concentration of pollutants already present in the atmosphere.

As humans on average spend approximately 90% of their time indoors, indoor air quality has a big impact on their health (Ohura et al., 2006). Current ventilation transfers polluted air indoors and products as paint and cleaning chemicals excrete additional pollutants inside buildings, aggravating indoor air quality even more.

The current practice of biofiltration is to prevent toxic substances from entering the atmosphere through the exhaust of contaminating instances. However, as illustrated earlier, to prevent further climate change, we also ought to remove the particles already present in the air. These particles not only boost climate change, but also impact our health, with the most exposure in the inside environment. If biofiltration was to be used in the opposite way, by preventing toxic substances from the outside entering inside, the concentration of pollutants in the atmosphere would lower and the indoor air quality would increase. By switching the current principle of biofiltration, one would kill two birds with one stone, or two pollutants with one filter. As the knowledge of microorganisms and their metabolism has increased greatly, a filter could be assembled that removes the most hazardous pollutants from the atmosphere and prevents them from entering buildings. This led to the following research question:

What is the optimal biofilter for removing pollutants entering buildings through ventilation?

A literature study will be done to answer the research question. The first step will be to dive into air pollution and the specific toxins harmful for humans and nature. Using this information, microorganisms capable of degrading these specific target pollutants are searched so that a consortium can be constructed. Literature regarding specifics and efficiency of different types of biofilters will provide the information needed to choose the most suitable one. Alterations and modifications that would increase the efficiency of the biofilter are then explored. Finally, the operating conditions are looked at, from settings to maintenance. Every piece of information will then contribute to a final conceptual design that answers the research question.

Chapter 1 Biofiltration of indoor pollutants

Biofiltration is the cleaning of air using organisms and one of the oldest bioremediation techniques (Srivastava et al., 2021). Bioremediation is defined as “…the process whereby wastes are biologically degraded under controlled conditions to an innocuous state, or to levels below concentration limits established by regulatory authorities.” (Mueller et al., 1996). It primarily uses microorganisms to break down local contaminants into less toxic products. The first use of biofiltration stems from 1923, when H. Back used it to limit hydrogen sulfide emission from wastewater treatment plants using a soil bed filled with microorganisms (Leson & Winer, 1991). The technique gained more attention and since 1950 it has been used for the filtration of off-gasses (Marycz et al., 2022). The first models consisted of open spaces filled with soil, through which air from perforated pipes were pumped. Because of this configuration, these systems required lots of space due to their low specific activity of microorganisms and had many disadvantages such as acidification, uneven air distribution and drying out. Continuous research led to the models we have today, which will be discussed shortly. All models have a couple of requirements: a chamber in which the gas flows, microorganisms that break down contaminants and nutrients for the microorganisms to function properly. However, the different biofilters do differ in their configuration and mechanism. In order to come to the optimal biofilter for removing toxins that enter buildings, first the most prevalent indoor pollutants will be discussed.

Indoor pollutants

Indoor air quality has not been getting the same amount of attention as outdoor air quality has in respect to health. However, the concentration of air pollution indoors is often higher than outside due to accumulation and long-time exposure to pollutants (EPA, 2020). Complications of long-term exposure to bad indoor air quality has become more apparent in the last decades. In order to save money on heating, buildings became more sealed to the outside, leading to poor ventilation (EEA, 2019). The WHO listed the most hazardous pollutants for human health with dangerous concentrations in the atmosphere, which will now be discussed.

Ozone (O_3) is primarily known for its protective ability in the ozone-layer, but at ground-level, it has an opposite effect (Chen et al., 2007). Here it is a major component of smog and formed by photochemical reactions with other pollutants. Long-time or excessive exposure causes asthma, problems with breathing and even lung diseases. As a contaminant, ozone is hard to break down, but its precursors are more easily degraded. That is why this paper from here on focuses on the abatement of precursors of ozone. The main precursors of ozone are VOCs, nitrogen dioxide and methane.

Volatile organic compounds (VOCs) are released primarily by anthropogenic activities and, as explained in the introduction, are dangerous due to their long mean lifetime and toxicity (Montero-Montoya et al., 2018). Long-time exposure to these chemicals, even in low concentrations, can lead to neurotoxicity, immunotoxicity and reproductive toxicity, especially in children who still need to develop (Grandjean et al., 2019). The substances can be categorized into hydrophilic- and hydrophobic groups, and in addition to their toxicity for humans, they also contribute to the formation of particulate matter and ozone (Fuller et al., 2022). The most prevalent VOCs are benzene, toluene, ethylbenzene and xylene (BTEX) (Davidson et al., 2021). The local BTEX concentration is often used to measure the level of toxicity by VOCs in the atmosphere.

Nitrogen dioxide (NO_2) is emitted from fossil fuel burning in vehicles, heating and power generation (WHO, 2021). Exposure can initiate and aggravate respiratory diseases, even at low concentrations (Chen et al., 2007). Children and elderly are mostly vulnerable to its adverse health effects. It can also react with other air pollutants to produce the toxic ground-level ozone.

Methane (CH_4) was responsible for 45% of the net atmospheric warming effect of all anthropogenic activities and approximately constitutes a third of the green-house effect from all exhaust mixed gasses (Kuylenstierna et al., 2021). It also is one of the main precursors to ground level ozone.

Sulfur dioxide (SO_2) comes from the combustion of coal and oil (Chen et al., 2007). Main sources are coal-fired power plants, cement factories and transportation. It has been thought that it catalyzes pre-existing cardiovascular- and initiates respiratory diseases. Additionally, the oxidation of SO_2 plays an important role in global atmospheric chemistry leading to haze pollution, acid precipitation and climate change (Liu et al., 2021).

Particulate matter (PM) are solids or liquids in the air and come in different sizes: PM_{10} has a width of 10 μm or less and $PM_{2.5}$ a width of 2.5 μm or less. The more coarse PM_{10} can irritate airways, but $PM_{2.5}$ can diffuse through lungs into the bloodstream (Atafar et al., 2019). A component of $PM_{2.5}$ is black carbon, or otherwise called soot. These particles have a global warming potential that is 460-1500 times higher than CO_2 (Kuylenstierna et al., 2021). However, biofilters that use microorganisms are not able to degrade PM, in contrast to biofilters using plants. A small part of PM is absorbed in the biofilm, but this is not substantial enough to rule out health consequences.

Now that the most hazardous indoor pollutants are clear, the three types of bioreactors will be discussed and compared in their abilities to break down these pollutants.

Types of biofilters

Conventional biofilter

The first biofiltration system has a roster or bed with organic materials in which the microorganisms reside (Delhoménie et al., 2005). Air is pumped through the bed and the immobilized microorganisms degrade the contaminants. The ascended air is then clean and transferred away. A solution with nutrients is occasionally sprayed on the bed to provide the microorganisms with vital micro- and macronutrients (see Figure 1).

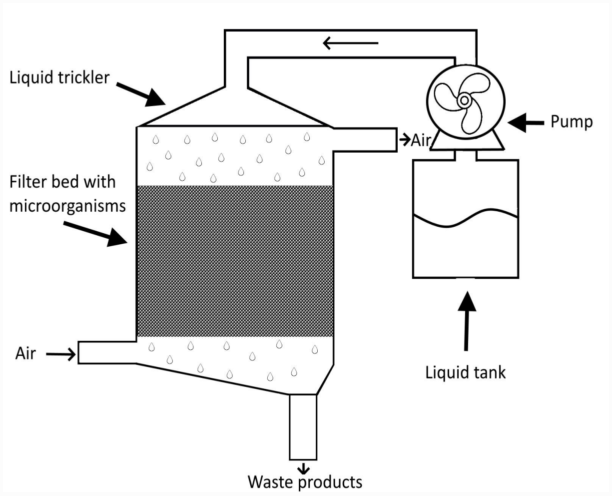

Figure 1: Schematic of different components and airflow of a conventional biofilter. Source: own

Bioscrubber

The second biofiltration system is called a bioscrubber. Contaminated gas is still pumped upwards, but transferred into a liquid phase, or ‘aerosol’ (Delhoménie et al., 2005). The droplets containing the pollutants are pumped into a separate bioreactor, containing the degrading microorganisms. The microorganisms are suspended in the liquid, together with vital nutrients (see Figure 2). Bioscrubbers are most often using microorganisms in sludge retrieved from wastewater treatment plants. The residence time for aqueous solutions in bioreactors range between 20 and 40 days (Delhoménie et al., 2005).

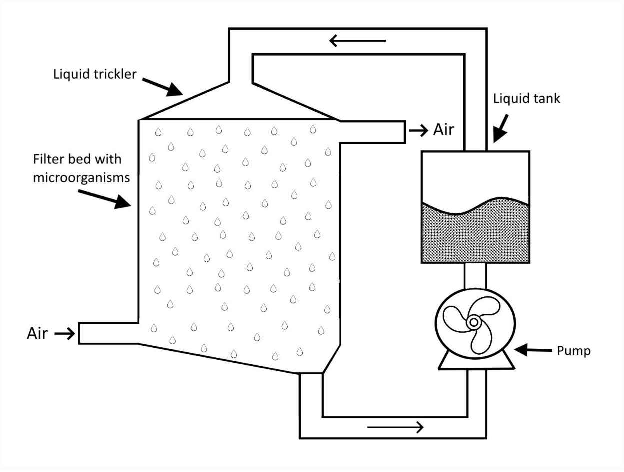

Figure 2: Schematic of different components and airflow of a bioscrubber. Source: own

Biotrickling filter

The final type of biofilter is a combination of the prior two configurations (see Figure 2), as it has an inert bed with inoculated microorganisms while an aqueous solution is continuously sprinkled down (Delhoménie et al., 2005). This aqueous solution contains nutrients and provide moisture, vital for growth and maintenance of microorganisms. The difference with the conventional biofilter is that the material of the inert bed can be chosen. It often consists of a type of plastic or ceramic, on which organisms grow and air is able to pass through. Contaminants need to transfer into the liquid phase and then the biofilm. Arrived here, they are degraded after some time. The continuous sprinkling of the nutrients allow close control of the pH, end product removal and temperature.

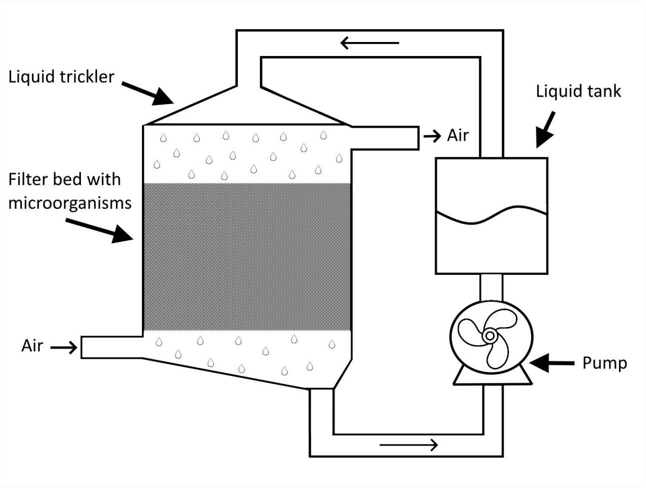

Figure 3: Schematic of different components and airflow of a biotrickling filter. Source: own

Comparison of the bioreactors

Now, the different types of bioreactors will be compared on a variety of criteria. These criteria are grouped into the ability to break down contaminants, and the operation efficiency and practicality. The first group of criteria is most important, as this determines the rate at which air is cleaned before entering a building. The second group looks at the practical aspects of the biofilter so that it has the potential to be widely applicable. Both of them have a subset of aspects which will now be discussed.

Ability to break down contaminants

A mixture of microorganisms determines what kind of contaminants can be broken down in a biofilter (Sonil et al., 2012). Every microbe has its specific metabolism, and a mixture of microbes is called a consortium. The setup and mechanism of a biofilter affects the performance of the biofilter greatly. This determines the removal efficiency of organic hydrophilic-, organic hydrophobic- and inorganic compounds from a gas. The setup and mechanism also determines at what concentrations the biofilter is able to break down contaminants. Lastly, the rate at which pollutants are transferred from the gas into the biofilm, is called mass transfer and differs between types of biofilters.

Table 1: Comparison of three types of biofilters on their degradation ability. Source: Gospodarek et al., 2019.

Function	**Ability**	**Conventional biofilter**	**Bioscrubber**	**Biotrickling filter**
Ability to break down contaminants	inoculate specific microorganisms	no	yes	yes
	degrade organic hydrophilic compounds	moderate	bad	very good
	degrade organic hydrophobic compounds	good	bad	good
	degrade inorganic compounds	moderate	good	good
	suitable concentration contaminants	low	high	moderate
	mass transfer	high	high	low

Operation efficiency and practicality

Efficiency and practicality are different aspects, but intertwined regarding the functionality of the different kinds of biofilters. The first aspect is the ability to control pH and nutrients. Because each microorganism performs at a different pH range and needs other nutrients, this factor is important to fit the chosen microorganisms. The accumulation of biomass/waste determines the efficiency of the biofiltration. Clogging and aggregation of residue products from filtration or biomass growth leads to less air being treated by the microorganisms. The ability to recycle nutrients is both influencing the efficiency as the practicality. Lastly, the operation and running costs are also considered.

Table 2: Comparison of three types of biofilters on their functional aspects. Source: Gospodarek et al., 2019

Function	**Ability**	**Conventional biofilter**	**Bioscrubber**	**Biotrickling filter**
Operation efficiency/practicality	control pH and nutrients	no	yes	yes
	accumulation of biomass/waste	no	yes	yes
	nutrients can be recycled	no	no	yes
	operation and running costs	low	high	high

The different aspects of the comparison are ranked on their importance for conceptualizing the optimal biofilter (Table 1 and Table 2). The ability to inoculate specific microorganisms is compared first, and only the conventional biofilter does not possess it. The optimal biofilter removes the earlier stated target pollutants, and each microorganism breaks down specific compounds. To achieve this, the ability to choose the specific microorganisms is fundamental. The conventional biofilter uses a bed with natural microorganisms. This way, a degree of control is lost, as the possibility to use specific microorganisms is absent in the conventional biofilter. Natural selection could stir towards a consortium that filtrates some target pollutants, but efficiency could be lost due to the simultaneous breakdown of less toxic compounds.

Next, is the degree of the ability of each biofilter to break down different compounds. The biotrickling filter scores best overall in the three categories. The minimal concentration at which a biofilter can successfully break down a contaminant is the lowest for the conventional biofilter, followed by the biotrickling filter and the bioscrubber.

Being able to control the pH and nutrients affects the efficiency of the biofilter, and the conventional biofilter is the only type that does not have this ability. In contrast, the conventional filter is the only filter that does not accumulate biomass or waste. Bioscrubbers produce waste water in its filtering process, and the microorganisms in biotrickling filters can grow to a point that the passage of gas is clogged. However, the biotrickling filter is the only biofilter in which the nutritional liquid is recycled. The last criteria are the costs for operating and running the biofilters. With this, conventional biofilter scores better than the bioscrubber and biotrickling filter.

By comparing all the different aspects of a biofilter, a substantiated choice can be made to find an answer to the research question. The first kind of biofilter that gets cut is the conventional biofilter, as the ability to choose inoculated microorganisms and control the pH and nutrients is crucial for finding the optimal biofilter for specific contaminants. In the choice between the bioscrubber and biotrickling filter, the bioscrubber wins it on the mass transfer, but the biotrickling filter wins it on all other criteria. Therefore, the biotrickling filter prevails over the other biofilters, which is supported by previous papers (Mirmohammadi et al., 2017; Lee et al., 2010; Barbusinski et al., 2017; Lee & Heber, 2010). This is why this report will focus on substantiating the optimal biotrickling filter from here on.

Current uses of biotrickling filters are in the paint industry (Naha et al., 2022), pharmaceutical industry (Hu et al., 2016) and landfill sites (Han et al., 2020). It is optimized to specifically filter the contaminants of the offgasses from exhausts. To find the optimal biotrickling filter for the use of removing contaminants from the air ventilating buildings, the different aspects of the biotrickling filter need to be explored and conformed to the goal of the study. The most important factors are the microbial consortium performing the biodegradation, setup of the biofilter and operation configurations. In the next chapters, each of these factors will be discussed to move towards the optimal biotrickling filter.

Chapter 2 Microorganisms

Microorganisms are the motor behind the remediation of contaminants in biofilters. From 1893 until 1982, conventional biofilters were most commonly used. This biofilter contained a medium in which microorganisms were thriving due to its nutritional content. Often, wood snippets, compost, or soil were used through which water or air flowed through (Hort et al., 2009). In this setup, the microorganisms inhabiting the medium were not known and thus not specific for the contaminants. From 1980 forward, more research was done on new types of biofilters. This gave birth to the biotrickling filter in which specific microorganisms could be inoculated. In this chapter, bacteria, fungi and a combination of both will be compared in respect to their removal capacity in biofilters, but first the microbial biofilm in which biodegradation takes place is discussed.

Biofilm

In biology, a biofilm is a mucous layer consisting of microbes embedded in secreted polymers. About 90% of biofilm mass consists of extracellular polymeric substances (EPS) while the microorganisms themselves are the other 10% (Gebreyohannes et al., 2019) Biofilms are a vital part in the maintenance of microbial communities. Its growth is determined by factors such as temperature, pH, nutrient- and oxygen availability and its development comprises the stages of attachment, maturation, and dispersal. First, cells attach to a biotic or abiotic surface in the biotrickling filter via nonspecific interactions such as Van der Waals-interactions (Flemming & Wingender, 2010). In the maturation stage, microbial cells adhere to a substrate creating an irreversible connection and start producing EPS. The third stage involves differentiation, multiplication and finally dispersion. The force behind dispersion differs between the types of biofilters due to their differences in design and mechanisms. In the biotrickling filter, bulk transport with the trickling liquid and gravitational forces lead to new surfaces of the biotrickling filter being colonized (Marycz et al., 2022). Microorganisms secrete EPS in their surroundings so that they embed themselves in the polymers. The nutritious liquid forms another layer on top of the biofilm. This means that the pollutants need to transfer from the gas to the liquid, and then from the liquid to the biofilm to reach the microorganisms where they can be broken down (Figure 4). This mass transfer differs per biofilm and biofilter (as discussed in Chapter 1).

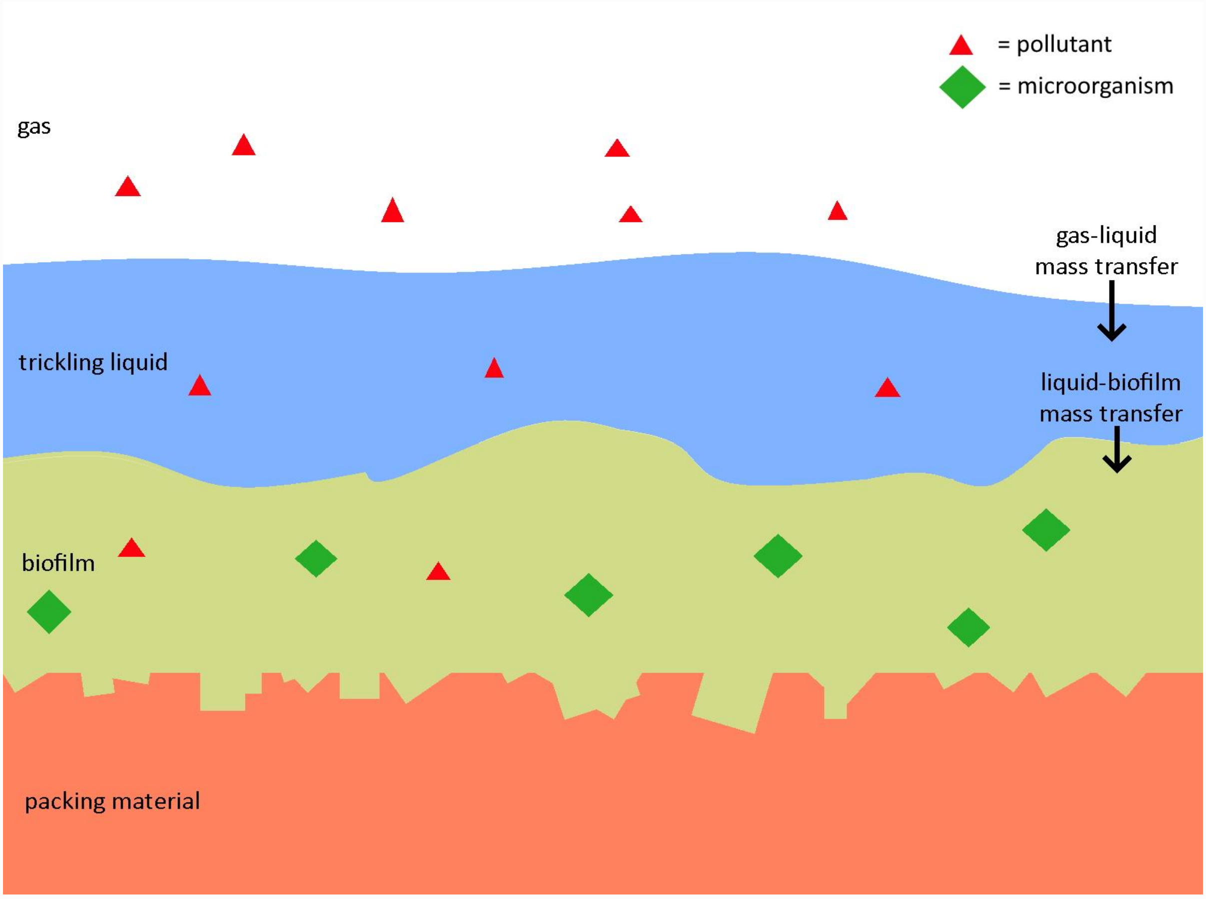

Figure 4: Schematic of transfer of contaminants through the trickling liquid into the bioflim. Source: own

The mass transfer rate is also related to Henry's law constant of pollutants. This law states that the amount of dissolved gas in a liquid is proportional to its partial pressure above the liquid, and each pollutant has its own proportional constant. Figure 4 illustrates the mass transfer into the liquid and biofilm. Once arrived in the biofilm, pollutants can be broken down in a couple of steps. First, the microorganisms catch gas contaminants on their cell surface. The gas contaminants diffuse into the cell, while metabolites and enzymes leave the cell, thereby creating a bidirectional exchange. In the cell, the pollutants are decomposed by enzymes into simpler and less harmful molecules that diffuse out of the cell. Thus, the rate of biodegradation is controlled by both the mass transfer from air to biofilm and by the biodegradation reactions (Li et al., 2012).

Bacteria

Until 1995 it was thought that bacteria were the sole actors in the remediating ability of biotrickling filters. Their catabolic activity happens in the biofilm. To do this properly, different microorganisms need different water activities in the biofilter. Water activity is the amount of free water in a substrate that is actually available to microorganisms. It decreases when solids or chemicals are added to an aqueous phase. A water activity above 0.90 is optimal for growth of most bacteria (Kennes et al., 2004). The pH of the environment is also important, since most bacteria perform best at a near neutral pH (Kennes et al., 2004).

Fungi

In 1996, Weber and Hartmans found that fungi also contribute to bioremediation in biotrickling filters. The catabolic activity of fungi also happens in biofilm, but also in its aerial hyphae. These aerial hyphae grow through the water film, making it possible to directly contact pollutants in the gaseous phase (Vergara-Fernández et al., 2018). It does this by producing proteins called hydrophobins that form a hydrophobic layer of the hyphae. These proteins lower the surface tension so that the hyphae can grow through the water. Additionally, the hydrophobins facilitate direct uptake of pollutants from the gaseous phase, making fungi effective for degradation of hydrophobic pollutants. Water activity for appropriate fungal activity ought to be above 0.60 and the pH can range between 3-8 (Ali et al., 2017).

Some papers report of biofilters efficiency increasing when the bed is inoculated with a combination of bacterial and fungal species (Kennes et al., 2004; Cheng et al., 2016). As each organism can break down a specific compound, a toxic pollutant can be degraded in stages in which different microorganisms contribute. This co-metabolism could make a consortium of both bacteria and fungi the most promising solution.

The next step is to look into the ability of microorganisms to break down the indoor contaminants (Table 3). For each group of pollutants, it was shown that a single or multiple microorganisms degrade the pollutant to a less toxic metabolite. From this, the optimal consortium can be assembled to degrade pollutants from the atmosphere.

Table 3: The different species of fungi and bacteria that degrade pollutants. Sources: Qi & Kinney, 2002; Yoon et al., 2009; Raboni et al., 2017; Rybarczyk et al., 2019; Liew et al., 2020; Vergara-Fernandez et al. 2020; Xie et al., 2021.

Pollutant	***Fungal species***	***Source***	***Bacterial species***	***Source***
VOCs hydrophobic (BTEX)	*Cladosporium sphaerospermum* *Fusarium solani* *Paecilomyces variotii* *Paecilomyces* *Phanerochaete chrysosporium* *Aspergillus versicolor* *Exophiala*	Raboni et al., 2017 Qi & Kinney, 2002	*Pseudomonas putida* *Pseudomonas fluorescens* *Ralstonia pickettii* *Rhodococcus erythropolis* *Acinetobacter*	Raboni et al., 2017
VOCs hydrophilic			*Pseudomonas sp.* *Bacillus sp.*	Rybarczyk et al., 2019
NO_2			*Thauera* *Vulcanibacillus*	Xie et al., 2021

			Anaerovorax *Defluviimonas* *Simplicispira*	
CH_4	*Fusarium solani*	Vergara-Fernandez et al. 2020	Methanotrophs: *Methylosinus trichosporium* *Ganoderma lucidum* *Methylomicrobium album* *Methylocystis sp.*	Yoon et al., 2009 Liew et al., 2020 Vergara-Fernandez et al. 2020
SO_2			*Desulfobulbus* *Desulfosarcina* *Sulfurovum* *Chlorobium* *Thauera*	Xie et al., 2021

Consortia in biofilters

As explained earlier, pollutants diffuse into the cell of a microorganism in a bidirectional exchange. Inside the cell, enzymes break down the pollutants through different mechanisms into simpler molecules. The most common catabolic pathways of microorganisms are hydrolysis, oxidation, and reduction (Xu et al., 2021). Microorganisms use the metabolites as their source for energy and carbon. Products from these reactions are converted into biomass or diffuse out of the cell.

From the previous biofiltration experiments, hydrophilic VOCs, NO_2 and SO_2 are broken down by bacteria, while hydrophobic VOCs and CH_4 can be broken down by either bacteria or fungi (Table 3). For the hydrophobic VOCs, Raboni et al., (2017) found that a two-staged biofilter first treating gas with a bacterial filter and then with a fungal filter, had a higher average degradation efficiency (92.5 %) than the bacterial filter (76.4%) or fungal filter (68.3%) alone. This can be explained by the synergy between fungi and bacteria and the widening of the action spectrum, as intermediate metabolites can be broken down by one another. The most dominant bacteria found in this filter was *Pseudomonas putida*, having multiple catabolic pathways in order to degrade BTEX (Otenio et al., 2005). Apart from this, *Pseudomonas putida* has been proven to degrade a broad scale of other persistent organic compounds (Zhou et al., 2021). In the fungal filter, multiple different strains of fungi were found (see Table 3). From these, *Cladosporium sphaerospermum* and *Exophiala lecanii-corni* were found most abundantly and both have proven able to degrade aromatic hydrocarbons (BTEX), ketones and organic acids (Qi & Kinney, 2002).

For the group of hydrophilic VOCs, all experiments in biofilters used activated sludge for inoculating microorganisms. Activated sludge are clumps of microorganisms that form during remediation processes of wastewater and contain numerous species of bacteria

(Rajasulochana et al., 2016). There is not much research into specific microorganisms that break down hydrophilic VOCs, but often *Pseudomonas sp.* and *Bacillus sp.* were found in the treatment of hydrophilic VOCs (Rybarczyk et al., 2019).

The same synergy between fungi and bacteria in the remediation of hydrophobic VOCs was found for the remediation of methane. In research from Vergara-Fernandez et al. (2020), the removal efficiency was compared between a biofilter inoculated with the biodegrading fungus *Fusarium solani* and a biofilter with *F. solani* combined with methanobacterial strains *Methylocystis sp. and Methylomicrobium album.* In the latter biofilter, the removal efficiency was twice as high, arguing for a mixed consortium for the degradation of methane. Lebrero et al. (2016) found the same result when he compared the removal efficiency of a consortium of the fungus *Graphium sp.* with different methanobacteria with the removal efficiency of a consortium solely with methanobacteria.

Xie et al. (2021) found that a consortium of bacteria was able to degrade CO_2, NO and SO_2. Using sludge from a sedimentation tank, they constructed a biofilter in which 82.81% of NO, 100% of SO2 and 75.23% of CO_2 were removed from an air stream. One of the metabolites in the degradation pathway of NO was NO2, one of the target pollutants. Additionally, they were able to break down 100% of the supplied SO_2. So, with this single consortium, two of the earlier stated target pollutants can be broken down. On top of this, the consortium can fixate carbon from CO2. While this was not a target pollutant originally, it would be advantageous to remove this from the ventilated air in buildings.

From these experiments, specific microorganisms were shown to break down target pollutants. The choice of the final consortium is based on previously tested consortia aiming at degrading specific pollutants. Different microorganisms broke down hydrophobic VOCs and a selection was made based on papers from Qi & Kinney (2002) and Raboni et al. (2017). The bacteria from the consortium of the results from Rybarczyk et al., (2019) were chosen for the hydrophilic VOCs and the fungal-bacterial consortium from Vergara-Fernandez et al. (2020) for the breakdown of methane. Finally, the consortium of Xie et al. (2021) was adapted for the breakdown of CO_2, NO and SO_2 (Table 4). Now that the consortium of microorganisms is substantiated, the next chapter will dive into the practical aspects of bringing together all these parts into a functional biotrickling filter.

Table 4: Final substantiated consortium of microorganisms for the biofilter

Pollutant	**Fungi**	**Bacteria**
VOCs hydrophobic (BTEX)	*Cladosporium sphaerospermum* *Exophiala lecanii-corni*	*Pseudomonas putida*
VOCs hydrophilic		*Pseudomonas sp.* *Bacillus sp.*
NO_2		*Thauera* *Vulcanibacillus* *Anaerovorax* *Defluviimonas* *Simplicispira*
CH_4	*Fusarium solani*	*Methylocystis sp.* *Methylomicrobium album*
SO_2		*Desulfobulbus* *Desulfosarcina* *Sulfurovum* *Chlorobium*
CO2		*Longilinea* *Cloacibacillus*

Chapter 3 Form and configuration

A biotrickling filter is a combination of a bioscrubber and conventional biofilter (Figure 3). The influent gas enters a chamber in which microorganisms are inoculated on an inert bed, while a liquid is sprayed over the filter in which pollutants are absorbed. Unique to its design is the fact that the aqueous solution is recycled in the bottom and re-used for another round nurturing the microorganisms. The biotrickling filter is one of the most recent designs in the field of biofiltration and a lot of research is done to increase its efficiency (Ergas & Cardenas-Gonzalez, 2004). Figure 5A, 5B and 5C illustrate alternative configurations of the biotrickling filter. Figure 5A has two separated inert beds, in Figure 5B the aqueous solution moves co-currently with the inlet gas and Figure 5C has two consecutive chambers.

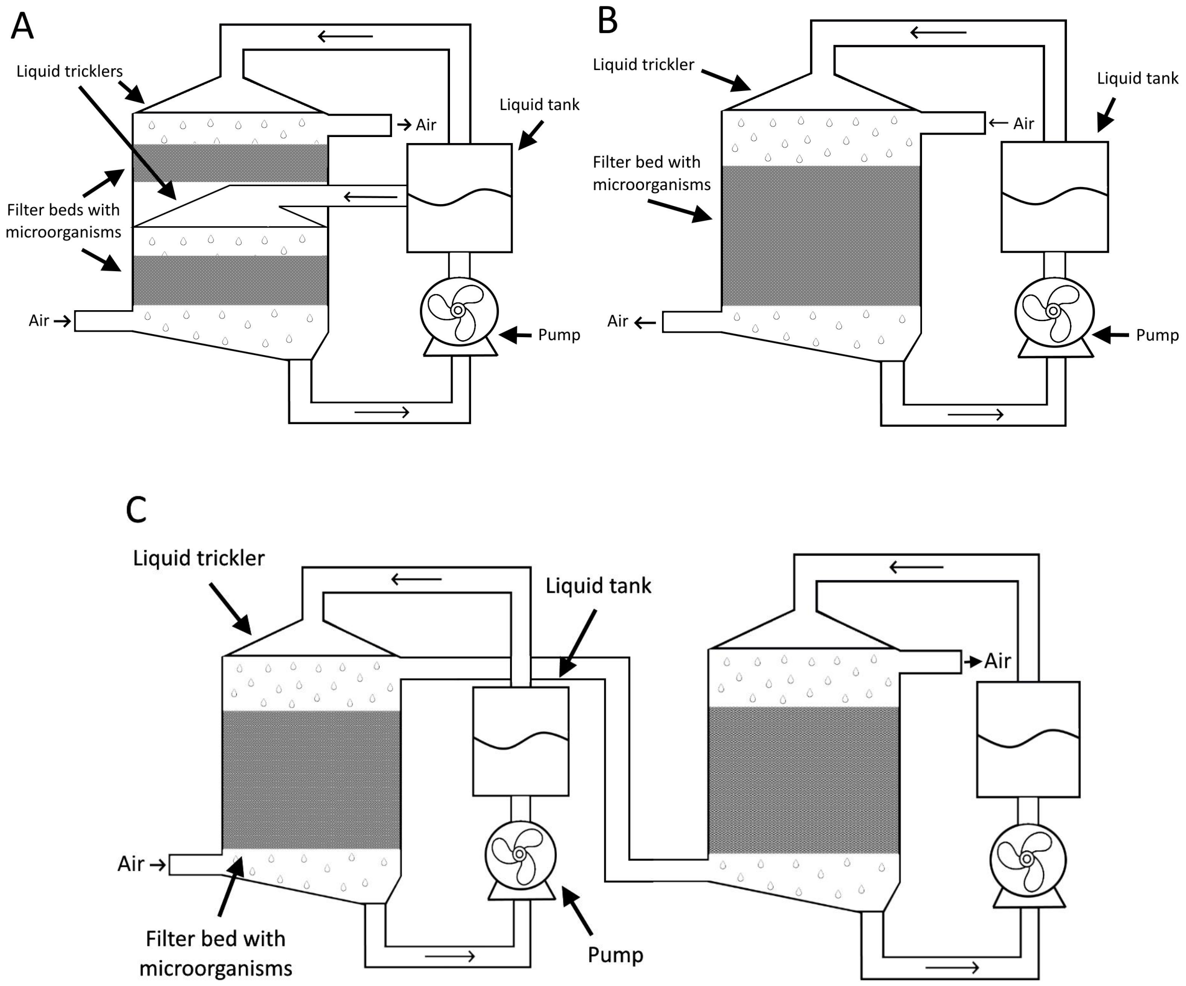

Figure 5: Schematic of alternative configurations of the biotrickling filter. Figure 5A illustrates a biotrickling filter with multiple beds, 5B illustrates a biotrickling filter with con-current movement of gas and trickling liquid and 5C a biotrickling filter with two consecutive chambers. Source: own

Assignment consortia

Chapter 2 concluded with a final consortium of microorganisms that are preferentially used in the biotrickling filter. These bacteria and fungi were chosen from previous remediation experiments combined with the consortium of Xie et al. (2021) that were able to degrade CO_2, NO_2 and SO_2. To prevent possible interference between the different consortia, a biotrickling filter will be used with two consecutive chambers, being chamber I and chamber II (see Figure 5C). Chamber II includes the functional microorganisms from the experiments of Xie et al. (2021) and chamber I contains the functional microorganisms from the experiments of Qi & Kinney (2002), Raboni et al. (2017) and Rybarczyk et al., (2019). The removal efficiency will tell whether a stable equilibrium forms in chamber I. If not, then the microorganisms in chamber I should be divided into additional chambers. This way, the consortia from previous research are grouped together. The order of both chambers is also important for cleaning the air. Biodegradation of VOCs produces CO_2, and the adapted consortium of Xie et al. (2020) breaks down CO_2. Therefore, the consortium degrading CO_2, NO_2 and SO_2 should after the consortium degrading the VOCs and methane (see Table 5).

Table 5: Assignment of different microorganisms to the two chambers.

	Fungi	**Bacteria**
Chamber I	*Cladosporium sphaerrospermum* *Exophiala lecanii-corni* *Fusarium solani*	*Pseudomonas putida* *Pseudomonas sp.* *Bacillus sp.* *Methylocystis sp.* *Methylomicrobium album*
Chamber II		*Thauera* *Vulcanibacillus* *Anaerovorax* *Defluviimonas* *Simplicispira* *Desulfobulbus* *Desulfosarcina* *Sulfurovum* *Chlorobium* *Longilinea* *Cloacibacillus*

Packing material

Microorganisms that degrade pollutants in a biotrickling filter are adhered to an inert bed via EPS (Rybarczyk et al., 2019). This bed is crucial for the functioning of microbes, as it facilitates growth of a biofilm and promotes the high contact area between biofilm and gas. The filter bed consists of a packing material. Requirements for an optimal packing material are as follows:

- Robust and non-degradable, so that maintenance is minimized
- Volume that consist for 35 to 40% of air cavities, providing efficient distribution of a gas throughout the bed
- High surface area which enables biofilm development, thereby increasing mass transfer from gas to biofilm
- Adequate bed drainage for the by-products of the reactions to flow away
- Adequate buffer capacity to ensure a favorable pH
- Odorless
- Hydrophilic to allow retaining of water vital for the biofilm and microorganisms

Packing materials can be made from organic, inorganic or mixed materials (Wu et al., 2018). Organic materials, such as peat or wood chips, were primarily used in early biotrickling filters and conventional biofilters. Due to the tendency of organic packing material to clog biofilters, synthetic organic packing material such as rubber particles and plastic pall rings were developed as alternatives (Park et al., 2011).

Commonly used inorganic packing materials are lava and perlite, as their irregular shape have good surface properties (Wu et al., 2018). Mixed packing material was developed to solve the problem of clogging in organic packing material. It is produced by adding inert packing materials with large pores, such as polystyrene spheres, lava rocks and glass beads, to organic packing material.

In order for a packing material to function for a long time in a biofilter, it should not degrade. Therefore, biofilters most commonly use chemically non-reactive (inert) materials (Kim & Deshusses, 2008). This leaves the choice between inorganic- and synthetic organic packing material, of which both are inert. Previous research by Kim & Deshusses (2008) has looked at different materials and their influence on the degradation of pollutants. They compared the gas-liquid mass-transfer coefficients and the liquid-biofilm mass-transfer coefficients of different packing materials. These materials included Pall rings, porous ceramic Raschig rings, compost-wood chips mixture, polyurethane foam cube and porous ceramic beads. Out of these materials, the porous ceramic beads had the highest gas-liquid and liquid-biofilm mass-transfer coefficients, hypothesized due to its density (Kim & Deshusses, 2008). These materials all possess the previous stated requirements, so with the result from this experiment, porous ceramic beads will be chosen as packing material in the optimal biotrickling filter.

Settings of operation

For maximal performance of the biotrickling filter, optimal settings of operation have to be specified. The factors that influence the removal efficiency in biotrickling filters are listed in Table 6. Each compound has a specific configuration of factors for its breakdown. Therefore, operation settings may be suboptimal if mixtures of compounds or contaminants need to be degraded, especially when the different compounds affect each other. Concentrations of pollutants around the world also differ, so optimal settings are also dependent on the location.

Table 6: Different factors that influence the removal efficiency in biotrickling filters

Settings in biotrickling filter
temperature
pH
amount of trickling liquid
consortium of microbes
amount of gas in chamber
time of gas residence in chamber (EBRT)
nutrients in trickling liquid
order of different chambers
size of chamber

To find the optimal configuration of settings for a specific mixture of contaminants, the settings in Table 6 should be altered. During these alterations, the concentration of target compound in influent- and effluent gas should be measured, using a small gas chromatograph. Comparing each variation in the settings, with the removal efficiency, will eventually give the optimal configuration for a specific mixture of compounds. As both chambers have different consortia of microbes, their settings will assumably differ. Using previous literature, a proposal of some settings will be substantiated.

Xie et al. (2021) experimented with the optimal pH and gas-trickling liquid ratio. Throughout the experiments, they kept a temperature of 25 °C, found the ideal pH to be 9 and gas-trickling liquid-ratio to be 1:3. As these configurations obtain the highest removal efficiency of their consortium, and this exact consortium is adopted, these settings are adopted for chamber II in the optimal biotrickling filter. Chamber I houses fungi as well as bacteria that are expected to function at different optimal pH. Fungi have a wide rage of pH, from 3 to over 8, but generally thrive in an acidic environment with an optimum pH of around 5 (Ali et al., 2017). Bacteria function best at a near-neutral pH. The pH in chamber I will be kept at 6 and changed to see what pH works optimal for this chamber. Maintaining and changing the pH will be done with the trickling liquid. The temperature and gas-trickling ratio of chamber I will start at the same values. After the biofilter is functional, changing both factors can lead to optimal settings.

The time that the gas resides in a chamber of a biofilter is called the empty bed retention time (EBRT) (Xie et al., 2021). The longer a gas stays in a biofilter, the more contaminants will be degraded. Xie et al. (2021) found that the minimal retention time for their experiment was 2.85 minutes. As the settings from their experiments were copied to obtain similar removal efficiency, the EBRT is kept at 2.85 minutes. For chamber I, the EBRT of the experiments that the microorganisms were chosen from, will differ from chamber II. Vergara-Fernandez et al. (2020) found that their consortium of *F. solani* with *Methylomicrobium album and Methylocystis sp.* broke down approximately 19% of total influent methane during an EBRT of 24 minutes, and approximately 14% of total influent methane with an EBRT of 6 minutes. The experiments of Vergara-Fernandez et al. (2020) were done in a PVC-column of 105 cm in length and 7.9 cm in diameter. When the length of the chamber is increased, the removal efficiency does not increase linearly. This is because other factors, such as the production of metabolites, could hinder degradation. However, as buildings require lots of fresh air, a larger chamber is proposed. This way, the contact surface is increased so that the EBRT can stay low. A cylindrical chamber of 200 cm in height and 100 cm in diameter would have a volume of approximately 1500 L, which is about 735 times the size of the experiments of Vergara-Fernandez et al. (2020).

At the start, an EBRT of 6 min will be chosen for the first chamber. Because the EBRTs differ in both chambers, a valve between them is installed to ensure constant EBRTs. Alternating the EBRTs combined with the results from measuring the concentration of each gas, will lead to final optimal EBRTs for both chambers. The amount of gas in the chambers, is dependent on the ventilation system present in buildings, as this powers the inlet of air in the biofilter. Important hereby is that the amount of gas will not impede the removal efficiency.

Trickling liquid

Nutrients in biotrickling filter-liquid typically are nitrogen, phosphorus, potassium, and sulfur (Rybarczyk et al., 2019). However, the goal is to obtain a consortium in which all microbes are part of the breakdown pathway of the target pollutants. Therefore, minimal nutrients will be added so that natural selection transforms the consortia containing only the essential microorganisms. The target pollutants for chamber II contain nitrogen and sulfur, so the nutrient liquid here will only contain phosphorus and potassium. Chamber I was not tested for the breakdown of the target pollutants containing nitrogen and sulfur, so this liquid includes nitrogen, phosphorus, potassium, and sulfur. When removal efficiency lowers with the chosen nutrient mixture, concentrations will be altered and nitrogen and sulfur can be added so that the microbes grow more.

The trickling liquid in a biotrickling filter also regulates the pH. In chamber I, a lot of CO_2 will be produced due to the breakdown of VOCs, which will lower the pH. The trickling liquid should counter this by having an alkaline nature. The pH of chamber II should also be higher, so the trickling liquid in chamber II should be more alkaline than the trickling liquid in chamber I. After the biofilter is functional, tuning of the pH of the trickling filter is possible to find the optimal version. Lastly, the continuous sprinkling of the liquid also prevents spores of the fungi from exiting the chamber (Saucedo-Lucero et al., 2014). This finalizes the operating settings for both chambers (Table 7).

Table 7: Proposed settings operation for the start-up of the biotrickling filter

	Chamber I	Chamber II
Temperature	25 °C	25 °C
pH	5	9
Amount of trickling liquid (ratio gas:liquid)	1:3	1:3
Consortium of microbes	See Table 4	See Table 4
Time of gas residence in chamber (EBRT)	6 min	2.85 min
Nutrients in trickling liquid	Nitrogen, phosphorus, potassium and sulfur	Phosphorus and potassium
Size of chamber (length x diameter)	200 cm x 100 cm	200 cm x 100 cm

Maintenance

While biofiltration is a passive process, biotrickling filters require maintenance. As the microbes convert contaminants into biomass, the biofilm accumulates. This can lead to a pressure drop, in which the gas moves more slowly through the filter, and eventually the filter could clog. To prevent excessive biomass formation, mechanical, chemical and biological methods are available (De Vela & Gostomski, 2018). Mechanical methods include stirring or water draining, while chemicals either detach the biofilm from the bed or inhibit growth of microorganisms. Biological control of excess biomass focuses on introducing predatory species that feed on the microorganisms to limit biofilm growth. The ideal method should be sustainable, prevent long-time loss of efficiency, and require minimal effort. Mechanical methods depend on the size of the bioreactor, as actions like stirring and washing need to be intensified to treat bigger reactors. It also demands a lot of energy, and the biotrickling filter is switched off during maintenance. Chemical methods proved more aggressive. For example, with minimal amounts of 0.001 M NaOCl, removal efficiency is brought down to 10% of the maximal capacity and requires 10 days to recover back to 90% maximal capacity. Residual chemicals can inhibit growth and slow down recovery. The biological method of introducing natural predators is the least energy demanding procedure, as the predators do the work after their introduction. Downside of this method is that it is harder to control the amount of biomass that is removed. Together, the biological method is the most preferred since it is sustainable, prevents long-time loss of efficiency and requires minimal effort. Zhang et al., (2020) looked into the mechanism of biological biomass control, and found that larvae of the mosquito species *Bradysia odoriphaga* successfully removed biofilm through peristaltic movement. Biofilm was then washed away by the nutrient liquid. As this method has all the previously stated requirements, the problem of lack of control maintains. However, Won et al., (2004) found a solution by lowering the amount of gas flowing through the chamber. This resulted in a lack of oxygen that reduced the population of mosquitos. The frequency and duration should be carefully tested so that the net growth of biofilm is zero at the maximal removal capacity of the biotrickling filter.

Discussion

In this study, the optimal biofilter was substantiated for cleaning air that enters the building through ventilation. Using literature from previous research, the most harmful pollutants for nature and humans were determined. Then, microorganisms able to break down these target pollutants were found, and a consortium was proposed. Finally, the type of biofilter, setup and operating settings were discussed to come to the optimal biofilter (Figure 6).

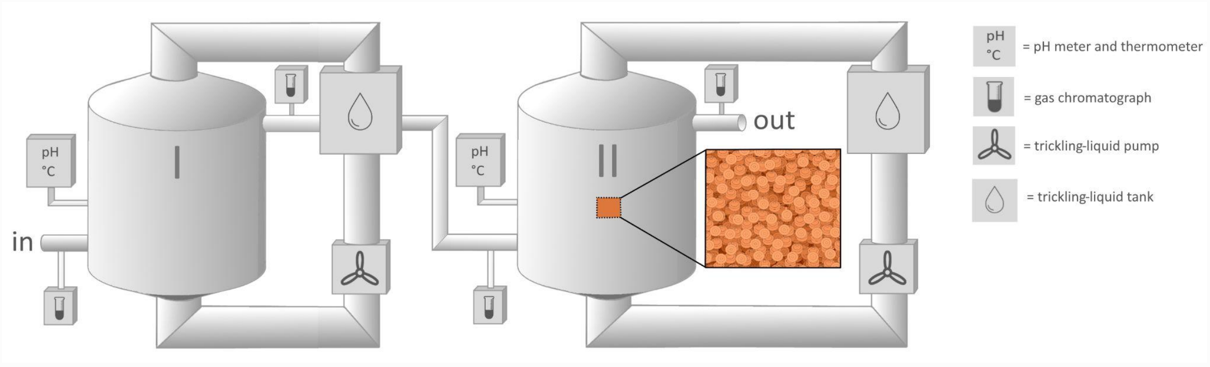

Figure 6: Schematic of the substantiated optimal biotrickling filter. Polluted gas enters chamber I in the bottom left, moves through the biotrickling filter and exits chamber II purified in the top right. The chambers are filled with ceramic beads onto which the microorganisms are adhered via the EPS. Source: own.

The most harmful pollutants for nature and humans present in the atmosphere are VOCs, NO_2, CH_4 and SO_2(WHO, 2021). Qi & Kinney (2002) and Raboni et al. (2017) found consortia that were able to break down VOCs and Vergara-Fernandez et al. (2020) found a consortium able to break down CH4. These consortia were adapted to be inoculated in chamber I. Xie et al. (2021) found a consortium able to break down CO_2, NO_2 and SO_2, which was adapted and placed in chamber II. While $CO_2$was not an initial target pollutant, it is broken down in the adapted consortium in chamber II. Because the breakdown of VOCs produces additional CO_2, these microorganisms are placed in chamber I, so that the consortium in chamber II can remove it. The biotrickling filter is chosen because of its ability to break down different types of pollutants and degree of control that can be exercised (see Table 1 and Table 2). The chambers are proposed to be 200 cm in length and 100 cm in diameter, to ensure a low EBRT, crucial for the amount of air ventilated into buildings. The EBRT in chamber I is 6 min, while the EBRT in chamber II is 2.85 minutes, so a valve between the chambers regulates this. Local concentrations of the target pollutants differ from place to place, causing the optimal configuration of the biotrickling filter to be place-dependent. By changing each setting and measuring a change in removal efficiency, an optimal configuration is achieved. The biofilter is installed to a (present) ventilation system. This powers the inlet of gas, while preventing the amount of gas from lowering the total removal efficiency. To prevent the biofilm gaining excessive mass, a predator will be added to the filter. A group of *Bradysia odoriphaga* mosquitoes removes biomass through peristaltic movements, so that the amount of biomass remains constant. When the mosquitoes remove too much of the biofilm and removal efficiencies decrease, lowering the amount of inlet gas suffocates the excess mosquitoes. A substantiated default configuration of settings is given in Table 7.

This study presents a new insight into biofiltration, as previous research primarily focussed on treating the exhaust off-gas of polluting instances, such as factories and land-fill sites (Ferdowsi et al., 2022). By reversing the process, incoming air in buildings can be purified using the biotrickling filter in this study. To the knowledge of the author, this is the first study looking into this application of biofiltrating incoming air.

When this system is distributed on a large scale, every building serves as a cleaning station for filtrating air in the atmosphere. Due to the built-in adjustability, it can function efficiently in different parts of the world. When solar energy is used, either through generating electricity to power the machine or heat the chambers, the system has the potential to function completely passive. The design is commercially interesting, as companies would not only improve the working conditions in their buildings, but also contribute to preventing further climate change.

A major assumption in the design is the formation of a stable equilibrium in chamber I. As this consortium contains a mixture of microorganisms of which the interrelations are unknown, the kinetics of biodegradation can interfere with the removal efficiency. Removal efficiencies will tell whether an equilibrium forms, or that the consortium needs to be split into an additional chamber. This is a considerable aspect, as the total costs of the setup and operation of the biotrickling filter increase significantly with additional chambers.

Another assumption is that the chosen consortia were tested at high concentrations of isolated contaminants in gas. Isolated compounds flowed through biofilters, from which the removal efficiency of the consortium was calculated. A mixture of gas may have a different result, as the compound can interfere in degradation pathways. Additionally, the concentration of pollutants in the atmosphere is lower than in the experiments, and further research should point out whether the same removal efficiency is obtained with the atmospheric concentrations.

The optimal configuration of the biotrickling filter is dependent on the concentration of pollutants in an environment. To fine tune the configuration for the highest removal efficiency, a lot of time has to be spent on experimenting with all the settings. During this, the biotrickling filter will only require energy while unable to filter air.

With the current setup, the initial setup costs are fairly high, as big chambers are used, requiring powerful ventilation systems. This means that the conceptual design has a small target group, solely being individuals or companies with a large budget for sustainability.

This study provides a first step in the direction of sustainable air filtration by reversing the current method of biofiltration. The microbial consortia are the motor behind the biotrickling filters, and future research should focus on improvements in which microorganisms complement instead of interfere with each other. This way, fewer chambers can be used, which significantly reduces the costs of the biotrickling filter. A different approach for finding a functional consortium is by using activated sludge obtained from instances such as waste water plants. When this sludge is isolated and subjected only to the target pollutants, present microbes able to feed on it survive while others die. Having more knowledge on the functional microorganisms and their interactions in the consortia also leads to a better understanding of the optimal settings for operation. It would also contribute to finding more suitable packing materials that would better support biofilms, mass transfer and contact area. Real-live monitoring of all operating parameters and removal efficiency would make it able to improve settings more quickly.

All these aspects will contribute to the biotrickling filter being more efficient and affordable, so that in the future, every house can be provided with the system. This way, we all contribute to preventing further climate change, one breath at a time.

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