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Abstract: Cities are producers of high quantities of secondary liquid and solid streams that are still poorly utilized within urban systems. In order to tackle this issue, there has been an ever-growing push for more efficient resource management and waste prevention in urban areas, following the concept of a circular economy. This review paper provides a characterization of urban solid and liquid resource flows (including water, nutrients, metals, potential energy, and organics), which pass through selected nature-based solutions (NBS) and supporting units (SU), expanding on that characterization through the study of existing cases. In particular, this paper presents the currently implemented NBS units for resource recovery, the applicable solid and liquid urban waste streams and the SU dedicated to increasing the quality and minimizing hazards of specific streams at the



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). source level (e.g., concentrated fertilizers, disinfected recovered products). The recovery efficiency of systems, where NBS and SU are combined, operated at a micro- or meso-scale and applied at technology readiness levels higher than 5, is reviewed. The importance of collection and transport infrastructure, treatment and recovery technology, and (urban) agricultural or urban green reuse on the quantity and quality of input and output materials are discussed, also regarding the current main circularity and application challenges.

Keywords: circularity challenges; nature-based solutions; supporting units; urban streams; circular cities

1. Introduction

Cities are centers of human and economic activity and, in this flurry of activity, produce high quantities of discarded materials and products, effectively functioning as concentrators of natural resources, perpetuating the current linear system of "take-make-dispose" [1]. In order to combat this huge resource consumption, there has been an ever-growing push for the adoption of better resource management and waste prevention in urban areas, in line with the concept of the circular economy [2]. This paper is a product of an interdisciplinary cooperation among researchers from all 28 EU countries and 11 third countries within the EU-funded COST Action 17133 "Implementing nature-based solutions for creating a resourceful circular city" (https://www.cost.eu/actions/CA17133/, accessed on 30 September 2021) that attempts to contribute to this discussion of the implementation of a circular economy in cities, particularly by the use of specific technologies, interventions, and units based on natural principles.

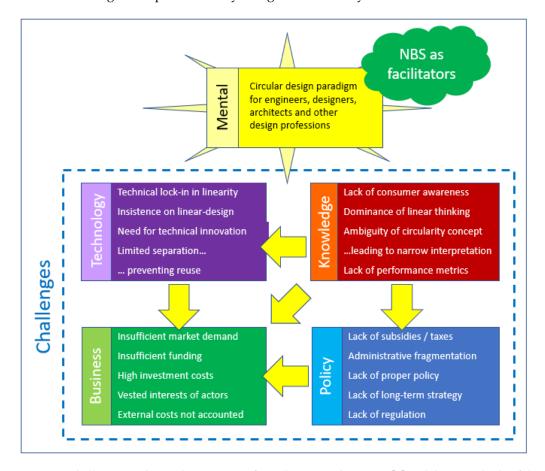
The definition of a circular economy provided in the first paper of the COST Action Circular City [3] is based on previous definitions provided by other sources [4,5], describing it as an economic system that aims at minimizing the waste and input of energy and returns as many resources as possible.

However, the implementation of circularity in urban areas comes with added challenges which need to be addressed (Figure 1). Paiho et al. [6] attempted to develop a comprehensive list of these challenges, which were subdivided into four main categories, a "Business" category, a "Policy" category, a "Technical" category and a "Knowledge" category.

In addition, a fifth "mental" category (Figure 1) is the necessary shift from the prevailing linear approach in problem-solving towards a more holistic, circular design approach [7]. Engineers, designers, architects, and other design professionals are playing a key role in creating the built environment. In the traditional linear design process, the effects that occur outside of the system borders are generally considered as a separate problem. As a consequence, it is in the best case tackled in a separate design process, or else directly handed over to nature (which mostly means that it is not addressed at all). This practice is inherently prone to creating new environmental challenges, as the development of wastewater management in the last two centuries demonstrates [7]. The development of a circular design paradigm for the above-mentioned professions must therefore also be part of the development of circularity practices.

To deal with these circularity challenges, Langergraber et al. [3] proposed to apply the concept of a circular city as a basis for the application of nature-based solutions (NBS).

NBS can serve as cost-effective, resource efficient, and locally adapted application tools of circular economy within cities [3,8]. In general, by their definition, NBS can serve both as replacements for the grey infrastructure that is based on linear principles, and also as complementary systems that can help in the transition towards circularity [8,9]. The COST Action Circular City also attempted to define the specific circularity challenges in cities approachable by NBS. These technical challenges were defined by Atanasova et al. [10] as follows:



- Preserving natural resources by reducing their import;
- Minimizing waste production by using resources in cycles.

Figure 1. Challenges to the implementation of circularity in urban areas [6] and the critical role of the mental category—a new circular design paradigm—to address them, using nature-based solutions as facilitators.

Atanasova et al. [10] as well as Langergraber et al. [11] attempted to specify these challenges towards the specific resource streams treated by NBS (water, food, materials, energy). To that extent, a detailed list of urban circularity challenges (UCC) could be obtained as follows:

- 1. Restoring and maintaining the water cycle (by rainwater management) (UCC1);
- 2. Water and waste treatment, recovery, and reuse (UCC2);
- 3. Nutrient recovery and reuse (UCC3);
- 4. Material recovery and reuse (UCC4);
- 5. Food and biomass production (UCC5);
- 6. Energy efficiency and recovery (UCC6);
- 7. Building system recovery (UCC7).

Additionally, however, NBS can provide other advantages in urban settings, such as the enhancement of their environmental and ecological status, addressing the demand of the populations for natural resources, and climate change mitigation and adaption, among others. In this way, human well-being is improved and the societal challenges of urban living are ameliorated, ensuring approval of the local populations [8]. This entire process, therefore, ensures a systemic transition towards circularity in cities, which guarantees not just economic and environmental harmony, but also societal support.

This movement towards circularity will promote the recovery and the closing of the loops of both water within cities and several nutrients as well as other resources carried by city waters [12]. The water itself can be recovered, reclaimed, and reused in order to obtain

a more sustainable water management system [13]. Many of the water flows within cities are characterized by high carbon, nitrogen, phosphorous, and potassium contents [14].

Challenges are posed by other elements contained in wastewater, such as metals, pharmaceuticals, pesticides, etc., which require treatment and recovery [9]. The origins of these flows are primarily derived from management systems, namely sewage systems, rubbish bins, and exhaust pipes, that are ubiquitous in modern cities. Therefore, these secondary resource streams (urban and industrial wastewater, municipal solid waste, and gaseous effluents) are the key aspect in the development of a solid closed loop economic model confined to city boundaries [9,14]. Zeller et al. [2], after an analysis of the waste flows of various sources in the city region of Brussels, concluded that wastes with the lowest market value accumulated at high density and high unit cost and transportation by treatment (such as municipal solid waste or organic waste) are more suited for local material recycling and energy recovery than high-market-value waste such as metal and glass. Thus, the highest circular economy valorization potential for these secondary bioresource flows, that is, organic waste containing nutrients and biomass, comes in the form of technologies capable of integrating these waste flows within the urban metabolism.

In order to determine an adequate balance of the material and energy flows in organic waste and wastewater systems in urban areas to ensure circularity, first, these flows must be identified and studied, both as inputs and outputs. The bibliography on this matter still considerably lacks in characterization of these flows. This is primarily due to the absence of a uniform characterization model for resource flows in urban areas and a lack of defined geographical boundaries to limit the urban areas [6]. Urban metabolism studies have used several different approaches, from material flow analysis (MFA), life-cycle assessment (LCA), input-output (IO) analysis, cost-benefit analysis (CBA), spatio-temporal modelling with geographic information system analysis, and many others [6,15-17]. Some studies have tried to perform a flow balance of the waste/resource flows within urban areas and their surrounding regions [2,18], with only rare cases considering specific material and energy flows of organic waste and wastewater management [19]. Alternatively, other studies focused on some of the specific technologies applied in urban areas, such as sanitation systems [17,20], which defined the following inputs: total phosphorous (P), total nitrogen (N), potassium (K), total solids, and water. Water is considered as an increasingly scarce commodity in urban areas due to human and industrial pressure, which needs to be saved or reused. Both P and N were defined as important macronutrients, while total solids were in turn used as a proxy from which either energy could be recovered in the form of biochar or biogas, or organic matter could be recovered as a soil amendment. However, P, N, and total solids can also be considered pollutants, as their mismanagement and accumulation in water bodies can lead to algal blooms, eutrophication, and hypoxic dead zones [14].

Coupled urban agricultural systems bear huge potential to contribute to circular cities, with NBS as enablers. A substance flow analysis of the city of Vienna concluded that, with a treatment system using NBS, the wastewater and biodegradable kitchen waste from 77,250 people could be processed to supply the N and P fertilizer needed for the entire vegetable production within the greater city boundaries, which currently supplies one-third of the city's vegetable consumption [21].

Another example of an analysis of a coupled urban-agricultural system and the material flows of P and N is provided by Firmansyah et al. [22]. Overall flow analysis showed that the agricultural system was a significant source of N and P nutrients lost through erosion/run-off and leaching. The urban systems also had a considerable negative impact on this isolated ecosystem local due to N and P losses from domestic waste and wastewater by leaching and atmospheric emission. The authors of this study concluded that the nutrient management was clearly unbalanced. Approaches to rebalancing the situation within the island come mostly by changing the current sanitation system, ensuring the retrieval of N and P present in domestic waste and runoff for application in all sub-systems.

Based on the information gathered on the definition of the input and output flows of the bioresources and equivalent materials, even in circular systems, it must be understood that a circular urban system will never be fully self-sufficient: much of the nutrients in cities such as nitrogen and phosphorous enter in the form of food, which is heavily produced outside city boundaries [2,6,22]. Although some food production can be developed inside cities in the form of urban farms or equivalent units [23], the import of food is still and will remain dominant. Therefore, in order to achieve a circular bioresource system in cities, it must be ensured that the flows of nutrients must be subdivided into separate fractions: (i) one that will remain in the city environment and helps in subsisting both the natural ecosystem and some of the human activities; and (ii) another fraction must be adequately exported out of the city for agricultural and other purposes. The recovered nutrients can be transformed into composts or fertilizers with high nitrogen (N) and phosphorus (P) content. In this paper, we discuss NBS supporting the flows of nutrients in the city environment while the fraction of flows for agriculture and other purposes outside the city borders is not addressed.

Considering the necessity of maintaining a sustainable management of water, nutrients, and other meaningful flows present in the urban water and biowaste sector, it becomes obvious that novel or already used technologies need to be evaluated, not only for their efficiencies and economic output, but also for their potential to separate and recover these elements in the same or novel forms to achieve circularity. In that way, both environmental and economic value can be derived, and a successful transition of this sector towards circularity can not only be possible, but attractive. The NBS methodology, of which the development of a holistic, circular approach for problem solving is a part, is extremely interesting, as the inherent focus on resource recovery ensures an improved management of water, carbon, nutrients, energy, and potentially other elements that can be used in interconnected systems [8]. It may become the key facilitator for the implementation of circularity (Figure 1). Contaminants (pathogens, organic micro-pollutants, potentially toxic metals, etc.) can be kept out of the waste stream or removed to an extent that the product is safe for reuse. It is therefore necessary to study the various NBS units and its input and output resource flows derived from the urban activities. Several NBS units in combination with several supporting units (SU) can together form a resource recovery system to help the recovery of above-mentioned elements and mitigate risks associated with contaminants. In that way, the methodology introduced with the concept of NBS can be proved to fulfil the goals previously set out. While biological processes are the foundation for NBS, other units based on chemical and physical principles may be required to effectively "close the loops". The mass and energy balances of SU also need to be studied in detail to comply, when necessary, with the demand on quality output streams and a reduction of footprint. Within this study, those NBS units including the SU that have been applied as part of a circular system in a local, city environment, are discussed.

This publication serves as a follow-up to previous contributions of the COST Action Circular City since it expands on their findings. A previous COST Action Circular City publication in particular [9], which provides a list of NBS that perform resource recovery activities, is the basis for the NBS and SU selected in this publication. The selective criteria are based on a novel methodology expanded upon in the following chapter. Resource flows (water, nutrients, energy, bioresources) passing through selected NBS and SU are provided, expanding them on the characterization of real-life cases already implemented in cities. The purpose is to provide a detailed guide of the possible resource recovery solutions, mostly technological, alongside any limitations or challenges to be resolved in order to achieve the circularity by implementation of NBS in cities. As a result, we provided a compelling archive for consultation on the merits of these novel solutions as good options to be implemented further in urban areas, in order to guarantee the sustainability of cities within and outside the European Union.

2. Methodology

The present paper applies the definition for NBS units of Langergraber et al. [3], viz. "technologies that bring nature into cities and those that are derived from nature, using organisms as principal agents if they enable resource recovery and the restoration of ecosystem services in urban areas".

When building NBS systems for resource recovery, next to NBS units mostly described by Langergraber et al. [11], often physical/chemical SU are needed to enable the production of high-concentration products such as precipitates from phosphorus or ammonium salts, or to remove pollutants such as pharmaceuticals, personal care products, or pathogens. Based on Langergraber et al. [11], a selection of these SU is also discussed in this paper. All selected NBS and SU are analyzed for potential city input and output streams and systematically presented in Supplementary Materials A and B, respectively. Additional NBS units as well as other SU that contribute to resource recovery have been introduced in the present paper.

The criteria for selecting the NBS and SU, analyzed in the present paper are:

- Relevant for the recovery of resources such as water, CO₂, nutrients, energy, organics, and metals from city (waste) streams;
- Already applied (TRL > 5) as a unit in a local (decentral) circular system (micro-(household), meso- (district) and macro- (city and above) scales; [3,9] in the city;
- Applicable in an urban environment.

3. Results

3.1. Liquid Incoming Streams

3.1.1. Treatment Wetlands

Working Principle

Treatment wetlands (TWs) (also called constructed wetlands) comprise a series of engineered systems designed and constructed to mimic natural processes found in natural wetlands involving vegetation, soils, or gravel and their associated microbial communities to provide treatment for various wastewater streams. TWs are divided into two main hydrologic categories: (a) open-water surface wetlands, which are shallow sealed basins (one or a sequence) with open water areas planted with floating, submerged, or emergent wetland plants (similar to the appearance of natural marshes); and (b) subsurface flow wetlands, which consist of one or more deeper sealed beds filled with gravel and sand. Water flows below the surface level of the filter bed, either horizontally (horizontal flow or HF wetlands) or vertically (vertical flow or VF wetlands) [24,25].

The application of subsurface flow wetlands are the most appropriate in cities. TWs can be applied in micro-, meso- and macro scales, which also result in different end usersfrom individuals and local communities to water utilities. Although the majority of TWs are applied in rural areas to provide on-site or decentralized wastewater treatment, their application in urban settings is gaining attention (e.g., TW in Orhei for 20,000 PE as the main wastewater treatment plant of a city; [26]). However, since cities face very limited space and TWs require a large area, which is their biggest constraint, new types of TWs are being developed, such as rooftop wetlands or vertically oriented systems. Vertically oriented systems that treat wastewater are also called (intensive) green walls and have been investigated mainly for greywater treatment [27–29]. Implementations often aim for treated water reuse in the form of onsite fertigation or toilet flushing, for example, in the stepwise aligned indoor/outdoor vertECO $^{\! \odot}$ system installed at a touristic resort in coastal Lloret de Mar, Spain [30]. Current setups at TRL7 prove the applicability of green wall systems, even for the liquid-phase of household wastewater (HOUSEFUL project; EU Grant Agreement ID: 776708; https://cordis.europa.eu/project/id/776708, accessed on 30 September 2021).

In- and Outputs

In terms of incoming wastewater flows, most of TWs receive primary treated domestic wastewater. Primary treatment includes various sedimentation units (SU 7). Primary treated domestic wastewater contains 30–40% initial suspended solids and 60–75% initial BOD₅. TWs can also receive secondary treated domestic wastewater, which has low organic and suspended solids content and high nutrient content, and acts as a tertiary treatment step. In addition, TWs can also be used for the final treatment of tertiary treated water. The content of components in secondary and tertiary treated water depends on the national regulatory requirements for secondary and tertiary treatment in the country in which the system is used. In addition, TWs can be used to treat greywater, industrial wastewater, and rainwater. Greywater has the following characteristics: COD 200–700, BOD₅ 100–400, TN 8–30, TP 2–7 mg L⁻¹ [31,32]. As mentioned above, most green wall TWs built and investigated so far treat greywater. However, the treatment of the liquid phase of household blackwater has been tested successfully in the ongoing HOUSEFUL project (EU Grant Agreement ID: 776708; https://cordis.europa.eu/project/id/776708, accessed on 30 September 2021).

The key output of the TWs is secondary, tertiary, and finally polished treated water with the characteristics according to the respective national legislation and the solids retained in the primary treatment in the form of primary sludge. A special type of VF TW—French reed bed—is designed to receive raw domestic wastewater. In this case, the solids are not removed in a primary settler, but accumulate on the top layer of a vertical filter bed. The accumulated partially mineralized and dewatered sludge is removed every 10–15 years [33].

By planting herbaceous or woody plants, TWs provide plant biomass. The biomass production of *Phragmites australis*, the most common plant used in TWs, is 19 ± 13 t ha⁻¹ y⁻¹ when used for the secondary treatment of domestic wastewater [34]. A special type of TW, so-called evapotranspirative systems, wherein short-rotation willows are planted in the treatment beds, can produce more biomass as reeds, e.g., 22–26 t wood chips ha⁻¹ y⁻¹ [35,36].

Connected Units

Regarding liquid in-/outputs, TWs can be connected to phosphorus precipitation SU because phosphorus can be recovered or removed. For the further removal of specific pollutants, TWs can be combined with activated carbon units, advanced oxidation processes, and membranes. The reclaimed water can be used for the irrigation or fertigation of street trees and urban parks (NBS 39, 40, 41), urban agriculture (NBS 47, 49, 51) [37], or any other unit to cover water needs.

Regarding solid in-/outputs, TWs are usually connected to solid-liquid separation (settling tank) as an SU. The sludge can be further treated in an anaerobic digestor to produce biogas and in sludge drying bed to produce soil amendment. The biomass can be composted to produce fertilizers or a compost matrix; the woody biomass can be used for river revitalization elements (NBS 28), for energy production, or as a source of lignocellulose for the production of composite materials.

Case Studies and Literature

Within the city, TWs have recently been applied mainly for greywater treatment in sustainable housing estates or public institutions. The treated water is used for groundwater recharge (Lübeck, Germany), toilet flushing (Hannover, Germany), or the irrigation of vegetable gardens (Lima, Peru—https://www.susana.org/_resources/documents/default/ 2-70-en-susana-cs-peru-lima-sanchristoferus-2009.pdf, accessed on 30 September 2021). In the latter case study, a separate TW is also used to treat liquid fraction of blackwater, and the treated water is used for the irrigation of lawns, fruit trees, and flowers. The listed examples apply on a micro and meso scale and are also presented in Table 1.

Case Study	NBS Units	Supporting Units	Product and Reuse	References
Sneek—Noorderhoek, the Netherlands	Anaerobic BW treatment (UASB) Aerobic GW + effluent BW treatment OLAND (nitritation/anammox)	Vacuum toilets, struvite precipitation, membrane filtration	Biogas, Struvite, reclaimed water	https://www.saniwijzer.nl accessed on 30 September 2021
Lübeck, Flintenbreite, Germany	Vertical flow treatment wetland for greywater, anaerobic treatment for blackwater and biowaste	Vacuum toilet	Liquid biofertilizer for farmlands and gardens, reclaimed grew water for discharge and groundwater recharge	https://www.cyclifier.org, project/flintenbreite- neighborhood/ http://www.susana.org/ _resources/documents/ default/2-59-en-susana-cs germany-luebeck- ecological-housing-bobx. pdf accessed on 30 September 2021
Hannover, Germany	Treatment wetland for greywater	Vacuum toilet, urine-diverting toilets	Reclaimed water for toilet flushing	https://www.susana.org/ _resources/documents/ default/2-1986-en-ecosan- pds-007-germany- hannover-oekotechnikpark 2005.pdf accessed on 30 September 2021
Lima, Peru	Composting of organic waste, vermicomposting for solid fraction of blackwater, treatment wetland for greywater, treatment wetland for liquid fraction of blackwater	Urine-diverting toilets, solid separation unit	Treated blackwater for irrigation of lawns, reclaimed greywater	https://www.susana.org/ _resources/documents/ default/2-70-en-susana-cs peru-lima-sanchristoferus 2009.pdf accessed on 30 September 2021
Grecia Salentina area, Italy	Composting from house organic waste; vermicomposting; home composting; community compost		Compost/vermicompost for home and city use	http://www. compostcommunity.it/wp content/uploads/2021/01 brochure_inglese.pdf accessed on 30 September 2021
Vienna, Austria	Anaerobic treatment and composting of kitchen waste and green waste from urban green areas		Compost for urban green areas, gardens	https: //www.wenigermist.at/ biomuell-und-speisereste- richtig-entsorgen accessed on 30 September 2021
Ljubljana, Slovenia	Anaerobic treatment and composting of kitchen waste and green waste from urban green areas		Compost for urban green areas, gardens	http: //www.rcero-ljubljana.eu accessed on 30 September 2021
Mālpils, Latvia	Vermicomposting from different types of biowaste (e.g., sewage sludge, manure, leaves)		Compost for urban green areas, gardens	https://smartcitysweden com/best-practice/192/ biowaste-treatment-by- vermicomposting/ accesse on 30 September 2021
Culemborg, EVA-Lanxmeer, the Netherlands	VF treatment wetland (greywater)		Reclaimed water	https: //edepot.wur.nl/180531 (In Dutch) https://www. urbangreenbluegrids.com, projects/eva-lanxmeer- results/ accessed on 30 September 2021
Cressy, Switzerland	Composting unit VF treatment wetland (greywater)	Dry toilet	Compost, reclaimed water	https://www.cooperative equilibre.ch/projets/cressy accessed on 30 September 2021

 Table 1. Summary of the NBS case studies reported in the present paper.

Case Study	NBS Units	Supporting Units	Product and Reuse	References			
Hamburg, Jenfelder au, Germany	Anaerobic treatment (CSTR)	Vacuum toilets	biogas	[38]			
Helsingborg, H+, Sweden	Anaerobic treatment (UASB); aerobic GW treatment	Vacuum toilet; struvite precipitation; ammonium stripper	Struvite, organic fertilizer, ammonium sulfate, biogas	http://run4lifeproject.eu/ accessed on 30 September 2021			
Sneek Lemmerweg, the Netherlands	TAD (Thermophilic anaerobic treatment) (UASB)	Ultra-low flush vacuum toilet	Hygienized effluent containing fertilizers	[39]			
Wageningen, NIOO, the Netherlands	Anaerobic treatment (UASB) Pilot Photobioreactor	Vacuum toilets	Biogas, algae biomass, reclaimed water	https://www.saniwijzer.nl/ (In Dutch) accessed on 30 September 2021			
Oslo, Klosterenga, Norway	Septic tank Aerobic biofilter Horizontal subsurface flow treatment wetland		Reclaimed water	https://www.susana.org accessed on 30 September 2021 https://www.susana.org/ _resources/documents/ default/2-248-jenssen- urban-greywater-oslo-en. pdf accessed on 30 September 2021			
Gent, Belgium	Anaerobic treatment Aerobic treatment	Struvite precipitation, Vacuum toilets, Membranes	Struvite; reclaimed water, biogas	http://run4lifeproject. eu/accessed on 30 September 2021 Democase Gent—Nereus Project (nereus-project.eu) accessed on 30 September 2021			
The Hague, Rijnstraat, the Netherlands	Anaerobic treatment	Struvite precipitation vacuum toilets Waterfree urinals	Struvite, biogas	http://www.saniwijzer.nl (In Dutch) accessed on 30 September 2021			
Hamburg, BIQ, International Building Exhibition (IBA), Germany	Photobioreactor	Flotation unit, heat exchanger, an external biogas plant	Algae biomass, heat for heating and sanitary water, heat and sound insulation	https://www.archdaily. com/339451/worlds-first- algae-bioreactor-facade- nears-complet accessed on 30 September 2021			
Tampere, algal ponds, Finland	Algae ponds	Source-separation of urine	Algal biomass for fertilizer use	https://www.vanajavesi.fi/ levasieppari-hanke- ravinteet-talteen-ja- kiertoon- luonnonmukaisesti/ accessed on 30 September 2021			
erlin, Block 6, Germany	Blackwater treatment Greywater treatment Urban farming	Heat recovery from greywater	Heat and reclaimed water, organic fertilizer	http://www.roofwaterfarm com/en/block-6/ accessed on 30 September 2021			
Pogradec Albania- mineralized soil and Prrenjas, Elbasan Albania contaminated soil	Phytomining-Agromining	-	Nickel salt, energy, inorganic fertilizer	https: //www.life-agromine.com/ accessed on 30 September 2021 [40–42]			

Table 1. Cont.

Observed Co-Benefits and Limitations

The co-benefits of TWs arise mainly from the presence of plants, which contribute to the mitigation of heat islands via evapotranspiration and provide habitat for insects, birds, and other wildlife, thus increasing biodiversity, sequestering carbon in their biomass and enabling its reuse. Plants in TWs also play an important role in the aesthetic appearance of the plant and its integration into the landscape [43]. TWs can also be used for the mitigation and treatment of combined sewer overflow, thus reducing floods [44]. An additional benefit in the case of green walls is the added insulation effect for buildings

when placed on the exterior walls as well as a potential thermal regulation effect if systems are operated indoors [28,45,46].

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

TW are addressing numerous urban challenges: via wastewater, they provide reclaimed water for irrigation or fertigation, thus contributing to the restoration and maintenance of the urban water cycle [47]. Additionally, the produced plant biomass can be composted, contributing to nutrient recovery and reuse, or used for energy production [36,47]. TW plant biomass can be used as construction material in ecologically oriented construction as raw material for rooves, but different materials can also be produced from it, such as composite panels or insulation material [48–50].

3.1.2. Photobioreactors

Working Principle

Photobioreactors (PBRs) for nutrient recovery from wastewater are autotrophic wastewater treatment systems housing organisms such as microalgae, which tolerate high loads of wastewater, enable pathogen inhibition, carbon dioxide (CO_2) capture, and oxygenation, as well as valuable biomass production [51,52]. Mainly two types of PBRs can be utilized as NBS units: open raceway ponds or closed panel systems (tubular/flat). Open raceway ponds are low-cost systems with shallow water depth units and paddlewheels or blower pumps for aeration. Natural sunlight is usually preferred for illumination; however, greenhouse settings supplemented with artificial light (LED or similar) are common. Closed systems are generally more expensive. Controlled units with CO_2 supplements and continuous monitoring of system parameters such as light intensity, dissolved oxygen concentration, temperature, etc. are needed, which may increase operational costs. In PBRs, microalgae and aerobic bacteria can have symbiotic interactions with the exchange of different organic and inorganic compounds, such as minerals, vitamins, and gases. Green microalgae are primarily autotrophs; however, some species can grow as heterotrophs in the absence of light and thus compete with bacteria for organic sources or carbon. Microalgae growth depends on the temperature, concentration of mineral nutrients, pH, and intensity and duration of illumination [53,54]. Thus, domestic wastewaters usually lack the carbon required to remove all nitrogen by assimilation into algal biomass, indicated by elevated daytime pondwater pH, resulting from inorganic carbon assimilation causing a shift in the carbonate system equilibrium and release of hydroxide ions, which can increase the pondwater pH to >10. However, the optimum range of pH and dissolved organic carbon can be regulated by the injection of CO_2 , enhancing algal production, promoting aggregation and bio-flocculation of algae with bacterial flocs to further enhance algal settling [55] and nitrogen removal by providing the necessary carbon to stimulate algal growth and reduce pH [56]. In PBRs, optimal conditions for microalgae growth should be maintained to achieve maximum efficiency of the system including vital algae inoculum, sufficient light availability for algae growth, hydraulic retention times (up to 20 days), and the surface area needed for algae ponds (20 g dry weight $m^{-2} d^{-1}$), while avoiding contamination by fungi and zooplankton [57–59].

In- and Outputs

In terms of inputs, PBRs can receive primary, secondary treated wastewater, digested effluent of municipal wastewater, and anaerobically treated blackwater and urine [60–62], liquid digestate from a biogas plant, and different sources of CO_2 from, e.g., tailpipe CO_2 from cars/buses/trucks or combusted CH_4 gas if located in the city. PBRs can also treat the effluent of tertiary treated water. However, the very dark color of the wastewater can limit light availability for algae growth, and it should be considered regarding inputs. The quality of the listed influents depends on the requirements of national legislations. Under optimal operational conditions, the main outputs are treated municipal wastewater, grey/blackwater, urine, treated digestate, and efficiently harvested algae or

algae-bacteria biomass, which can be used as an organic fertilizer in agriculture, biofuels, biopolymers, animal feed, bio-stimulants, and substances such as pigments for cosmetic and pharmaceutical industries [52]. Algae biomass contains micro- and macronutrients, especially N, P, and K, and might be considered as an organic slow-release fertilizer [63]. Prior to reuse, algae biomass can be anaerobically digested for energy recovery [64,65].

Connected Units

Anaerobic treatment can be connected to produce an input stream for the PBR containing mainly nutrients and non-biodegradable chemical oxygen demand (COD). PBR effluents can be connected to a primary settler of raw wastewater to remove organic solids as a pre-treatment unit and harvesting unit to separate algae or algae-bacteria biomass from liquid, followed by additional effluent polishing if required by the legislation or end-users by (a) UV disinfection, (b) sand filter for final solids removal, (c) activated carbon unit, (d) advanced oxidation processes, and (e) membrane filter for efficient removal of specific pollutants or to provide a high-quality effluent, suitable for many re-use applications. After harvesting, the biomass can be additionally treated to meet specific requirements in a maturation pond for further solar-UV disinfection, used as storage before discharge or subsequent re-use [66]. The biomass, if not used as fertilizer or in industrial use, can be further treated in an anaerobic digestor to produce biogas and in a sludge drying bed to produce a soil amendment. The final effluent from PBRs can be used for the irrigation of street trees and urban parks, urban agriculture, while algae or algae-bacteria biomass can be used in urban agriculture. Both products can be used in any other NBS unit to cover water and fertilizing needs.

Case Studies and Literature

PBRs are applied on micro-, meso-, and macro scales with different end users, from communities to water utilities. However, due to the high surface size requirements of PBRs (e.g., $0.3 \text{ m}^3 \text{ m}^{-2}$), their application in the city is rare. However, Sutherland et al. (2020) report that the optimum size for maximum productivity is considerably smaller than the current full-scale systems, suggesting that a combination of mixing frequency and higher photosynthetic potential under low light conditions was the main driver of enhanced productivity. This has implications for commercial-scale systems also located in the city, with respect to capital and operational costs (e.g., comparison between different PBRs scales of 5 m^2 , 330 m^2 , 1 ha; [62]). It was reported that a full-scale pilot project, SolarLeaf façade, was installed on the BIQ house in Hamburg in 2013 (https://www.archdaily.com/3394 51/worlds-first-algae-bioreactor-facade-nears-complet, accessed on 30 September 2021), consisting of bioreactors to form a secondary façade, providing around a third of the total heat demand of the 15 residential units in the BIQ house.

Observed Co-Benefits and Limitations

Harvested algal biomass has several multi-valorization pathways, such as bio-stimulants and fertilizer for soil amendment, feed for different animal groups, and bio-composites for construction purposes. There are also several co-benefits while treating wastewaters such as microplastic, contaminants of emerging concern (CEC), and pathogen removal. As a matter of fact, biodegradation and photodegradation are the most important removal pathways for CECs, achieving up to 90% removal efficiency [67]. However, several risks can influence the outputs. Among them, fungi and zooplankton contamination, inappropriate pH range, inefficient CO_2 injection and inefficient harvesting, seasonal algae die-off, and self-shading are the most usual ones.

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

Photobioreactors allow sustainable biomass (algae or algae-bacteria) generation while upcycling nutrients that are readily available in urban wastewater. As a result, there is a dual benefit of urban circularity, avoiding emissions for making new fertilizer or feed compounds while achieving treated wastewater with reduced costs. When photobioreactors are operated in autotrophic mode, significant amounts of CO_2 capture can be achieved, helping to reduce urban emissions generated as industrial flue gas or from transportation-related activities.

3.1.3. Anaerobic Treatment

Working Principle

Anaerobic digestion combines the treatment of contaminated waste streams with the production of energy and nutrients in a recoverable form. In oxygen absence, different groups of microorganisms (the biomass or sludge) cooperate to transform complex organic matter in four sequential steps to methane and CO_2 (biogas). In the process, the complex organic matter is firstly hydrolyzed and subsequently fermented (acidogenesis and acetogenesis) to substrates (acetate, H_2/CO_2 , C1 compounds) which can be converted to methane (methanogenesis). In the process, nutrients bound to the organic matter are released in their water-soluble forms (ammonium, phosphate), which are not further converted under the conditions applied. These nutrients are available for recovery and reuse as, for example, fertilizers (provided that the salts do not precipitate in the sludge). For soluble organic matter such as glucose, amino-acids, and volatile fatty acids, the conversion to biogas takes place in a similar way, starting with (depending on the nature of the organic matter) acido-or acetogenesis [68].

Usually, two different systems are distinguished for continuous anaerobic treatment: low-rate systems without biomass retention, which are completely mixed and applied for input flows that have high suspended solid and COD concentrations (>50 g/L), and high-rate systems with biomass retention that are fed with input streams with lower COD concentrations.

In- and Outputs

A large variety of input streams can be treated anaerobically, if certain conditions are met. Organic matter should be biodegradable for an important part and more or less free of inhibitory compounds. Anaerobic systems are currently operating for a variety of inputs ranging from domestic and industrial wastewaters, agro-industrial plant residues, manure, sewage sludge, and (fractions of municipal) solid waste. The application scale is also highly variable; large-scale systems are operating for industrial and municipal wastewater treatment. UASB is one of the suitable processes for both carbon removal and energy recovery from domestic wastewater streams [69,70]. In the past decades, smaller systems have been installed for black (toilet) water (BW) treatment as a result of the implementation of source-separated sanitation concepts [9]. The production of volatile fatty acids (VFA) from, for example, primary sludge [71] and also polyhydroxyalkonates production [72] can be an alternative to biogas production.

Connected Units

Conventionally collected, low-concentrated domestic wastewater can be treated anaerobically when tropical conditions are prevailing [73]. For low-temperature climates, lowflush toilets such as vacuum toilets need to be installed, in combination with the separation of greywater from blackwater, to provide a concentrated blackwater suited as an influent for a heated (mesophilic) anaerobic treatment system [74]. Regardless of the end-product, anaerobic treatment usually requires post-treatment. In general, effluent COD concentrations are too high to allow for direct use of the effluent, for example, as a nutrient-rich solution for fertilization of continuous (all year round) crop systems. Supporting units such as struvite precipitation for P recovery, ammonia stripping/absorption for nitrogen recovery, and aerobic treatment can be included after the anaerobic treatment. Moreover, pathogens can be present (depending on the input substrate) in high concentrations. Posttreatment for organic micropollutant and pathogen removal is needed to ensure high end-product quality. Gas treatment needs to be installed (for desulfurization) and odor control needs to be ensured. Different post-treatment supporting units are presented in Supplementary Materials B.

Literature Case Studies

Various NBS systems on a micro-, meso- and macro scale (Table 1, based on Kisser et al. [9]) have been implemented in cities for the recovery of resources (N, P, energy, organics, water) from household waste (water) streams, in which the anaerobic mesophilic NBS unit is the core technology of the system. Recently, on a scale of ca. 2000 people (meso scale), apartment buildings in Helsingborg (H⁺ project) were equipped with source separated sanitation and mesophilic anaerobic treatment of blackwater and (in a separate unit) food waste. Energy and organic fertilizer are produced during anaerobic treatment while struvite and ammonium-sulfate are recovered from the anaerobic effluents via, respectively, a struvite precipitation and a stripping/absorption unit (Table 1). In Sneek, the housing project at the Lemmerweg [75] was recently upgraded with ultra-low-flush vacuum toilets connected to a thermophilic anaerobic system for hygienized fertilizer recovery from blackwater.

In Stockholm, primary settled wastewater from Henriksdal WWTP was pre-treated in UASB reactors followed by partial nitrification/anammox for mainstream nitrogen removal [76].

Observed Co-Benefits and Limitations

Energy is produced in the form of biogas, and it can be applied to increase reactor temperatures, minimizing external energy demands. Nutrients are retained in the effluent and can be recovered for use as a fertilizer in agriculture either as is (directly) or after the application of a recovery supporting unit (e.g., struvite SU 3 and ammonium after stripping SU 4 in Supplementary Materials B). Sludge can be reused in agriculture after a disinfection step. Streams with a low COD concentration and low temperature (for example, conventionally collected domestic sewage) produce too low amounts of biogas to increase the process temperature. Therefore, large reactor volumes are needed. Source separation of blackwater using low-flush toilets (e.g., vacuum) can tackle this issue.

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

Our food is grown in agriculture with nutrients or comes from animals fed with agricultural products, and these nutrients are excreted after consumption via urine and feces or end partly in kitchen waste. The recovery of nutrients through the application of anaerobic treatment of domestic waste and wastewater streams followed by the abovementioned NBS units and subsequent use in agriculture contributes to a circular economy. Since anaerobic treatment is used with the aim of reaching energy neutrality, no additional energy is required, and in some cases, an excess of energy is produced.

3.1.4. Aerobic Treatment

Working Principle

Aerobic treatment is based on the oxidation of organic material and nutrients (for example, nitrogen) by micro-organisms. Carbon is oxidized to CO_2 , and biomass (sludge) and nutrients are removed via a combination of denitrification/nitrification for nitrogen and via chemical or biological P removal. In some cases, the process is limited to mainly nitrification as it is in the VUNA process for recovery of nutrients from human urine [77].

The process can be performed in several types of aerated reactors. Often, it is combined with a settler or a membrane process (SU 7 or SU 8 in Supplementary Materials B) to ensure high effluent quality. Due to the recirculation of effluents from the membrane and biological processes, this combination is sometimes described as one technology.

In- and Outputs

Aerobic treatment is most suitable for diluted wastewater streams, for instance, separately collected domestic greywater; the effluent from blackwater treatment or a combination of the two can be used as input streams. The main output of the process is the treated water. This water has a high quality and with further polishing can be safely reused. Furthermore, sludge is produced.

Connected Units

The incoming stream is most often first treated or collected in a settling tank. The aerobic treatment is sufficient to reach wastewater effluent standards. However, the aerated tank can further be connected to a membrane filter or micropollutant removal system (e.g., UV) (SU 5 or SU 8 in Supplementary Materials B) for high effluent quality to increase possibilities for reuse. Furthermore, because greywater can have high temperatures, it has great potential for heat recovery; therefore, heat exchangers can be integrated in the system.

Literature Case Studies

Various aerobic treatment systems on mesoscale have been implemented in cities for the recovery of water from diluted household wastewater streams (Table 1). In Helsingborg (H+ project), a membrane bioreactor is used to treat domestic greywater, and in Gent, a similar system has been implemented to treat a combination of domestic greywaters and effluents from blackwater treatment (Run4Life EU project—Recovery and utilization of nutrients 4 low impact fertilizer-https://cordis.europa.eu/project/id/730285, accessed on 30 September 2021, n.d.). The effluent produced in Gent will be reused in a soap factory (Nereus Project-new energy and resources from urban sanitationhttps://www.nereus-project.eu/, accessed on 30 September 2021, n.d.). In Berlin, a moving biofilm bed reactor has been applied in combination with heat recovery and UV disinfection to reuse heat and water for toilet flushing [78]. In the neighborhood Klosterenga in Oslo, the greywater of an apartment building is treated by an aerobic bioreactor in combination with a porous media filter and a subsurface TW. The effluent is used in a local garden with a playground for kids (Peter, D.J.; n.d.). In Sneek, the Netherlands, an aerobic treatment in combination with nanofiltration was tested for 6 months and reached high effluent quality as well (Sanimonitor, n.d. https://www.sanimonitor.nl/rapportageprojecten/noorderhoek-grijswaterbehandeling---nanofiltratie/detail_data=495, accessed on 20 April 2021). These case studies show that in combination with different connecting units, aerobic treatment is well suited for reuse purposes of water in the city.

Furthermore, aerobic treatment can be used for the treatment of separately collected urine. As part of a fully operational urine-separating sanitation system (TRL 7) at EAWAG, Switzerland, two moving bed bioreactors (MBBR) have been in operation for several years [77]. Regarding the removal of pharmaceuticals from the treated urine, a post-treatment step is necessary, e.g., by powdered activated carbon (PAC) [79].

Observed Co-Benefits and Limitations

The benefits of using aerobic treatment are: the high effluent quality and therefore possibilities for reuse of wastewater; the compactness of the reactors; and due to the removal of most of the organic substances, the lower chance of the re-growth of micro-organisms and odor problems occurring [80]. Furthermore, aerobic treatment is mostly suitable for the treatment of greywater. By separately collecting greywater from residential buildings, a high-temperature stream is created, which makes aerobic treatment ideal for combination with heat recovery.

Constraints of aerobic treatment are the usually high sludge production and often the need for an external C source dosing for denitrification. When treating the effluent of a blackwater treatment, this last constraint can be solved by applying a nitritation/anammox on the blackwater effluent before it enters the aerobic treatment. As the nitritation/anammox can remove 70–90% of N of the blackwater effluent stream [81–83], it reduces the need for an external C source in wastewater treatment. Further constraints are mainly found in financial, legal, social issues: high costs for the implementation of wastewater separation in existing buildings, the legal obligation of water quality for reuse, and the willingness of end-users to use recycled water. Another issue that can arise is the pharmaceutical content. In Sneek, it was seen that aerobic greywater treatment was not sufficient to remove certain pharmaceuticals [84]. This is an area for further consideration.

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

Aerobic treatment (in combination with connecting units) provides the possibility to reuse diluted wastewater streams. This allows for the circular use of water instead of linear use. Moreover, it can supply a constant flow (all year round) of clean water which can be beneficial in reuse purposes in, for instance, industries. Furthermore, the circular economy aims at minimizing the input of energy. As aerobic treatment is ideal for use in combination with heat recovery from wastewater, it decreases the need for energy consumption for heat production in residential areas.

3.2. Solid Incoming Streams

3.2.1. Composting and Vermicomposting

Working Principle

Composting comprises (principally) aerobic processes for the oxidation of organic matter or biosolids in (mainly) end product amenable to resource recovery with a minimum capital investment and relatively small operating commitment, with the aim to stabilize the organic matter in the product (compost), reduce the number of pathogens, and to obtain a relatively dry product [85,86].

At the beginning of the process, mesophilic bacteria naturally present in the input waste or inoculated decompose the readily biodegradable fraction of the organic matter. During these initial stages, the temperature of the compost keeps increasing up to 60 °C [85,86]. Thereafter, both thermophilic bacteria and fungi take over degradation of the remaining biodegradable matter. The high temperature ensures a hygienization of the compost product [85]. After most of the readily biodegradable matter has been modified, during the cooling stage of the process, mesophilic bacteria and higher organisms continue the breakdown of the organic matter to finally reach a maturation phase in which the compost is completely stabilized. The process as a whole can take several weeks to months [85,87] and high-quality compost is related to its stability and nutrients content. Vermicomposting also involves the oxidation of organic matter, resulting in smaller volumes, but in that process, worms (e.g., *Eisenia fetida, Perionyx excavatus, P. sansibaricus, E. andrei, Eudrilus eugeniae*) are the main actors (alongside normal microbial biomass; [88]). As worms, in general, are not heat tolerant, vermicomposting usually does not include a thermophilic phase [89].

In- and Outputs

Composting can be a simple process on small-scale (home composting) to controlled large-scale operations. For home composting, usually, a bin (or heap) is filled up with fresh material and compost is used as the starter. For larger-scale composting, confined boxes or tanks and tunnels could be used.

Typical waste streams to be composted are vegetable materials, crop residues, dry (no water or urine) feces, biological sludge from wastewater treatment plants, and green cuts, with a dry matter content higher than 40% and a C/N ratio ranging from 25–30 [90]. Aeration is performed manually (e.g., by waste overturning) during the process and the operator manages the input by ideally alternating the addition of readily biodegradable material with more resistant lignocellulosic inputs. The temperature of the bin/heap is not controlled (but can be steered by addition of readily biodegradable organic matter), nor is the humidity.

In the case of vermicomposting, worms need to be added. The temperature needs to be controlled in the mesophilic temperature range. Vermicomposting relies on the worms to mix/aerate and fragment the input. The worms are light-sensitive, so conditions should be controlled.

Larger-scale composting facilities decrease the size of the input material to (usually) <5 cm, and the input material is mixed with an inoculum (compost), and bulking material with a high C/N ratio to increase the passive aeration or facilitate the active one. Humidity is controlled in these systems. Off gas air is usually treated in biofilters. The composition of the output material depends largely on the quality of the input materials but largely 30–60% of the input carbon is oxidized to CO_2 during composting [91].

Case Studies and Literature Case Studies

Thousands of municipalities in Italy apply so-called "kerbside collection programs", focusing on food waste collection. This approach is based on small-volume kitchen caddies fitted with biodegradable bags (i.e., compostable bioplastic or paper liners); collection is performed at the kerbside (or door-to-door collection) and adopting convenient frequencies aimed at enhancing citizens' participation in composting. This strategy is also used in the absence of a plant dedicated to the treatment of organic waste. In these cases, some municipal administrations (i.e., the province of Lecce, which does not have a dedicated plant) have equipped themselves with community composters, which allow to autonomously treat part of the organic waste produced on a community level by reducing both the production of waste and the costs of transport [92] (Community Composters, 2021).

In other EU countries, home composting is a common practice. For example, 48% of people in Slovenia were reported to have home composting systems [93]. The home process is also used alongside urban composting plants (Spain; [94]). A community composting project in the city of Bratislava, Slovakia, demonstrated the importance of cooperation among the various stakeholders and citizens interested in composting their own bio-waste, and resulted in a reduction in the amount of bio-waste in mixed municipal waste.

The Malpils Biotechnology Centre (in Latvia) is involved in biowaste treatment using the method of vermicomposting and production of organic fertilizer from it. The main aim is to study the problem of how to process biowaste in all its complexity in order to produce a high-quality product from different types of biowaste (e.g., sewage sludge, manure, leaves) by covering the whole treatment cycle, from the collection of the biowaste to the final treatment and selling of the fertilizer. As the method of vermicomposting is not widely used around the Baltic Sea, further studies should be carried out to find the most efficient ways to adapt it to Latvia's waste management needs. The pilot project has elaborated the technology for the preparation of substrates suitable for feeding the earth worms used for improving compost quality [95].

Observed Co-Benefit and Limitations

Composting inevitably generates some emissions, such as gases and bioaerosols. The gases include CO_2 , CH_4 , N_2O , sulfur compounds, and many other volatile organic compounds (VOCs) that can be odorous [96] and should be considered as a potential nuisance for the neighborhood [97]. In composting facilities, there is a huge bioaerosol production; however, currently, there is no evidence of the toxicity of these bioaerosols, and the risk to nearby residents thus cannot be quantified [98].

Composting may also be a source of microplastics in the environment. Gui et al. [99] found that "rural domestic waste compost was a significant source of microplastics in soils, and the microplastics in compost products were closely related to the quantity and type of plastic waste present in rural domestic waste".

Composting and vermicomposting has various effects on heavy metal concentrations in the end product. Some papers demonstrate an increase in metal (Cd, Cu, Pb, Zn) concentrations, others show a decrease [100–103]. Composting also tends to stabilize metals [104] with redistribution from relatively labile to more immobilized states. For this reason, all compost should go through quality control before use. Finally, issues related to leachate infiltration and runoff must be considered since they can be emitted during the process if not well managed [97].

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

Compost application is a way to improve soil health by enhancing its organic matter (critical for most soil functions such as soil structure, water purification and regulation, carbon sequestration and regulation, biodiversity, and nutrient cycling), and microbial diversity, as well as soil fertility and soil health, even in cities. Moreover, it is also a way to prevent the waste of raw materials and to reuse them. The compost produced by households or small communities can be used at the local level. In this way, citizens may benefit from a good-quality fertilizer and soil improver, such as compost/vermicompost, for use in their gardens or vegetable plots, avoiding disposal. This is a typical example of closing loops locally. However, home composting requires people to have some knowledge of good composting practice in order to avoid unnecessary environmental impacts and to ensure good-quality of waste separation and citizens' management of the composting process (EEA, 2020), and to teach how to compost waste materials [105]. The challenge in cities is to use it as a soil improver and fertilizer and the acceptability of producing it in urban areas because of its smell and low handleability.

3.2.2. Decentralized Solid Waste Anaerobic Treatment in Urban Areas Working Principle

Solid waste anaerobic digestion (SWAD) is a biological process that breaks down residual organic material (OM) via microorganisms in the absence of oxygen. The AD of organic material basically follows hydrolysis, acidogenesis, acetogenesis, and methanogenesis biochemical steps. Volatile fatty acids (VFA) formed after the acidogenesis step are intermediates in the process of conversion of biodegradable OM to methane. By the inhibition of the last steps (i.e., acetogenesis and methanogenesis) in the anaerobic conversion, VFA can accumulate in the system and as such can be harvested as an end product. SWAD produces biogas, a methane-rich gas that can be used as a fuel, and digestate, which is a source of nutrients that can be used as a fertilizer. Biogas can be converted to heat and electricity through combined heat power (CHP) engines, while the digestate can be further processed to separate water from the solid containing nutrient fraction using techniques such as a settling tank or electro-coagulation. The use of AD on a microscale is very much implemented in low- and middle-income countries; however, nowadays, one sees a trend regarding its application in urban areas of developed countries (e.g., A DECentralized management Scheme for Innovative Valorization of urban biowastE (DECI-SIVE) H2020 EU project http://www.decisive2020.eu/, accessed on 30 September 2021; https://cordis.europa.eu/project/id/689229, accessed on 30 September 2021).

In- and Outputs

Organic waste represents one of the largest fractions of the municipal waste mass: from 14% to 47% in the European countries, and more than 60% in developing countries. Urban biowaste such as food waste, and the organic fraction of municipal solid waste and co-substrate (lignocellulosic biomass: green waste from private gardens, green waste from public areas, paper towel from mass and commercial catering, etc.) can be used as feedstock. In some cases, urban waste is mixed with blackwater in order to generate a slurry [106]. Regarding the outputs, two types of streams are generated: the digestate that needs to be further processed to separate the liquid from the solid fractions and the biogas (a mixture of methane and CO₂ with traces of impurities such as hydrogen sulfide).

Connected Units

An efficient collection, storage, and pre-treatment network is required to supply a constant quantity of organic waste with the best quality to the AD process. Therefore, after collection, the organic waste needs to be stored and pre-treated in order to improve its digestibility while minimizing potential odorous nuisances [106]. The management of the output streams involves the post-treatment of digestate and biogas. Usually, digestate

cannot be used directly in most urban areas, and this requires the implementation of a solidliquid separation supporting unit able to generate a liquid stream as well as a solid stream. The liquid fraction could be treated for fertilizer recovery (i.e., struvite precipitation and/or ammonia stripping). In addition to the chemical process, TW or classical aerobic treatment, the liquid digestate may be further processed with PBRs and aquaponics [107,108]. Solid digestate is composted before being used for urban applications [108,109]. For the valorization of biogas, the most common application is the production of heat and electricity by a CHP unit that usually requires to upgrade the quality of the biogas, mostly by using a H_2S filter.

Literature Case Studies

Over the last decade, there have been several examples of moving from goods import and extra-urban waste management to a more urban network allowing circular local and decentralized valorization of biowaste, enabling energy and bioproduct production for local uses (e.g., DECISIVE H2020 EU project https://cordis.europa.eu/project/id/689229, accessed on 30 September 2021). While waiting for the outcomes of that project, several recent publications have reported on the application of decentralized anaerobic digestion of urban organic solid waste at a pilot scale (see Angeli et al. [106] for an overview and Nguyen et al. [110], Walker et al. [111] and Gonzalez et al. [112] for more recent pilot-scale studies). For instance, Walker et al. [111] reported the implementation of micro-scale AD fed on food and catering waste in London (UK). The pilot system was a 2 m³ singlestage digester containing an automated mechanical mixer and heated by an internal water heat exchanger, operated with the necessary input and output process units allowing to store/feed (average OLR of 1.6 kg VS m⁻³ d⁻¹) and safely manage the output streams. The biogas plant monitored over 319 days could process 4574 kg of food waste while producing 1008 m³ of biogas at an average of 60.6% methane. Nguyen et al. [110] operated a two-stage anaerobic digestion system in Ho Chi Minh city (Vietnam) which included a feed tank (0.4 m^3) , a hydrolysis reactor (1.2 m^3) , and a methanogenic reactor (4.0 m^3) . The reactor was fed with biowaste diverted from municipal solid waste collected from households and restaurants with an organic loading rate (OLR) ranging from 2.5 to 3.8 kg vs. $m^{-3} d^{-1}$. The highest biogas yields 263 ± 64 L kg⁻¹ t COD removed were obtained at an OLR of 2.5 kg vs. $m^{-3} d^{-1}$. It is expected that a full-scale 2S-AD plant with a capacity of 5200 tons day⁻¹ of biowaste collected currently from municipal solid waste in Ho Chi Minh city may create daily electricity of 552 MWh, thermal energy of 630 MWh, and the recovery of 16.1 tons of NH_4^+ -N, 11.4 tons of organic N, and 2.1 tons of TP as both organic liquid and solid fertilizers.

Observed Co-Benefits and Limitations

A pre-digester tank to store the feedstock collected and to feed the AD is required to buffer the irregularly collected volume of biowaste; however, the storage duration that may affect the AD performance and odor should be controlled [113]. Gonzalez et al. [112] reported that a temperature increase of the feed to process conditions requires a significant amount of thermal energy, which strongly affects the efficiency of the process when operated at a low organic load. However, the amount of energy consumed may be limited if the micro-scale AD process is operated in a greenhouse in temperate climate conditions [111]. With this approach, Walker et al. [111] reported a net positive energy balance and potential coefficient of overall performance (COP) of 3.16 and 5.55 based on electrical and heat energy inputs and outputs, respectively.

On-site heat and electricity production can fully benefit the housing infrastructure surrounding the micro-scale AD [114]. Walker et al. [111] reported that the most important contribution of micro-scale AD was the limitation of greenhouse gas emissions by the avoid-ance of on-site fossil fuel use, followed by the diversion of feed waste from landfill and that the plant could result in a carbon reduction of 2.95 kg CO_{2eq} kW h⁻¹ electricity production.

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

If the AD of the urban organic materials is combined with the development of urban agriculture, the biogas can be burned in a combined heat and power unit, allowing to comply with the heat demand of greenhouses while the digestate separated in liquid and solid fractions could be valorized. The liquid fraction is available for hydroponic facilities and the solid fraction is amended on soils after further treatment [107,108]. All these outputs may potentially mitigate several urban circularity challenges, as detailed by Atanasova et al. (2021). However, all stakeholders shall be involved in the project design and implementation for ensuring the success of decentralized urban organic waste treatment and valorization [115]. Finally, a method to design decentralized and micro-scale anaerobic digestion efficient networks in urban areas is still needed [116].

3.2.3. Insect Farming

Working Principle

Instead of composting or vermicomposting, nutrients in organic waste can also be converted by applying insect larvae [117]. There are several types of insects suitable for insect farming, e.g., mealworms, black soldier flies (BSFL), houseflies, crickets, waxworms, etc. [118]. To upcycle organic waste with insect larvae, the waste is ground into small particles or converted to a liquid or pasty state. The insects are bred in a nursery wherein the eggs take a certain time to hatch; for BSFL, it takes around four days. The recently hatched larvae (1–5 days old) are put together with the pretreated waste and start to feed on the organic matter. Depending on the quantity and quality of the waste, the larvae will need at least 12–16 days to reach the full size of around 0.5 cm width and 2.5 cm length. To treat 60 kg of waste, approximately 40.000 BSFL and 1 m² of space is needed. The larvae consume 100–125 mg/feed/day [119]. In the last larval stage, they are harvested. The larvae are quite resilient and can withstand changes in the environment, but for a fast waste conversion and high product yield, optimal surroundings are of advantage. Optimal conditions are influenced by the chosen insect, container dimensions, temperature, larval density, humidity, feeding rate, feeding interval, and type of feed [120]. The ideal operating conditions for BSFL are temperatures between 24 and 32 °C, a moisture content of 60–90%, and a shady environment [121]. The eggs or larvae can easily be bred on the production site. The space needed for 1 ton of incoming waste per day is around 50 m^2 for the breeding facility and 100 m² for the waste-processing spatiality [121].

In- and Output

Many insects can grow on a variety of biowastes such as animal manure, human excreta, fruit and vegetable waste, municipal organic solid waste, and milling and brewery side streams [122,123]. Those are therefore all possible input materials, although for the use of the larvae as animal feed or food, the selection is smaller due to regulations. Protein-rich larvae or extracts from those are the main output and can be used for feeding fish in hydroponics, poultry and pets, as a delicacy or food supplement [124,125]. The residual biowaste (mixture of unassimilated material and larvae excrement) can be used as a fertilizer and soil amendment in urban gardens and farms [126] or can also be converted in biogas units. Depending on the type of input, sometimes, some liquid fraction is also produced [122]. Low-value waste is thus transformed into high-value products with diverse potential applications.

Connected Units

Beforehand, depending on the incoming waste, some pretreatment of the input stream can be required to obtain the ideal composition for the insects (e.g., shredding, separation, moisture content adjustments) (Figure 2). Usually, the waste is shredded to a particle size of less than 1–2 cm, which can be performed with a simple gadget such as a hammer mill. After feeding in the main unit, the larvae are separated from the substrate through sieving. This can be performed manually on a small scale or with automatically shaking sieves.

The obtained larvae need to be further processed before they can be sold, either by drying, mixing them with other ingredients and producing pellets, or by extracting proteins and fats with more complicated processes [121]. The residual biowaste can be directly used as fertilizer but can also be further treated in an anaerobic digestion for the production of biowaste.

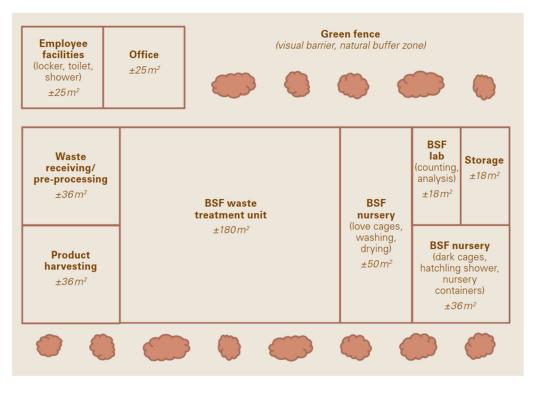


Figure 2. Draft of a BSFL farm and size estimation for the conversion of 2 tons of biowaste per day (from Dortmans et al. [121] (CC BY 4.0)).

Literature Case Studies

In Europe and North America, there is more resistance towards insect farming, and the legislation is an additional restriction [127]. Due to legislation, there are no cases to be found in Europe of insect farming on domestic waste. On the contrary, in many countries in Asia, Latin America, and Africa, the use of insects is widely accepted and even already applied; however, most examples of insect farming concern larger operations [128]. In the University of Catolica de Santa Maria in Peru, a research center has been created to form a basis for the BSFL industry in the country. The company Nasekomo in Sofia, Bulgaria, is producing BSFL on agricultural by-products for feed, oil, and fertilizer production on a larger commercial scale (Nasekomo, n.d., [129]). Furthermore, Biobuutz, in Tanzania, has created the Kuku Bonge, which is a home bin for BSFL production on a household scale. InsectiPro in Kenya is producing BSFL and crickets from organic waste for feed and food purposes on a large scale outside of the city (InsectiPro, n.d., [130]).

Observed Co-Benefit and Limitations

This process of breeding insects on waste can be applied on a variety of scales, which makes insect farming an NBS unit that is suitable for circular city ambitions around the globe. The construction of an insect farm does not require advanced materials, as they can easily be farmed in containers or boxes and several species can be used. Therefore, it is a simple, inexpensive way to recover nutrients in low- and middle-income countries as well [121,124]. A further benefit of using insect farming is that it results in higher value products, for instance BSFL consists of 32–58% proteins and 15–39% lipids based on DW [131]. The products can also find instant application in other NBS such as nearby

aquaponic systems as feed for the fish. The only by-product emerging is a compost-like substrate that can be utilized as a soil amendment and fertilizer [124,125]. Furthermore, some insects have additional benefits, e.g., BSFL are not bioaccumulating pharmaceuticals and pesticides but instead accelerate their half-life time [125,132]. A reduction in viruses and *Salmonella* spp. Can be observed after fly larvae composting [133].

The drawbacks are the loss of nitrogen through degassing of ammonia [133], and for the BSFL, the high contents of unsaturated fatty acids [125]. While there was no strong bioaccumulation of Zn, Cr, Cu, and As in BSFL fed with pig manure, the bioaccumulation factor of Cd was significantly higher. The speciation of the metals differed between the pig manure and the residual biomass, and pathogens from the pig manure were reduced in the BSFL feces [134,135]. The main limiting factors of insect farming lie in the social and legal constraints. In Europe and North America, there is still resistance towards the use of insects in food. However, these perceptions are changing, and in a study in Flanders, Belgium, it was found that using insects in animal feed and the foods obtained from animals fed on insects are generally accepted [127]. Legal issues are more persistent; in most European countries, the products from insect farming are only allowed to be fed to fish and/or poultry. Furthermore, insects and are not allowed to be grown on domestic waste, only on verified industrial waste, for example, potato peels [136].

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

Insect farming (in combination with connecting units) provides the possibility to create high-value products from organic waste streams. This allows for a truly circular solution for organic waste as the organic waste becomes food again [108].

3.2.4. Soil Conservation and Phytomining

Working Principle

Around 340,000 contaminated sites and around 2.5 million potentially contaminated sites are located in the EU [137]. Ultramafic and brownfield sites provide unfavorable conditions for plant growth, primarily due to phytotoxic concentrations of metals, such as Ni, Cr, Co, Cu, Mn, Ni, Pb, and Zn, but also due to low nutrient availability, low organic matter content, poor soil structure, the absence of topsoil, erosion, surface instability, compaction, and often high acidity. Ultramafic substrates cover large areas in the Balkans [40]. This region is a potential target for agromining activities and also has the highest diversity in Ni hyperaccumulator plants in Europe and one of the highest globally together with Anatolia in Turkey [40,138–140].

Phytomining, or agromining, describes the technique of growing plants to 'mine' metals contained in such soils. This technique comprises a chain of processes covering the improvement of soil quality (phytoremediation) and the incineration of the biomass produced in order to obtain the metals from the ashes of the hyperaccumulator plants, which can be considered as a bio-ore [140]. Thus, metals are extracted by plants and recovered for further use. This non-destructive approach is applied to recover high value metals (e.g., Ni, Co, rare earth elements) from sub-economic (low-grade) ores [141]. Currently, over 1000 plant species with the ability to hyperaccumulate metals and metalloids are known, with most of them accumulating either Al, Ni, Mn, or Zn [142], but also Au [143].

Phytoremediation is a technology that uses tolerant plants to clean up soil, water, or air contaminated by pollutants [144]. It can be applied to restore contaminated or degraded soils while producing biomass for industrial use, such as energy, fiber, and phytomining. Phytoextraction uses accumulating or hyperaccumulating plants to improve the biological quality of a soil by accumulating trace metals and metalloids from metal-rich soils or substrates (technosols) and transporting them to the harvestable aboveground shoots [141,145].

In- and Output

Inputs, as described in Supplementary Materials A, include the growing substrate, plants, soil amendment for biostimulation purposes, and additional microbial strains for bioaugmentation. Outputs include improved soil, recovered metal bio-ores (metals-enriched biomass of hyperaccumulator plants), and energy from biomass combustion.

The typical edaphic properties of ultramafic soils can severely limit plant growth (e.g., nutrient deficiency, poor soil structure, low organic matter). In areas affected by mining activities, these edaphic properties can be especially severe. Organic residues (composts, manure, biosolids, mulch, wood chips, biochar) are commonly applied to such contaminated soils to improve the physical soil properties, water infiltration, and water holding capacity, as well as to provide essential micro- and macronutrients for plant growth, and to decrease bulk density.

Connected Units

Connected NBS units include treatment wetlands, which could provide reclaimed water and nutrients contained in fertigation water applied to phytomining plots. As mentioned above, compost is a common soil conditioner applied to support plant growth on the unfavorable conditions of metal-enriched soils or substrates.

As described in Supplementary Materials B, bioengineering techniques can be applied to support phytomining, in particular for land stabilization to mitigate the movement of contaminated soils, as well as to mitigate landslides on slopes or to stabilize riverbanks. Sustainable drainage systems can help to optimize soil water and nutrient retention.

Case Studies and Literature

In Albania, ultramafic soils account for 11% of the land area and are the richest in the number of endemic plant species, including several Ni-hyperaccumulating plants [40]. Phytomining field plots have been operating since 2005 in Pojske, Pogradec (ultramafic), Prenjas serpentine quarries, and Elbasan (contaminated by industrial activities). Consequently, cropping systems have been designed. The Ni hyperaccumulator *Odontarrhena chalcidica* (synonym *Alyssum murale*) cultivated on ultramafic plots in south-east Albania under organic and mineral fertilization reached a biomass production 9.96 t ha⁻¹ and Ni yields of 145 kg ha⁻¹. The Ni hyperaccumulator *O. chalcidica* has real potential to become a cash crop [40].

Zhang et al. [146] obtained ammonium nickel sulfate hexahydrate (ANSH) with 99% purity by applying the hyperaccumulator *Alyssum murale* on ultramafic soils in Greece and Albania, drying and incinerating the Ni-rich biomass and a sequence of treatments of the ashes. Koppolu et al. [147], Zhang et al. [146], and Houzelot et al. [148] obtained 5–13% of Ni in the ash from incinerating nickel (Ni) hyperaccumulator plants, significantly higher than the Ni-concentrations in common (primary) ores (3%) [149].

The LIFE-AGROMINE project (completed in 2021) has provided reference cases on ultramafic agricultural land, ultramafic quarries, and technosols based on industrial waste at sites in Greece, Albania, Spain, and Austria, demonstrating the full phytomining cycle including the recovery of Ni-rich products and bioenergy [40] (Figure 3).

The principle of phytomining can also be applied to municipal and industrial solid waste streams [150], if the metals are bio-available or made bio-available through appropriate additives [151]. Brownfield restoration at the city level can also be combined with phytomining. Additionally, metals can be leached from the waste body through the application of suitable microorganisms [152]. The further refining and recovery can then also be performed through conventional metallurgical means.

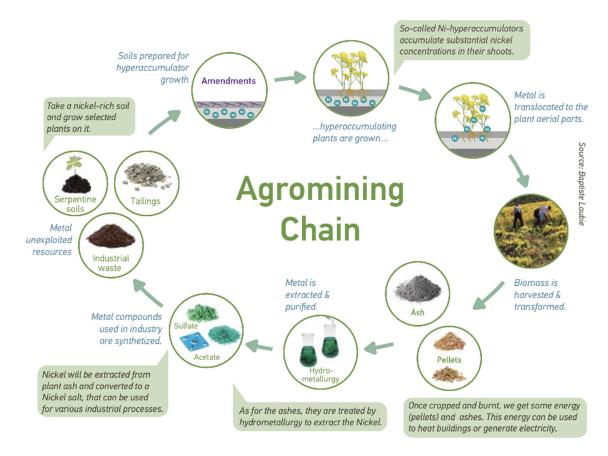


Figure 3. Agromining chain, source: Baptiste Laubie (2019), Layman's Agromine Report published in frame of Life AGROMINE project (https://www.alchemia-nova.net/website2018/wp-content/uploads/2019/08/laymans_agromine_EN-s.pdf, accessed on 28 August 2019).

Observed Co-Benefits and Limitations

Agroecological phytomining cropping systems permit the parallel cultivation of phytomining crops with conventional crops, which could provide additional benefits to farmers. Plant intercropping or co-cropping can enhance habitats and biodiversity, as well as stimulate the microbial communities and improve soil quality and functions. Incorporating N₂-fixing legumes into the cropping system can result in less dependence on fertilizers and can thereby enhance the resource efficiency, CO₂-footprint, and economic viability. Furthermore, hyperaccumulator plants are strongly resistant to pests and thus help to reduce the need for pesticide application. Farmers could apply this technique to metal-rich land to recover metals as a source of income. In particular, nickel agromining is considered an economically viable technique applied to ultramafic land, including ultramafic quarries, and technosols containing industrial waste. Plants that accumulate more than 2% Ni in aboveground biomass yield 200–400 kg Ni per ha, which has a greater value than all common agricultural crops [153]. In addition, renewable energy can be produced from the biomass (combustion or pyrolysis).

Breeding of improved strains with higher yields of the phytoextracted element as well as the improvement of methods to recover the agromined element(s) from plant biomass would further enhance the phytoextraction yield and financial feasibility [138,139].

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

Phytoremediation is an NBS which can be applied to any brownfields in cities to revitalize the valuable resource that healthy soils represent, thus counteracting "linear" land use, and enabling urban greening and the exploitation of its co-benefits for the living quality in urban areas, as well as for urban agriculture. Phytomining is likely less

widely applicable in cities because mining and smelting sites are typically located in rural areas; however, the cases of cities located in ultramafic areas where there is industrial or mining activities cannot be excluded [41,42]. Nevertheless, phytomining can contribute to supplying metals required for product value chains that ultimately reach cities and thus reduce imports of primary resources into the urban system. Consistently, by extracting metals from contaminated soils, phytomining enables the reuse of metals otherwise not utilized (wasted) and adversely impacting ecosystems.

3.3. Liquid and Solid Streams

3.3.1. Street Trees/Pocket Garden/Large Parks

Working Principles

Street trees, pocket gardens, and large urban parks are recognized as NBS by the following European projects URBANGREENUP—New Strategy for Re-Naturing Cities through Nature-Based Solutions (https://cordis.europa.eu/project/id/730426/fr, accessed on 30 September 2021), NATURE4CITIES—nature-based solutions for re-naturing cities: knowledge diffusion and decision support platform through new collaborative models (https://cordis.europa.eu/project/id/730468, accessed on 30 September 2021), UNALAB— Urban Nature Labs (https://cordis.europa.eu/project/id/730052, accessed on 30 September 2021) and in the scientific literature [154]. Street trees are defined as single or multiple trees planted, renewed, or maintained along roads, cycle paths, and footpaths. Suitable species must be selected for specific locations. Trees may be placed on one side of the road as single row trees and on both sides of the road to form a boulevard, if appropriate. In this case, the canopies of opposite trees can form an (almost) closed canopy. Pocket or garden parks are publicly accessible compact green spaces or small gardens (<0.5 ha) around and between buildings planted with ornamental trees, grass, and other plant species. Large urban parks are large green spaces (>0.5 ha) within a city with a variety of active and passive recreational facilities that meet the recreational and social needs of residents and visitors to the city. They are open to a wide range of audiences. Additionally, these plants offer a wide range of additional services and can enable resource recovery including liquid, solid, and gaseous streams (CO₂, etc.).

In- and Outputs

To function properly, street trees and plants established in pocket and large parks require a regular supply of nutrients and water. Nutrients can come from compost, organic or mineral fertilizers, or nutrient-rich irrigation water. Irrigation or fertigation water can be secondary treated municipal wastewater (e.g., output 2 of TW), rainwater, or other types of non-potable water. The amount of nutrients needed depends on the plant species and their nutrient requirements, as well as soil properties.

Output streams from street trees and parks are primarily cut branches, grass clippings, fallen leaves, seeds, and fruits that can be composted and returned to urban green spaces. While the above-ground woody biomass is not expected to accumulate pollutants [155,156], the leaf biomass may contain dust particles, heavy metals, and PAHs [157] therefore, the compost produced may not be suitable for food production and its quality would need to be analyzed prior to further use. Leaves, seeds, and fruits that fall on roads are removed by street cleaning and usually treated as mixed waste.

Case Studies

In many cities (e.g., Vienna, Ljubljana; see Table 1), the municipal composting facility or waste utility collects green waste from park maintenance and organic waste from public spaces and organic waste containers. The residents can also deliver garden waste on their own. The composting facility provides anaerobic digestion of organic matter and produces fresh compost, heat, and biogas. Heat and biogas converted to electricity are used to run the composting facility. Fresh compost is available to residents free of charge or for a reasonable price. Observed Co-Benefits and Limitations

Street trees and large- and pocket urban parks provide environmental, social, and economic benefits. They reduce heat island effects as they are cooler than the surrounding due to evapotranspiration and shading; however, the size of the park and the tree species used impact the temperature differences [158]. Parks and street trees reduce air pollution by adsorbing particulate matter onto tree and shrub surfaces, absorbing gases (O₃, NOx, SO₂, CO) and enabling bio- and photodegradation of organic pollutants such as PAH, thus reducing their further migration along the urban cycles and food chains [159,160]. Furthermore, they contribute to noise reduction; however, the density of vegetation, species, distance from the noise, ground surface features, as well as the subjective noise perception of residents impact the size of noise reduction [161,162]. Street trees and parks significantly reduce rainwater runoff, thus reducing urban flood risks and pressure on the sewage/stormwater collection and treatment systems [163]. By regulating the microclimate, urban water cycle, and water treatment, street trees and urban parks can also mitigate extreme weather events and their consequences [164].

Large urban parks and pocket gardens provide space for recreation and social gatherings and events, contributing to human physical and psychological health [165].

While biomass from urban parks and street trees is still a mainly unexploited resource, the studies show that it can significantly contribute to renewable energy needs [166,167]; moreover, selected parts of parks can be used for growing energy crops, which can reduce the maintenance costs and increase the renewable energy provision of the park [168].

Trees in pocket gardens, parks, and tree-lined streets can temporarily contribute to capture and store CO_2 emissions and thus reduce the city's carbon footprint. Trees in cities can sequestrate 0.61% of the annual traffic emissions, as shown on the example of Meran in Italy. This result also depends on the further biomass use/treatment [169]. Chen [170] estimates that the green infrastructure of 35 cities in China's major cities could, in summary, sequestrate 0.33% of the fossil fuel carbon emissions. The carbon storage calculations of urban trees can be very inaccurate and vary vastly depending on the management of the green spaces [171].

A comprehensive overview of regulating, provisioning, habitat, and cultural ecosystem services and disservices of street trees including the suggested management approached to maximize the benefits and reduce the limitations is provided by Sämuel et al. [172].

Street trees and urban parks have numerous co-benefits; however, if not designed and operated in terms of a circularity approach, they can also present certain disservices such as ecological (high water and nutrient demand), economic (leaf litter removal), social (undesirable insects and invasive plants), and public health (allergenic pollen) [173] issues; thus, urban planning needs to find a balance between providing as many co-benefits as possible while at the same time minimizing the disservices to acceptable levels [174].

Contribution of This NBS Unit to the Mitigation of Urban Circularity Challenges

The most recognizable contribution of street trees, large and pocket parks to urban circularity challenges is their mitigation of urban runoff and thus restoration and maintenance of the urban water cycle. Stormwater treatment and retention ponds, swales, and other measures of sustainable urban drainage can be integrated with street trees and urban parks, creating a multifunctional urban ecosystem. Especially in water-deficit areas, the irrigation of urban parks with reclaimed water, providing water and nutrients for plant growth, is a common practice [175,176].

Trees and parks are also applied to restore degraded building or district areas and recover their socio-economic function [177]. Additionally, street trees and urban parks are a low-cost source of lignocellulose-rich wastes that can be up-cycled to produce bio-composites [178], thus contributing to material recovery and reuse. Parts of urban parks can be arranged as community gardens, providing fruit, vegetables, and herbs, addressing the urban challenge of food production [179].

3.3.2. Gaseous Streams Working Principle

The ever-increasing growth in urban populations significantly intensifies anthropogenic effects on ecological systems, increases aerosols, particulate matter, and greenhouse gas emissions from heating, ventilation, and air conditioning (HVACs), traffic, and power generation, resulting in thermal hotspots and a continuous rise in CO_2 levels in cities.

 CO_2 is an essential ingredient for photosynthesis. Vegetation as well as (micro)algaebased technologies can turn CO_2 into biomass. Carbon capture mechanisms (CCM) of algae (including cyanobacteria, i.e., blue-green algae) supersedes the CO_2 utilization of higher plants. When connected with photobioreactor systems, algae-based CO_2 capture can go above ambient atmospheric levels. As such, higher CO_2 releases in urban settings such as industrial flue-gas, transportation exhaust, broilers, etc. can be mitigated.

Meanwhile, waste from pruning vegetation and algae biomass can also be harvested and processed to fertilizer (compost, biochar) or bioenergy [171]. Russo et al. [180] reported that trees in the streetscapes of the city of Bolzano, Italy, annually offset 0.08% of the amount of CO₂ emitted by the transportation sector. CO₂ sequestration by trees per m² of canopy cover were reported from 0.56 kg/year in bicycle lanes to 0.92 kg/year in streets [180].

Anderson and Gough [181] conducted a field study in Ontario, Canada, evaluating the impact of multiple green infrastructure applications on reducing ozone, nitrogen dioxide, and CO₂ concentrations across urban, suburban, and peri-urban morphologies. Data were collected from June to October over nine sites with mixed categories of five green infrastructure including green rooves, green walls, urban vegetation and forestry, urban agriculture systems, and tree-based intercropping systems. The results suggested that the application of green infrastructure across different urban, suburban, and peri-urban morphologies is beneficial in reducing CO₂, ozone, and nitrogen dioxide [181]. Though limited to one summer season, they detected an average reduction of 0.01, 0.11, and 23.4 ppm for ozone, nitrogen dioxide, and CO₂, respectively, across all sites and green infrastructure applications [181].

 CO_2 exhaust gas from industrial plants can be used to enhance plant growth. Increasing CO_2 concentrations in greenhouses is a commonly accepted technique to promote photosynthesis [182], causing more sugars and carbohydrates to be produced by the plant, resulting in shorter production times as well as increased yields and profits [182–186]. CO_2 supplementation can help to balance out CO_2 deficiencies that occur during the day in poorly ventilated greenhouses and thereby accelerate plant growth. This CO_2 could be added using waste exhaust gases, in particular, enrichment from exhaust gases compared to pure CO_2 [183], thus mitigating carbon emissions by capturing CO_2 in plant biomass [182]. Additionally, the use of purified exhaust gas from biogas combustion for CO_2 supplementation in greenhouses has been demonstrated [184]. This contributes to closing the carbon cycle by capturing and utilizing CO_2 for the production of food and industrial crops.

Reforestation and reducing deforestation and forest degradation (REDD) are eligible for carbon trading [187] (IPCC, 2007) and could thus represent an additional pathway to valorize CO_2 that is metabolized by urban or peri-urban forests. Nath et al. [188] highlight the high potential of timber bamboos for carbon farming and carbon trading due to the fast growth of bamboo and hence fast biomass accumulation. Timber bamboo captures 4.9–6 times the carbon that wood does [189].

In- and Outputs

Potential in- and output flows of vegetation for CO_2 capture correspond to those as outlined in Section 3.3 above. With respect to gaseous "resources", inputs include CO_2 and other air pollutants, introduced with the ambient air or exhaust gases directed to enclosed greenhouses. When photobioreactors are utilized for the cultivation of algae as described in Section 3.1.2, CO_2 -enriched air supply, NOx, SOx, and VOCs can be managed inputs. With

proper process control (pH, temperature, light, etc.), significant amounts of CO₂ capture can be achieved.

In addition to the outputs listed in Section 3.1.2, the outputs include O_2 and biomass, which can be utilized for biomass-to-bioenergy routes.

Connected Units

NBS units providing inputs to vegetated CO_2 capture include those producing soil amendments (e.g., composting), treatment wetlands and photobioreactors, which can provide treated wastewater for irrigation, as well as anaerobic treatment units, which can provide treated wastewater or digestate (nutrient source). NBS using residues of urban greening include composting. Photobioreactors include tubular or panel type designs as well as open pond designs. For output connections, units can vary depending on the final usage. When algal biomass is considered for liquid biofertilizer applications for city parks and other vegetation applications, no additional NBS units are required. However, anaerobic treatment units will be required for biogas/biomethane and subsequent compost applications.

Literature Case Studies

The famous 'vertical forest' (Bosco Verticale), an apartment building in Milan, features 20,000 plants, including 800 trees. It annually absorbs 40 tons of CO_2 and 1.5 tons of fine particulate matter each year and generates 90 tons of oxygen per year [190].

The discharge of CO₂-enriched exhaust gases into greenhouses for yield increase has been demonstrated, e.g., by Jaffrin et al. [184], who directed landfill biogas into a combustion boiler that directed the CO₂ inside a greenhouse after being purified. Thus, the waste gas was used both for heating the greenhouse and as a source of CO₂ supplementation to enhance plant growth [184].

The famous algae house (i.e., BIQ house) is a great example of CO₂ mitigation of urban buildings wherein broiler exhaust was in photobioreactors designed and installed as a facade (https://www.buildup.eu/en/practices/cases/biq-house-first-algae-powered-building-world, accessed on 30 September 2021). PhotoSyntheticaTM initiated by London-based Synthetic Landscapes Lab has several case studies demonstrating oxygen generating such as the Algae Curtain displayed in November 2018 at Dublin Castle during the week of Climate Innovation Summit in Dublin (https://www.photosynthetica.co.uk/cladding, accessed on 30 September 2021). Another notable case study is demonstrated by The Cloud Collective's Culture Urbaine Genève wherein photobioreactors were attached to the concrete siding of a viaduct highway to capture CO₂ from overpassing vehicles (https://inhabitat.com/overpass-algae-garden-turns-co2-emissions-into-combustible-biomass-in-switzerland/, accessed on 30 September 2021).

Benefits and Limitations

The plentiful co-benefits of urban greening are outlined in Section 3.3 above. The effectiveness depends significantly on design specifications such as a number of plants, growth conditions, and species. Velasco et al. [191] measured net CO_2 fluxes in subtropical and temperate urban areas, considering both vegetation and soil in combination. They found that urban greening reduced the total CO_2 flux by 1.4% in a neighborhood of Mexico City but added 4.4% extra CO_2 in a neighborhood in Singapore. They suggest that more complete assessments are needed to understand the lifecycle carbon reductions. Meanwhile, utilizing exhaust gases as a CO_2 source to measurably enhance crop production suggests that CO_2 is valorized that would otherwise be emitted to the atmosphere without further use.

As mentioned earlier, algae can mitigate significant amounts of CO_2 from urban environments, helping to decrease the overall carbon footprint of cities. Rather than costly carbon capture and sequestration (CCS) technologies, algae provide carbon capture and utilization (CCU), where additional value-added products such as biofertilizers and animal feed. These not only help municipalities to decrease their costs, but also provide additional CO_2 capture as emissions generated during the manufacturing of the replaced product is avoided.

Meanwhile, as fast-growing living organisms, algae-based NBS units require routine maintenance and equipment/processes in place to make use of generated biomass. Once a cycle is completed in set NBS units, seed cultures to initiate new batches must be available. Lastly, for building applications, appropriate measures must be taken to minimize or eliminate pumping noise of photobioreactors.

4. Supporting Units

4.1. Physical Separation Units

Starting from the conventional flush toilet (A, top left), Figure 4 categorizes the current existing toilet- and urinal types that can be used as supporting units in connection with NBS.

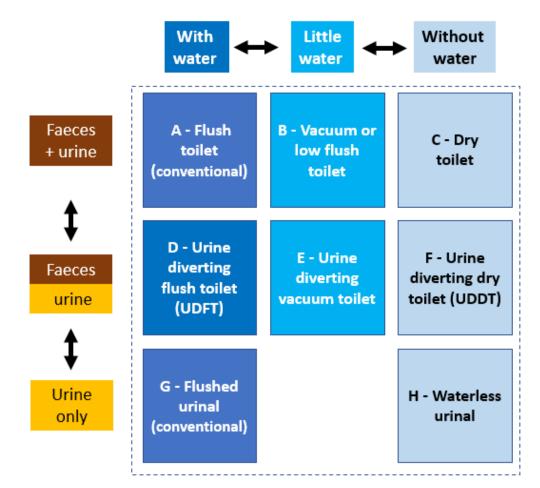


Figure 4. Toilets and urinals, categorized by water use and urine diversion. Grey: currently used toilet and urinal types.

4.1.1. Water-Saving/Water-Free Toilets without Urine Diversion Working Principle

The basic principles behind water-saving (Figure 4 Type B) and dry toilets (Figure 4 Type C) are the reduction or complete absence of water as a flushing medium and to produce pure(r) products.

In- and Outputs

The inputs are urine, feces, supporting materials (toilet paper, bulk material in dry toilets) and water (in low-flush toilets). The outputs of water-saving toilets are a mixture of

urine, fecal matter cleansing material, and water. The outputs of dry toilets are a mixture of urine, feces, cleansing materials, and bulk material. The fecal flow can be contaminated with other substances beyond the design purpose, such as vomit, pieces of plastics or hygiene articles, or unwanted materials (e.g., bedding for cat toilets). Blackwater and dry toilet material contain pathogens and microcontaminants, e.g., pharmaceuticals, hormones, and detergents.

Connected Units

The collected stream from water-saving or water-free toilets can be treated by anaerobic digestion (NBS 26) and, subsequently, nitrogen and phosphorus recovery (SU 3 and 4) or by hydrothermal carbonization (SU 6). In the case of dry toilets, the fecal matter plus urine can be transferred to a solid-state anaerobic digestion process (added NBS unit, derived from NBS 26), composting unit (NBS 23), or to a black soldier fly unit (added NBS unit). The dry fecal matter can also be dried and processed by pyrolysis (SU 6). The solid phase of anaerobic digestion (sludge), potentially after further drying and disinfection, or the processed dry toilet substrate can be used for soil improvement/slow-release fertilizer and conservation measures (NBS 33) or be subject to mechanical processing (added supporting unit) and, i.e., compost sieving (dry toilets).

Literature Case Studies

In Cressy (Geneva, Switzerland), the cooperative society 'Cooperative Equilibre' (CE) realized a three-story/thirteen-apartment building in 2011, which separates toilet waste from the water cycle by using dry toilets (Figure 4 Type C). Since 2011, CE has realized two more projects with a total of 103 apartments in Geneva following the idea of decentralized sanitation (including dry toilets) in an urban setting (https://www.cooperative-equilibre. ch/projets/cressy/, accessed on 30 September 2021, Kisser et al. [9]). Vacuum toilets followed by anaerobic treatment and nutrient recovery are applied in several so-called new sanitation projects in Germany, the Netherlands, Belgium, and Sweden [74,192].

4.1.2. Urine-Diverting Toilets

Working Principle

The common principle of all urine-diverting toilets (Figure 4, Types D–F) is the physical separation of the urine and feces flows within the toilet. This common principle is materialized in different ways, depending on cultural practices (e.g., sitting vs. squatting), the presence or absence of water as a flushing agent and by technical and design considerations [193]. The development is ongoing. Recently, a new and promising urine-diverting toilet has been developed and tested, based on computational fluid dynamics [194].

In- and Outputs

The inputs are urine, feces, supporting materials (toilet paper, bulk material in dry toilets) and water (in flush toilets). The outputs of urine-diverting dry toilets (Figure 4, Type F) are (a) separated urine and (b) fecal matter mixed with cleansing agents and bulk material. The outputs of flush toilets (Figure 4, Types D+E) are (a) separated urine with no or little water and (b) feces with toilet paper and water. The fecal flow can be contaminated with other substances beyond the design purpose, such as urine, vomit, pieces of plastic or hygiene articles, or unwanted materials (e.g., bedding for cat toilets). The urine can also become cross-contaminated with feces. Blackwater and dry toilet material contain pathogens and microcontaminants (e.g., pharmaceuticals and hormones) and, when contacted with fecal matter or excreted by people with a urinal infection, also pathogens.

Connected Units

The urine stream of urine-diverting toilets can be connected to a storage tank as a supporting unit for solid/liquid separation (SU 7) as described in Langergraber et al. [11]

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2021 and subsequently brought to a struvite precipitation unit for phosphorus recovery (SU 3). For nitrogen, ammonia stripping/adsorption (SU 4/SU 9) or the VUNA process (SU 4) are an option. The following connecting units have been identified for the processed urine: street trees and urban parks (fertigation, fertilization) (NBS 39, NBS 40, NBS 41), urban agriculture (NBS 49, NBS 50, NBS 51) and TW (NBS 21). For the solid phase, the connecting units are identical with those of water-saving/water-free toilets (see Section 4.1.1). The brownwater produced in flushed urine-diverting streams can be treated either anaerobically or aerobically, depending on the amount of water used for flushing. So far, this latter stream is discharged in the sewer for transport and treatment in a conventional central aerobic system. For full circularity, brownwater also needs to be treated locally and should include resource recovery and reuse.

Literature Case Studies

At the Forum Chriesbach office building in Duebendorf, Switzerland, a urine nutrient recovery system with UDFT for 220 people has been in operation since 2012 [195].

4.1.3. Water-Free Urinal

Working Principle

The common working principle of water-free urinals (Figure 4, type H) is the collection of urine without any addition of flush water. This common principle is materialized in different ways, depending on the design. The key characteristic of a water-free urinal is a device to allow free flow of the urine and at the same time prevent odor from the piping and storage to escape via the urinal.

In- and Outputs

The input to the system is urine. As so far only water-free urinals for men are available, the input is limited to male urine. The output is characterized by concentrated urine without any water, except for cleaning. Urine contains microcontaminants (e.g., pharmaceuticals and hormones) and, when contacted with fecal matter or excreted by people with a urinal infection, also pathogens.

Connected Units

While water-free urinals are widely used in Europe today, water-free urinals are applied and connected to a storage tank only in a few office buildings. There, the collected urine is subsequently brought to a struvite precipitation unit for phosphorus recovery (SU 3). Nitrogen is so far not recovered, but ammonia stripping/adsorption or the VUNA process could be an option (SU 4). In general, the connecting units are identical to those of water-saving/water-free toilets (see Section 4.1.1)

Literature Case Studies

Water-free urinals are installed in the office Rijnstraat, The Hague. Urine is, after storage, treated for struvite precipitation (https://www.nutrientplatform.org/en/success-stories/phosphorus-recovery-in-government-building//, accessed on 30 September 2021) and also at the EAWAG building Forum Chriesbach. Another case in the Netherlands with water-free urinals installed on a building scale is AFAS-LIFE. Urine is stored and transported to the wastewater treatment plant of Amsterdam, Waternet, for struvite precipitation (https://hollandcircularhotspot.nl/case/fosvaatje/, accessed on 30 September 2021). At the time of writing, it is not clear (yet) whether the recovery of P from AFAS-LIFE urine will be continued. Furthermore, the connecting units are potentially identical to those of the liquid phase of urine-diverting toilets (see Section 4.1.2).

4.1.4. Benefits and Limitations of Water-Saving and Water-Free Urinals and Toilets

One main benefit of water-saving (Figure 4, Types B+E), water-free (Figure 4, Types C+F) and urine-diverting toilets (Figure 4, Types D–F) is the reduction or complete avoid-

ance of flush water to enable energy-efficient recovery of the included resources. The quality of the outgoing flux depends largely on user behavior (e.g., proper use of the toilet).

Vacuum toilets and dry toilets are both dependent on electricity. Vacuum toilets rely on a working vacuum system. Dry toilets need a constant air flow to keep odors out of the building and regular handling for aeration and maturation of the compost, when directly connected with a composting unit. The main limitation is that the quality of the outgoing flux depends largely on user behavior (e.g., proper use of the toilet).

The main benefit of successful waterless urine-diverting toilets (Figure 4, Type F) or waterless urinals (Figure 4, Type H) is a much lower water consumption (on average, a person uses the toilet five times a day for urine production) and a concentrated, undiluted urine stock that can then be further processed by nitrogen and phosphorus recovery for fertilizer production. However, the quality of the outgoing flux depends largely on user behavior (e.g., proper use of the toilet) and to a minor extent on technical materialization (e.g., the existence of special toilet seats for kids). Urine contains microcontaminants (e.g., pharmaceuticals and hormones) and, when contacted with fecal matter or excreted by people with a urinal infection, also pathogens.

When combining water-free urinals and vacuum toilets in, for example, an office building, the vacuum collected blackwater is extra concentrated because it is not diluted by (half of) the urine and the associated flush water.

4.2. Bio-Physical Units

4.2.1. Bioengineering

Working Principle

Bioengineering uses vegetation within a "live" or "green" infrastructure system [196] and is applied as a building material for slope stabilization, e.g., for the mitigation of landslides; stabilization of riverbanks; to control sediment runoff, erosion, and flooding; and to enhance biodiversity [197]. Bioengineering techniques use live materials in combination with dead and inorganic materials (brush mattresses, geotextiles, fascines, wattle or wicker fences, hedge layers, branches, etc.).

In- and Outputs

Bioengineered structures may require irrigation, at least in the initial growth phase. This could be provided by rainwater or treated wastewater. Nutrients could be provided by secondary sources recovered by NBS such as water recovery from greywater treatment.

Bioengineered surfaces and slopes can retain rainwater and treat urban surface waters such as rivers [198], and thereby provide water fit for multiple re-uses. Bioengineering also provides a stable foundation for other NBS. Depending on the design, the vegetation can store large amounts of CO_2 as biomass [199], which could be used as a source of organic carbon and nutrients (as compost or biochar), or bioenergy.

Connected Units

Bioengineering can support soil conservation and phytomining (Section 3.1), as well as street trees, pocket gardens, and large parks (Section 3.2) by providing slope stabilization, preventing erosion and providing ecosystem services including nutrient capture from water sources. Bioengineering can also be a form of vegetated carbon capture system (Section 3.3).

Literature Case Studies

Bioengineering is applied to a larger extent in rural areas, but its benefits have been recognized also for cities. For example, for the construction of the Kartalpe metro station in Istanbul, Turkey, 0–25 m of topsoil was excavated and removed from a hill, resulting in serious erosion. A rehabilitation project applied bioengineering techniques to stabilize the slopes and re-vegetated them (ECOMED, 2017—https://ecomedbio.eu/case-studies-fluvial-coastal-slope, accessed on 30 September 2021).

Benefits and Limitations

Benefits include carbon storage as biomass [199], ecosystem services of urban greening, as well as nutrient removal from rivers in the case of vegetated riverbanks [198]. Compared to softwood riverbank stabilization (using brush mattresses, willow species), reed performed the highest nutrient retention and carbon sequestration in biomass in a study by Symmank et al. [200] in Germany. The recovered products should meet the demands with respect to quality (e.g., concentrations of pathogens and micropollutants, and requirements with respect to nutrient content).

4.3. Post- and Pre-Treatment Units

4.3.1. Disinfection (UV, Cavitation)

Working Principles

Disinfection is a process of adding chemical agent(s) into drinking water to inactivate pathogen microorganisms—parasites, bacteria, and viruses (EPA, 2021). Chlorine gas and chlorine dioxide are the most widely used disinfectants, while other forms of chlorine such as monochloramine (NH₂Cl) and dichloramine (NHCl₂) are used to a limited extent. The main principle of reaction is based on the formation of chlorine free radical. Besides chlorination, the most commonly used disinfection processes are UV radiation, solar disinfection, cavitation, multiple disinfectants (TiO₂/Ag⁺), and ozonation, amongst others [201]. Ozone disinfection, extensively used in Europe, is based on the fact that ozone is a strong oxidative agent ($E_{red} = 2.08$ V) and may react with substrates both via reactive O₃ species and hydroxyl radicals (\cdot OH) generated by the decomposition of ozone [202].

In- and Outputs

The amount/dose of applied chlorine depends on the type of chlorine disinfection: the added dose of active chlorine is between 2 and 5 mg L⁻¹, and after chlorination, the outlet should be between 0.2 and 0.5 mg L⁻¹ (PSATS, 2016—https://files.dep.state.pa.us/water/bsdw/operatorcertification/TrainingModules/ww05_disinfection_chlorination_wb.pdf, accessed on 30 September 2021). The advantage of chlorination over ozonation is the prolonged, residual effect of chlorine derivatives into the distribution system. Ozone is generated onsite because it is unstable and decomposes to elemental oxygen in a short time after generation [203]. Ozone may be added at several points throughout the treatment system, such as during pre-oxidation, intermediate oxidation, or final disinfection.

Connected Units

Various types of injection kits and pumps are used for water disinfection; a proper point of injection into the flow stream and thorough mixing is essential for full treatment (PSATS, 2016, https://files.dep.state.pa.us/water/bsdw/operatorcertification/TrainingModules/ww0 5_disinfection_chlorination_wb.pdf, accessed on 30 September 2021). The operational site for chlorination/ozonation has to be equipped by electric sources and adequate ventilation, but also requires a location relatively free of dust and dirt, protected from excessive sunlight or freezing. The disinfection area has easy access for maintenance and refilling and, if using a chemical tank, the tank has to be positioned as close as possible to the feeder.

Literature Case Studies

The occurrence and fate of carbonyl compounds as ozonation by-products at a full-scale drinking water treatment plant (DWTP) were studied for raw water and treated effluents (pre-ozonation, coagulation/flocculation, sand filtration, main ozonation, filtration through granular activated carbon and chlorination), on a monthly basis [204]. Pre-ozonation led to the formation of carbonyl compounds at concentrations of $67.3 \pm 43.3 \ \mu g \ L^{-1}$ (as a sum of 14 carbonyl compounds), whereas lower concentrations were determined after the main ozonation process, measured at $32.8 \pm 22.3 \ \mu g \ L^{-1}$. Moreover, the effective microbiological disinfection of drinking water may be also achieved with a lower concentration of ozone in a

shorter contact time compared to other disinfectants, such as chlorine, chlorine dioxide, and monochloramine [205].

Benefits and Limitations

The reaction between organic molecules and chlorine during water treatment results in potentially hazardous disinfection by-products (DBPs) [206]. Over 600 chemicals are classified as DBPs [207], among which the most hazardous compounds are known as trihalomethanes (THMs) [208]. According to the United States Environmental Protection Agency (US EPA), the maximum contaminant level of four chlorinated and/or brominated THMs in drinking water is regulated at 100 μ gL⁻¹ [203]. A range of low-molecular-weight carbonyl compounds (i.e., aldehydes, ketones, and carboxylic acids) are expected byproducts of this partial oxidation. Since ozone transformation products can either have a higher or lower tendency to generate DBPs than the starting material, contrasting effects on DBP formation are also unsurprising [201].

4.3.2. Activated Carbon

Working Principle

Adsorption is a chemical process used to remove a wide range of pollutants, both organic and inorganic, from liquid and gaseous flows. Most common adsorption systems use granular activated carbon (GAC) in column reactors because they are efficient and relatively cheap and simple to operate. GAC can be produced from different carbonaceous materials as wood, coke, coal, and agricultural residues [209]. The pressurized downflow columns are the most common solution for water treatment; in this case, GAC acts as filter, and more frequent backwashing is necessary [210].

In- and Outputs

Activated carbon adsorption can be adopted as the final step of plants treating municipal wastewater, greywater, blackwater, urine, or stormwater. It is mainly applied to remove organic micropollutants such as pharmaceuticals, personal care products, pesticides, and other industrial additives [211]. Treating secondary wastewater effluents with activated carbon results in a high-quality effluent that can be reused for many purposes. When the adsorption capacity of the GAC runs out, it is removed and sent to thermal regeneration.

Connected Units

GAC adsorption can be used to treat the effluent of TW (NBS 21), PBR (NBS 48), and wastewater aerobic treatment processes (NBS 26). It produces effluent that can be used to irrigate street tree/road vegetation, large urban parks, and pocket gardens (NBS 39, NBS 40, NBS 41).

Literature Case Studies

Bourgin et al. [212] investigated how WWTPs (upgraded by an advanced treatment for micropollutant abatement with (powdered) activated carbon treatment and/or ozonation could perform in reducing the discharge of micropollutants from WWTPs. The activated carbon filtration ensured a significant additional micropollutants abatement after ozonation due to sorption oxidation by-products (OBPs) such as bromate (BrO_3^-) and N-nitrosodimethylamine (NDMA), which allows to protect the ecosystem and drinking water resources in Switzerland.

The Pharmafilter pilot-scale installation (https://www.stowa.nl/publicaties/evaluationreport-pharmafilter, accessed on 30 September 2021) treats hospital wastewater for reuse and converts organic solid materials to energy. The core of the technical wastewater installation is the collection and treatment of wastewater to which other hospital waste flows have added and includes the use of single-use biodegradable solid products. The following processing steps take place in the installation: (i) shredding and separation, (ii) sieving over the grid, (iii) mixing/hydrolysis and digestion, (iv) membrane bioreactor, (v) high flux ozone installation, (vi) activated carbon, (vii) extraction and treatment of air, and (viii) monitoring and control. The ozone treatment may not remove all the micropollutants (pharmaceuticals, X-ray contrast fluids, etc.) and may convert an unknow number into metabolites, which may unfavorably affect the aqueous environment in which the treated wastewater is discharged. Activated carbon is therefore used as an extra stage to remove residues of pharmaceuticals, oxidation by-products, and hormone-disturbing substances that have passed through the ozone stage. Batelaan et al. [213] have reported that activated carbon filtration of ozone-treated effluent is acting as a good barrier to micropollutants. In another study, Duygan et al. [79] demonstrated that to reliably remove pharmaceuticals from treated urine, a post-treatment using adsorption to powdered activated carbon (PAC) was required. A risk assessment of the treated urine used as fertilizer on soil resulted in a risk quotient below 1 for the concentrations of trimethoprim, diclofenac, and sulfamethoxazole predicted in European countries and the USA. These results, and results from another study using granular activated carbon [214] have led to the production of a urine fertilizer (named Aurin) that is authorized for use on vegetables and flowers in Switzerland [215].

Benefits and Limitations

The main advantage of PAC/GAC adsorption is that it can simultaneously remove a large variety of inorganic and organic micropollutants, including disinfection by-products. It is also able to partially remove some pathogens. The main disadvantage is that the activated carbon runs out and must be replaced with a frequency that depends on the contamination degree of the fluid treated. Finally, it must be noted that a granular filtration section is necessary upstream GAC filters to remove total suspended solids.

4.3.3. Advanced Oxidation Processes (AOPs)

Working Principles

Advanced oxidation processes (AOPs) are frequently reported to be among the most suitable water treatment technologies to remove natural organic matter (NOM) and micropollutants (MPs) from wastewater [216]. The main principle of AOPs degradation is reaction of organic molecules (NOM and MPs) with hydroxyl radicals (\cdot OH), resulting in formation of smaller molecules (with a consequently smaller number of C atoms); \cdot OH radicals are defined as the strongest reactive species that can oxidize any compound present in the water matrix [202]. NOM, as a complex matrix of organic substances, is characterized by its variable molecular and physico-chemical properties caused by various solid-liquid interactions (bio-geologic formation and hydrologic cycle) [217]. MPs are usually found in aquatic medium at very low concentrations (ng L⁻¹– μ g L⁻¹) and known as xenobiotic compounds, such as pharmaceuticals, personal care products, steroid hormones, drugs of abuse, and pesticides, among others [218].

In- and Outputs

Despite the ability of a vast number of microorganisms to degrade a wide diversity of MPs (in conventional wastewater treatment plants), the residual concentration of these compounds in wastewater may be due to their low bioavailability in biological reactors [219]. Consequently, the secondary wastewater effluents of conventional activated sludge treatment still contain numerous MPs. In order to abate the presence of these compounds, advanced oxidation processes such as (i) UV/H₂O₂ [220], UV/chlorine [221] and/or ozone-based applications (O_3/H_2O_2 and O_3/UV) [222], and (ii) photo Fenton processes [223] and various electro-catalytic processes [224] are applied (or tested at pilot scale). Hence, the AOPs are found to fill the gap between the conventional physico-chemical and biological treatments and the limits set by environmental regulations (i.e., the degree of contamination of the treated wastewater determined by its end/use or site of discharge) [225].

Literature Case Studies

Developing countries, or even developed ones whose infrastructure is in decline, have dissimilar challenges concerning urban pollution prevention and control [226]. These range from providing basic access to safe drinking water and improving essential wastewater treatment. Outdated sewage systems that do not incorporate any wastewater treatment, as well as wastewater treatment infrastructure not designed to cope with an ever-growing number of MPs, are the main culprits in the deterioration of water quality. However, although AOP have been effectively tested (EU pilot scales) in the degradation of xenobiotic removal, particularly homogeneous photo-driven AOPs (e.g., UV/H₂O₂ and photo-Fenton) and heterogeneous photocatalytic processes (e.g., UV/TiO₂), have not yet found their application at full scale in urban wastewater treatment [227]. Ozone doses and contact times during advanced water treatment, which typically vary in the range of 1–5 mg L⁻¹ and 15–30 min, respectively, are usually insufficient to completely mineralize NOM [228].

Benefits and Limitations

One of the main advantages of AOPs is their capacity to simultaneously disinfect water. Hence, besides degrading organic pollutants (NOM and MPs), the mechanism for microbial inactivation used by AOP (i.e., the oxidative stress generated by ozonation) is also capable of reducing the microbial load of wastewater. Since ozonation may result in the formation of oxidation/disinfection by-products (e.g., N-nitrosodimethylamine (NDMA) a bromate), a polishing post-treatment step with a biologically active sand filter is recommended [229].

4.4. Resource Recovery Supporting Units

4.4.1. Phosphorus Precipitation

Working Principle

P-precipitation is generally established by the addition of multivalent metal ions such as calcium, magnesium, aluminum, and iron. Calcium and magnesium are generally applied for the recovery of P from concentrated streams, such as anaerobically treated blackwater or urine [83]. More information on struvite precipitation is reported in: https://run4life-project.eu/wp-content/uploads/2020/08/H2020-Run4Life-Factsheet-Technology-Struvite-Precipitation.pdf, accessed on 30 September 2021; Cunha et al. [230,231] show the possibility to produce calcium-phosphate granules in the anaerobic reactor (UASB) for treatment of blackwater, but the latter process is so far only applied at a laboratory scale.

In- and Outputs

High P input streams are needed for an efficient struvite recovery. Applicable urban streams are anaerobically treated, vacuum-collected blackwater with or without kitchen waste or separately collected urine, and rejection water from digested sewage sludge.

Connected Units

When P is precipitated from urine, water-free urinals or urine separation toilets followed by a storage unit are connected to the precipitation reactor, while anaerobic treatment is applied prior to the precipitation reactor when blackwater with or without kitchen waste is the phosphorus source. To ensure a sufficiently high concentration, the blackwater is collected with water-saving vacuum toilets (maximum one liter per flush). Prior to or after the recovery of phosphorus, nitrogen recovery/removal is needed. In Helsingborg, (H⁺) ammonia stripping/absorption is applied (see below), while in Sneek, nitrogen is removed via the OLAND process [81]. In Ghent, Nieuwe Dokkken, nitrogen is removed via conventional nitrification/denitrification, applying the COD from greywater as a carbon source plus a waste product from the nearby detergent industry (https://run4life-project.eu/demosites/, accessed on 30 September 2021).

Literature Case Studies

Case studies wherein struvite precipitation is applied in the urban environment are Waterschoon in Sneek, a housing estate of 250 houses with source-separated sanitation (https://www.stowa.nl/sites/default/files/assets/PUBLICATIES/Publicaties%20 2018/STOWA%202018-63%20NS%20Noorderhoek.pdf, accessed on 30 September 2021); Rijnstraat in The Hague, an office in which both urine and blackwater is separately collected (struvite is produced from urine) (https://www.nutrientplatform.org/succesverhalen/ rijnstraat8/, accessed on 30 September 2021); and H⁺ in Helsinborg (https://run4lifeproject.eu/demosites/, accessed on 30 September 2021) and Nieuwe Dokken in Ghent (https://run4life-project.eu/demosites/, accessed on 30 September 2021), both recently built housing estates with ca. 2000 inhabitants applying source separated sanitation with struvite precipitation from digested blackwater plus kitchen waste.

Benefits and Limitations

The benefits of the struvite precipitation process are the high recovery efficiency of phosphorus, a simple and stable process with a low energy input and a proven and well-known technology (https://run4life-project.eu/wp-content/uploads/2020/08/H2020-Run4Life-Factsheet-Technology-Struvite-Precipitation.pdf, accessed on 30 September 2021).

The product struvite is a slow-release phosphorous fertilizer, to be used in agriculture. However, the low N:P ratio does not meet the requirements of most crops; therefore, struvite is usually combined with nitrogen fertilizers (https://run4life-project.eu/wpcontent/uploads/2017/10/H2020-Run4Life-Factsheet-Product-Struvite.pdf, accessed on 30 September 2021; https://run4life-project.eu/wp-content/uploads/2017/10/H2020-Run4Life-Factsheet-Product-NPK-Pellet.pdf, accessed on 30 September 2021). Incinerated CaP granules, produced during anaerobic blackwater treatment, can directly replace phosphate rock in the fertilizer industry [230].

4.4.2. Ammonia Stripping/Absorption

Working Principle

In the ammonia stripping process, wastewater and air are brought into contact to transfer ammonia from the liquid to the gas phase. To ensure a high NH_3/NH_4^+ ratio, the pH of the wastewater is increased by adding a base. The water and gas flow in the opposite direction and the stripping tower generally contains packing material to enlarge the contact surface to maximize ammonia stripping.

To produce ammonium sulphate or ammonium nitrate, which can be used as fertilizer, the ammonia-rich air is scrubbed with nitric acid (HNO_3) or sulfuric acid (H_2SO_4). Further details can be found in: https://run4life-project.eu/wp-content/uploads/20 20/08/H2020-Run4Life-Factsheet-Technology-Ammonium-Stripping.pdf, accessed on 30 September 2021.

In- and Outputs

Urban inputs of a stripping unit are high-nitrogen-containing streams such as anaerobically treated, vacuum-collected blackwater or urine. Urine generally has a higher nitrogen concentration as compared to anaerobically treated, vacuum-collected blackwater.

Connected Units

The connected units are P-precipitation (struvite) and anaerobic treatment.

Literature Case Studies

So far, ammonia stripping for N recovery from urban streams applied in the city has only been executed in Helsingborg (https://run4life-project.eu/demosites/, accessed on 30 September 2021) for anaerobically treated, vacuum-collected blackwater. Urine generally has higher N concentrations as compared to vacuum-collected blackwater; however, so far, the ammonia stripping process for urine was only applied on a pilot scale [232].

Benefits and Limitations

The main benefit of the process is that a nitrogen fertilizer is being produced. For details on the product, see: https://run4life-project.eu/wp-content/uploads/2017/10/H2020-Run4Life-Factsheet-Product-Ammonium-Sulphate.pdf, accessed on 30 September 2021. The limitation of the stripping process is that it needs a stream with a high nitrogen concentration for an energy-efficient process performance.

4.4.3. Membranes

Working Principle

Membrane separation processes such as low-pressure microfiltration (MF) and ultrafiltration (UF), or high-pressure nanofiltration (NF) and reverse osmosis (RO), utilize a physical permeable barrier that enables to treat water while rejecting pollutants. During membrane filtration, the membrane allows the passage of certain constituents and retain other constituents found in the liquid. The membranes may be operated separately or in combination with other processes as a part of hybrid systems such as membrane bioreactors (MBRs), combining biological treatment (for example as activated sludge) with MF or UF [233,234]. The type of membrane and associated selective pore size influences the types of pollutants removed from the water. MF and UF are commonly used for solids, polymers, emulsions, colloids, and bacteria (for disinfection purposes) removal. In UF, viruses and proteins are also removed. NF and RO are used to reduce the effluent salinity or for the removal of organic and inorganic contaminants, including emerging contaminants and antimicrobial resistance control [216,235]. If the membrane is supplied with an aeration system, it may be used for nitrification, and a membrane-aerated biological reactor (MABR) can be used for nitrogen removal wherein both nitrification and denitrification (with external carbon dosage) can be achieved in one unit [236–238].

In- and Outputs

The incoming stream can be either treated or untreated urban wastewater, greywater, blackwater, or stormwater. The outcome streams are reclaimed water (effluent, also referred to permeate) and a concentrated stream with accumulated compounds not passing the membranes or, in the case of MBR, the solids.

Membranes can be employed as a polishing step for further removal of specific contaminants, and as such, support the TWs, PBRs, or anaerobic treatment units. By using an appropriate membrane type, membranes enable the recovery of water with a quality tailored to the needs of the reuse application, including potable water [239,240]. The reclaimed water produce can be used for irrigation or fertigation purposes (for example, in street trees, urban parks, and urban agriculture). Membranes may be also used for the harvesting of algal biomass in PBRs or solid/liquid separation (SU 7) in an anaerobic system without biomass retention [241].

Connected Units

Membranes are versatile and membrane filtration units can be incorporated with other units in multiple configurations. Membranes can act as a pre-treatment, post-treatment, separation, or up-concentration step.

Membrane units can be combined with other solid/liquid separation units (SU 7), including other type of membranes, which can provide a pre-treatment function. Membranes may also be followed by a disinfection unit, especially when membranes with more an open structure, such as MF, are used and/or when water disinfection is of particular importance. Other units such as AOP or activated carbon may be connected for post-treatment purposes to remove, for example, the remaining organic matter (COD), residual contaminants (e.g., persistent pharmaceuticals, chemicals, etc.), or salinity.

Case Studies and Literature

Among different membrane systems, MBRs are most commonly used for treatment of domestic wastewater, greywater, and/or a combination of domestic greywaters and effluents from blackwater treatment [242–245].

Observed Co-Benefits and Limitations

The main benefit of the membranes is the high and stable quality of the produced water, enabling water reuse, which contributes to closing the water cycle. Another benefit of the membranes is their small footprint and modularity, suitable for all scales, including single households [242,243]. The typical drawbacks are high energy requirements (which can be offset by the use of renewable energy sources or by gravity-driven systems), and the cost of membranes, membrane fouling, and generation of the concentrated stream containing the separated salts and other pollutants (which could be subsequently recovered with other potentially valuable materials).

4.4.4. Biochar/Hydrochar Production

Working Principle

Biochar and hydrochar are products of thermochemical processes of biomass conversion. Thermochemical processes include pyrolysis, torrefaction, gasification, or hydrothermal carbonization. For these processes, dry or wet organic carbon-rich material or C-rich biomass are required. The quality of their products (biochar or hydrochar production) depends on the type and process conditions of the thermochemical process. Processes that produce biochar include pyrolysis, torrefaction, and gasification; coproducts include water vapor, heat, condensable liquids (bio-oil, condensable tar (which goes to landfill), and syngas (combustible gases such as CO, CH_4 , and H_2 for energy production). In the case of hydrochar production, the reaction pressure (hydrothermal carbonization) is usually not controlled in the process and is autogenic with the saturation vapor pressure of water corresponding to the reaction temperature. At high temperatures, water with a high ionization constant can facilitate hydrolysis and cleavage of lignocellulosic biomass; water is responsible for the hydrolysis of organics, which can be further catalyzed by acids or bases.

In- and Outputs

Biochar is the char coproduct from the thermochemical processing of dry biomass. Biochar can be produced from different types of biomass residues, including crop plants (e.g., rice husk, wheat bran, etc.), tree cuttings, wood chips or dried fecal matter, such as composting toilet substrate [246], and as an intermediate product in bioethanol production (biowastes from the food processing industry).

The hydrothermal carbonization of C-rich biomass in the presence of water results in the production of a solid material that is referred to as hydrochar [247].

Literature Case Studies

Interest in biochar soil applications originated from the long-term fertility of terra preta anthropogenic soils in the Brazilian Amazon [248]. More recently, the recalcitrance of biochar carbon has attracted international attention as an inexpensive and effective way to sequester atmospheric carbon for centuries to millennia while simultaneously producing carbon-negative energy and improving soil quality [249]. Current research and demonstration cases focus on relationships between feedstocks, reaction conditions, biochar properties, soil and crop responses to biochar applications, and biochar economics [250].

Benefits and Limitations

In general, thermo-chemical processes are attractive and have certain advantages such as higher productivity, complete utilization of feedstocks, leading to multiple products, applicability to a wide range of feedstocks, independence of climatic conditions, and better control over the process relative to biological processes [251]. Biochar has been used primarily for soil remediation (e.g., [252]) and as an agent for carbon sequestration [253]. Both biochar and hydrochar have value-added industrial use, and both could increase carbon sequestration and nutrient recovery (because of the production of N-rich products). If used as soil physical and chemical improvers, biochar and hydrochar could both improve pesticide and nutrient management, increase soil carbon storage, enhance water infiltration and retention, encourage beneficial soil organisms, and prevent soil compaction [254,255]. On the other hand, they could be heavy metals sources, and if used in a high quantity as soil improvers, both biochar and hydrochar could increase albedo [256].

The advantage of hydrothermal carbonization processes is that it usually takes place at relatively low temperatures (150–350 °C, at about 2 MPa pressure) and wet feedstock can be directly used, including wet animal manure, sewage sludge, and algae [257]. It is a fast process that has a much shorter residence time than dry pyrolysis. However, there are contrasting data on the consistency of the eco-friendly nature of the process, even if tar is not produced and the ash content is reduced [258,259].

5. Discussion

The above-described NBS and supporting units present a broad set of technologies and solutions that can achieve a considerable level of resource recovery within urban environments, from water to nutrients to inorganic constituents and energy. Nevertheless, NBS still present some limitations that prevent further implementation of these solutions in many settings.

For one, defining NBS is still a work in progress, and different organizations have taken to develop their own definitions. The European Commission within its independent expert report defines NBS as: "inspired and supported by nature, which are cost-effective, simultaneously provide environmental, social and economic benefits and help build resilience (...) solutions [which] bring more, and more diverse, nature and natural features and processes into cities, landscapes and seascapes, through locally adapted, resourceefficient and systemic interventions" [260]. This somewhat differs from the definition developed within the COST Action Circular City framework in which this paper was developed. In it, NBS are described as: "concepts that bring nature into cities and those that are derived from nature (...) address societal challenges and enable resource recovery, climate mitigation and adaptation challenges, human well-being, ecosystem restoration and/or improved biodiversity status, within the urban ecosystems (...) definition we achieve resource recovery using organisms (e.g., microbes, algae, plants, insects, and worms) as the principal agents (...) [and] physical and chemical processes can be included for recovery of resources (...), as they may be needed for supporting and enhancing the performance of NBS" [3]. While the definition provided by the previous paper of the COST Action Circular City has to be considered within the context of achieving circularity in the specific environment of the city, this variation in definitions prevents a cohesive message from being conveyed to the wider community, which needs to finance, construct, and maintain these NBS in the first place. The lack of a uniform definition may also limit the development of a legal framework, which in turn also increases the bureaucratic problems associated with these types of interventions.

Another limitation for resource recovery using NBS is that, as observed in this paper, natural processes alone cannot achieve the necessary rates of recovery or provide the product in a retrievable form. In many cases, the resources used by the organisms of the NBS units are in normal conditions utilized for their natural growth (for example, plant growth); however, in most circumstances, NBS fail to provide the recovered nutrient or resource in pure form. Some notable exceptions do exist, such as anaerobic digestion, a process that results in the production of biogas and food production by urban farming. The COST Action Circular City takes this into consideration, and the inclusion of the need for supporting physical and chemical processes is particularly relevant for the group that developed this paper. The various supporting units described in this paper can

therefore provide alternatives that increase the rate of recovery of several nutrients for example, phosphorous-through processes such as struvite precipitation, with no biological input, but which result in a recoverable and easily applicable product. The supporting units in this paper have proven to be able to refine the output materials from some NBS units into higher-value products, and therefore provide greater efficiency and economic viability to a process which is still dominated by natural processes. However, a proper planning of the combination of NBS units and downstream supporting systems is required, in order to obtain good quality output materials. However, to achieve highquality end-products and high efficiencies, it is also necessary to have good source materials upstream. This can also be a limiting factor in achieving circular nature-based systems, as the modern configuration of many urban areas still approaches water resources as one stream, when, in reality, several higher-purity (and -quality) streams exist, such as stormwater, greywater, blackwater, etc. [261,262]. For many of the NBS described, the input wastewater streams must have specific compositions; otherwise, the systems do not function appropriately. At the same time, units such as urine-separating toilets require a separate drainage system to be implemented in order to separate streams, which requires investments in infrastructure [262]. This approach of separating waste/resource streams at the source has been a recent one which has not been implemented in many cities. Nevertheless, this challenge of avoiding contamination of the input streams is critical to obtain well-controlled systems for recovering and recirculating all components within the urban ecosystem and remains as the only effective solution to produce valuable resources without causing huge technological and financial challenges to minimize environmental and health risks.

Any NBS implementation plan in urban settings must also take into consideration the fact that, as previously described, there are limits to maintaining circular resource cycles within the cities. The greater population density of cities will always require some degree of resource importation from outside city boundaries. Solid material resources such as compost could be transported outside the city boundaries to provide nutrient resources to agricultural fields [22]. The output of these fields can later be transported inside the city. That way, the cycle does leave the city boundaries but maintains the necessary circularity. However, as long as the size of the market is sufficiently large to maintain specialized industries and to provide economically viable circular economy solutions, any kind of material can be recycled in the city-region boundaries [2]. It has been found that for any type of waste with low market value accumulated at a high density and high unit cost, transportation/treatment is more suited to local recycling [2], which means that urban wastewaters and organic waste are good sources to be used in urban ecosystems. Materials such as wastewater and solid waste, which are too voluminous and heavy and would require great energy expenditure to be feasibly exported back to rural areas, can in turn be used as sources of nutrients to close the existing urban cycles [20]. The internal urban resource cycles, which can be created by combining the several NBS and supporting units presented in this paper, will therefore be of great use to reduce environmental impacts. Given their characteristics, NBS and supporting units can fill this niche to maintain the resource use and recovery in urban environments, first by compensating for the flaws and limits of the dominant grey infrastructure, progressively replacing it entirely with natural systems, at different internal levels (household, district, and citywide) [3], integrating nature into cities in a sustainable way.

The contributions each NBS unit can provide towards solving the circularity challenges present in cities are varied, and in combination, they can solve all technical challenges that have been defined in previous actions of the COST Action Circular City. The water cycle can be restored and maintained by treatment wetlands, algal photobioreactors, and aerobic and anaerobic treatments, which treat wastewater and separate (micro)contaminants, materials, and nutrients. Material and nutrient cycles, in turn, can be closed as NBS units such as insect farming, phytomining, and composting upcycle them into products with added value, from protein-rich content for animal feed to natural biofertilizers to food products (thereby solving another of the technical challenges that had been found). In turn, most supporting units cannot solve the circularity challenges on their own, but it is their integration alongside the NBS units that can facilitate the closing of loops and connecting of the material and nutrient flows where there may be barriers. Here, complex (micro)contaminants such as pharmaceuticals and pathogens in wastewaters can be destroyed by disinfection processes such as UV, cavitation, or AOPs. Materials and nutrients particularly difficult to retrieve through purely biological processes can also be recovered by physical processing (e.g., membranes for wastewaters, ammonia striping), chemical processes (e.g., struvite precipitation), or thermal processing (biochar/hydrochar production). However, the efficient circularity of resources can be achieved only if cities implement urban farming or resources are used outside of city limits. All of these potential solutions fulfil the objectives of achieving resource efficiency and circularity in cities set out by several institutions such as the European Union in their Green Deal [263], their Action Plan for Zero Pollution for Air, Water and Soil [264], and ultimately, their Circular Economy Action Plan [265]. Moreover, the 2030 Agenda for Sustainable Development set out by the United Nations can also be fulfilled with the closing of loops and waste reduction provided by the combination of NBS and Supporting Units, including the goals of Clean Water and Sanitation (SDG 6), Sustainable Cities and Communities (SDG 11), Responsible Production and Consumption (SDG 12), and Climate Action (SDG 13), among others [266].

All the NBS and supporting units presented in this paper are proven systems (TRL > 5) with examples of application in urban areas. The great variety of solutions makes it feasible that connecting NBS and supporting units can form a circular network in which all resources present in solid and liquid waste can be reused, recovered, and recycled. The analysis performed by Diaz-Elsayed et al. [267] suggests that the life cycle impacts of resource recovery are generally decreased the higher the number of people served by them is, which means that by interlinking these units into larger-scale systems, the overall environmental impact can be reduced even further. This fulfils the objective to reach a natural resource system within and/or associated to the city, achieving the objective of the circular city. Nevertheless, the work of research and innovation in this field has continued, and in the coming years, new innovations and approaches are expected to continue to appear. By continually increasing reuse/recovery/recycling yields, the movement towards circular cities can continue to progress and provide natural solutions which improve urban ecosystems and provide human well-being and resilience towards climate change.

6. Conclusions

This paper attempted to provide an updated snapshot of the current characteristics and capabilities of nature-based solutions and supporting units based on physical and chemical processes. The data obtained enabled us to understand that, depending on the input and output of different systems, it is possible to create a network of technologies using mostly natural processes that can recover resources and reapply them in environmentally friendly ways. The limitations of many of these NBS can be overcome by integrating them into more complex but extensive systems with lower life cycle impacts. This enables the development of solutions that are not only good from an environmental standpoint, but which are also economically and socially beneficial towards communities living in cities, improving their well-being and resilience towards the coming challenges of climate change.

The great variety of possible combinations between NBS and supporting units is a testament to their versatility, and their application is recommended in further projects and pilot tests throughout Europe and beyond. To that end, we recommend that future studies in the field of NBS focus on the study of circular networks to achieve new circular resource management units in cities, using not only the above-described proven systems (TRL > 5) but also other systems with lower TRL but with associated potential.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/ 10.3390/w13223153/s1, Table S1: Supplementary Materials A Description of NBS Units, Table S2: Supplementary Materials B NBS supporting units. **Author Contributions:** All authors contributed equally to conceptualization, writing and reviewing. All authors have read and agreed to the published version of the manuscript.

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