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Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv



Urban nitrogen budgets: Evaluating and comparing the path of nitrogen through cities for improved management

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HIGHLIGHTS

• Household, wastewater, denitrification central for urban core

- Urban agriculture, export of goods, losses to environment central for surrounding
- Increasing Nr recycling and nitrogen use efficiency through Nr recovery from human excreta and waste is possible and needed

G R A P H I C A L A B S T R A C T



ARTICLE INFO

Guest Editor: Lex Bouwman

Keywords: Nitrogen Urban Environment Circularity Air pollution Water pollution

ABSTRACT

Reactive nitrogen (Nr) released to the environment is a cause of multiple environmental threats. While Nr flows are often only analyzed in an agricultural context, consumption and emission takes place in the urban environment, and opportunities for Nr recycling and effective policy implementation for mitigation often appear in cities. Since little information is available on the bigger picture of Nr flows through the urban environment, these opportunities often remain unexploited. Here we developed a framework to model Nr pathways through urban and surrounding areas, which we applied to four test areas (Beijing and Shijiazhuang (China), Vienna (Austria), and Zielona Góra (Poland)). Using indicators such as recycling rates and Nr surplus, we estimated environmental risks and recycling potentials based on Nr flows and their entry and exit points. Our findings show marked differences between the core and surrounding areas of each city, with the former being a site of Nr consumption with largest flows associated with households, and the latter a site of (agricultural) production with largest flows associated with industry (fertilizers) and urban plants. As a result, Nr transgresses the core areas in a rather linear

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https://doi.org/10.1016/j.scitotenv.2023.166827

Received 26 May 2023; Received in revised form 30 August 2023; Accepted 2 September 2023 Available online 6 September 2023

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manner with only 0–5 % being re-used, with inputs from Nr contained in food and fuels and outputs most commonly as non-reactive N_2 emissions to the atmosphere from wastewater treatment and combustion processes. While the peri-urban areas show a higher Nr recycling rate (6–14 %), Nr accumulation and emissions from cultivated land pose significant environmental challenges, indicating the need for mitigation measures. We found potential to increase nitrogen use efficiency through improved Nr management on cultivated areas and to increase Nr recycling using urine and sewage sludge as synthetic fertilizer substitutes. Hence our framework for urban nitrogen budgets not only allows for consistent budgeting but helps identify common patterns, potentially harmful flows and Nr recycling potential.

1. Introduction

Reactive nitrogen (Nr), as key plant nutrient, plays a critical role in sustaining human life on earth (Erisman et al., 2018). Naturally fixed nitrogen is insufficient to meet the growing demand for agricultural production worldwide, thereby necessitating the large-scale application of mineral fertilizers produced through the Haber-Bosch process, which substantially alters the natural N cycle (Smil, 1999; Fowler et al., 2013). Another part of this alteration of the natural cycle concerns Nr losses to the environment added from combustion processes and livestock husbandry. Altogether anthropogenic activities are estimated to account for around half of the Nr input to terrestrial and marine ecosystems (Fowler et al., 2013).

While facilitating the provision of food, this increase of Nr input is associated with a range of adverse effects (de Vries, 2021). Excess nitrogen use on soils can lead to eutrophication of water bodies or soil acidification but also has global effects due to the formation of nitrous oxides (N₂O), a greenhouse gas with a global warming potential nearly 300 times stronger than that of CO₂ (Fowler et al., 2013; Reis et al., 2016; Erisman et al., 2018). Increased atmospheric Nr deposition affects biodiversity as well as the health of marine and terrestrial ecosystems through additional contributions to eutrophication and acidification (Smith, 2003; Bobbink et al., 2010). These perturbations to the N cycle also affect human health (Townsend et al., 2003), with exposure to nitrogen oxides (NO_X) and particulate matter (PM) linked to combustion processes contributing to respiratory and cardio-vascular diseases, while elevated levels of nitrate (NO₃) in groundwater can lead to methemoglobinemia in infants and gastric and oesophageal cancers (Anenberg et al., 2022; WHO, 2011).

While most literature focusses on the global or national N cycle, the alteration of the natural N cycle and its consequences are particularly important in urban areas, due to their distinct landscapes, production patterns and high population density leading to concentrated Nr consumption (with 79 % of food produced for cities – FAO, 2023). High population densities also mean that a large population is exposed to the consequences of intensive Nr consumption and environmental losses from the N cycle. In 2019 it was estimated that globally two-thirds of the 1.85 million new asthma cases attributed to nitrogen dioxide (NO₂) occurred in urban areas (Anenberg et al., 2022).

Intensified Nr consumption leads to intensified losses to water and the atmosphere, potentially threatening not only human health but also nearby ecosystems. Faerge et al. (2001) found that, in Bangkok, a significant portion of Nr losses throughout the urban environment was released into the aquatic system. Gu et al. (2012) demonstrated that the quality of surface and groundwater within the greater Shanghai area exceeded the highest thresholds for Nr concentration of the Chinese government standard for water quality as emissions from the urban environment to the hydrosphere have increased. This means that the water is heavily polluted and not safe for human consumption. Pet excretion, particularly in municipal green areas, is a specific form of enriching urban soils with N. De Frenne et al. (2022) found an average deposit of 11 kg N ha⁻¹ of dog excreta on peri-urban forests and nature reserves around Ghent (Belgium), compared to 5-25 kg of nitrogen from agricultural, industrial and traffic sources. This nitrogen can form a soil deposit, infiltrate deep into the soil profile as ammonium NH₄ and be released to the atmosphere as N_2O or NH_3 (Petrovic, 1990; Sutton et al., 2000). The loss of soil nitrogen occurs under denitrification conditions (release to the atmosphere as N_2), resulting from a shortage of oxygen in the soil. Such conditions can arise from excessive soil compaction (a situation typical in cities), as well as intensive fertigation with sewage or cyclic flooding of the soil with water (Toor et al., 2020). However, urban environments also offer a potential for Nr recycling. Svirejeva-Hopkins et al. (2011) showed that Nr in wastewater being lost due to denitrification during treatment represents an annual resource of around 32 million Euros when used as fertilizer. This loss could be prevented by e. g. directly making use of urine, sewage sludge streams, and food-waste, while also reducing water consumption (Sutton et al., 2013).

The impact of urban areas on the environment and human health as well as their potential for Nr recycling are expected to become increasingly significant, due to the projected global urban population increase, with over 68 % of people living in cities by 2050 (UN, 2018). However, despite the increasing importance of understanding Nr in the urban environment, available research on this topic is limited and the lack of a harmonized methodology hampers comparability between studies (Winiwarter et al., 2020). Hence, we developed such a methodology for calculating and comparing N budgets for urban areas, aimed to be applicable under very different circumstances. As test areas, we selected four cities, two situated in China and two in Europe and used the results to draw conclusions about the Nr cycle in different urban areas, to compare specific output parameters among the cities, and to evaluate potential threats to the environment and human health. Additionally, our method enabled us to evaluate potentials to increase Nr recycling. The benefits and limitations of such comparisons include the identification of specific flow patterns and the opportunity to learn from other examples.

2. Methodology

2.1. The framework

In a first step, a stock/flow model was developed to quantify Nr flows between so-called 'pools', where Nr is converted or accumulated (stock) (see Fig. 1). This model was based on the guidance document for the calculation of National Nitrogen Budgets (NNBs) developed by the Expert Panel on Nitrogen Budgets (EPNB) (UNECE, 2013). The definition and names of some pools had to be modified to fit the purpose of representing an urban or peri-urban system and indicating a potential deviation from the initial NNB pool.¹ The aim of keeping a similar structure to that recommended for NNBs is to enable the downscaling from and comparison with country budgets and national air pollutant and greenhouse gas inventories.

The final urban model framework contains ten pools and 110 flows between the pools and beyond the system boundaries (supplementary material S1). The flows represent Nr encompassed in goods or Nr losses such as NO₃ leaching or NH₃ and N₂O volatilization as well as NO_X emissions. Conversion of Nr into non-reactive nitrogen, N₂, e.g. from

¹ A more detailed description of each pool's reference pool in the NNB can be found in section S1 of the supplementary material.



Fig. 1. Scheme of urban and peri-urban stock and flow framework as implemented in STAN. The grey line represents the system's boundary. Dark bars at the bottom of pools represent stocks. Dashed arrows represent N2 (non-reactive nitrogen) flows from the system to the system's environment. Arrows from and to the Import/Export pool symbolize exchange with the system's environment.

wastewater treatment and combustion, is considered an Nr sink and not further dealt with. The framework was implemented in the Material/ Substance Flow Analysis freeware 'STAN', which not only enables the graphic depiction of stocks and flows but also enables the consideration of uncertainties and missing data using mathematical statistical methods such as error propagation (Cencic and Rechberger, 2008).

2.2. Individual pools and connections

2.2.1. Import/export

The import/export pool deals with flows beyond the system's boundaries. All Nr in imported and exported goods as well as all Nr pollution that is not deposited locally passes through it.

2.2.2. Combustion

This pool represents all combustion processes within the respective study area including energy

production, transport, heating, and others. Inputs to the combustion pool come from the industry pool as fuel for industrial processes, energy production and transport, the household pool as fuel for heating, the waste pool as waste for waste incineration and the urban plants pool as fuel for agricultural machinery. Only the actual conversion of fuels through combustion processes takes place in the combustion pool. Outflows from the combustion pool are directed to the waste pool as combustion residues and to air as Nr emissions such as NO_X , or they are considered sinks when N_2 is formed from exhaust cleaning.

2.2.3. Industry

The industry pool encompasses all industrial processes (chemical, food processing, energy production, etc.) taking place in the study area. Input flows come from the import/export pool, the urban plants and urbans animals pool, depending on the type and extent of agricultural production in the area, and from the waste pool in the form of recycled waste. Outputs are directed to the combustion pool as fuel for combustion processes, to households as food or other products, to urban crops as synthetic fertilizers, to urban livestock and pets as feed and in terms of import/export in the form of exported products, and to wastewater and air (only as direct emissions, not emissions from combustion processes).

2.2.4. Households

The household pool represents local consumption with inputs such as food coming from agricultural land, horticulture or from the production

of other goods such as furniture, fuels and clothes in the industry pool or import from beyond the system boundaries. There is also an inflow from the air pool, representing nitrogen deposition on built-up urban or periurban areas. The main outputs are directed to waste and wastewater as well as combustion. There are also outflows to agricultural land, in the form of urine directly applied to soil surfaces, to urban livestock, in the form of food residues used as feed, and to urban greens, in the form of organic waste composted in private gardens. This pool is the only one with pre-existing stocks, rather than annual stock changes, representing Nr in furniture, food and other products that are stored in households.

2.2.5. Waste

The waste pool includes all processes linked to waste treatment, such as composting, incineration, recycling, landfill etc. Inflows come as waste from the household pool, the urban plant, urban animals (mainly pet excreta) and industry pools or in the form of sewage sludge from the wastewater pool and ashes from the combustion pool. Outflows are recycled waste directed to the industry pool, waste for incineration directed to the combustion pool, compost directed to urban plants, untreated waste that is exported outside the city boundary and emissions to air, water and wastewater depending on different waste treatment types. This pool has stocks, representing Nr remaining in landfill.

2.2.6. Wastewater

The wastewater pool represents all flows connected to wastewater (industrial and domestic) in the respective area. Inflows come from the household pool and the waste and industry pools. Outflows are directed to waste as sewage sludge, to water and air as emissions but also outside the system boundaries, as non-reactive nitrogen (N_2) is formed during denitrification in wastewater treatment.

2.2.7. Urban animals

The urban animals pool encompasses two sub-pools: pets and urban livestock. The main inputs to the pet pool come from the industry and import/export pools in the form of feed. Main outflows are directed to the waste pool and the urban green sub-pool as pet excreta. The main input to the urban livestock pool is feed from the industry and import/export pools as well as from the 'agricultural land' sub-pool and the household pool in the form of food residues used as feed. Main outputs are livestock products directed to the household, industry and import/export pools as well as manure Nr directed to agricultural land sub-pool, slaughter waste and manure Nr directed to the waste pool and emissions from manure management directed to the air pool. Both sub-pools have stocks, representing the Nr retained by livestock and pets.

2.2.8. Urban plants

The urban plants pool is divided into three sub-pools: (i) urban greens, encompassing parks, gardens, forests etc., (ii) horticulture, defined as smaller-scale production in gardens or greenhouses as opposed to large-scale crop production on fields (encompassing only flowers in the Chinese test areas, orchards and flowers in the Polish test area and vegetables and flowers in the Austrian test area) and (iii) agricultural land, encompassing crop- and grassland. The main inflow to all three pools is mineral fertilizer from the industry or the import/ export pool. Other inflows to the agricultural land pool are sewage sludge from wastewater processing, manure Nr from urban livestock, compost from waste processing and human excreta from the household pool used for fertilization, Nr in water used for irrigation, Nr deposition and biological nitrogen fixation (BNF). The latter two are both encompassed in the Nr flow from air to agricultural land. Main outflows from the agricultural land pool include agricultural goods directed to households, import/export, urban livestock, and industry as well as losses to the environment such as NO3 leaching and run-off into the water pool and N2O and NH3 volatilization to the air pool. On urban greens, additional input includes compost from households and pet excreta, while the only outputs are losses to air and water as well as

green waste. From the horticulture pool, there is an additional output to urban greens, representing flowers planted in public parks. All three subpools are assigned stocks, representing Nr remaining in the soil.

2.2.9. Water

The water pool represents all water bodies relevant for the respective study area (e.g., groundwater, rivers, lakes). Main inflows are from urban plants and wastewater processing, while major outflows are directed to the urban plant pools for irrigation.

This pool contains stocks, as Nr can accumulate in standing water such as lakes and groundwater.

2.2.10. Air

The air pool represents all emissions to the atmosphere. Inputs come from urban plants, urban livestock, wastewater processing, waste processing, combustion, and industrial processes. Outputs are in the form of Nr deposition, directed to urban plants and households (representing built-up areas) or transboundary emissions directed to the import/ export pool. This pool also includes BNF as flow from air to urban plants although the N fixed from air is non-reactive. Alternatively, it could also be accounted for in the stock calculation or urban plants.

2.3. System boundaries

After setting up this model template, all flow parameters were populated with regional-specific information of four test areas: Vienna (Austria), Zielona Góra (Poland), Beijing (China) and Shijiazhuang (China). The geographical delineations of the areas were intended to include all areas of high population density, while at the same time enabling access to the required data (Fig. 2). To ensure the latter, alignment of the area boundaries with administrative boundaries proved to be highly beneficial. The use of administrative boundaries may not in all cases fully capture the specificity of an urban situation with regard to Nr budgets, possibly hampering comparability between cities. To capture such effects, we define for each city the "core" and the "surrounding" area, assuming that at least one of them would adequately represent cities. The 'core' area covers built-up land (as much as this can be reflected by the need to adhere to the administrative boundaries). The 'peri-urban' area is defined by its very strong exchange with the core, e. g., by providing the urban workforce, being a primary source of fresh produce or similar – all characterized by fast transport infrastructure.

For Vienna, this split was carried out by taking advantage of the territorial classification used in the European Union, with the NUTS 2 areas for Vienna taken as 'core', and the two NUTS areas 'Vienna Surrounding – North' and 'Vienna Surrounding – South' as the surrounding peri-urban area. For Shijiazhuang, the area within the second ring road of Shijiazhuang was define as city center, with the area outside the second ring road being defined as peri-urban. The split between urban and peri-urban areas for Beijing was made according to distinctions used by city planners. Six central districts of Beijing were defined as core area and the other areas as surrounding. Zielona Góra was divided into the older town (core area) and the 'New District' (surrounding area). The



Fig. 2. Delineation of the four test areas and the differentiation between their core (dark green) and surrounding (light green) areas.

New District, a former rural community with only 7 % built-up area in total land area, was incorporated into the city of Zielona Góra in 2015.

2.4. Data collection and calculations

2015 was chosen as base year for data collection. Data collection for the calculation of the different flows was making use of the respective statistical national/urban data available for each test area, often using proxies such as population or livestock distribution to break down certain flows to the necessary level of detail.

For the urban and peri-urban area of Vienna, most information on combustion, livestock, agricultural and horticultural land was taken from Statistik Austria (n.d.). For the Vienna core area, more detailed data were often taken from information made publicly available by the city authorities or the departments responsible for waste and wastewater (MA48, 2015; ebswien, n.d.). N excretion and Nr volatilization rates were taken from the GAINS model (Amann et al., 2011). A full description of all calculations for the Vienna urban and peri-urban areas is available in Section S2 of the supplementary material.

For the Chinese test areas, the "coupled human and natural systems" (CHANS) and "NUtrient flows in Food chains, Environment and Resources use" (NUFER) models were used, which provide, among others, parameters such as Nr deposition rate, BNF rate, the ratio of crop yield to feed/food, Nr excretion rate, Nr volatilization rate, air emission rate and

2.5. Indicators

Several indicators were used to evaluate the Nr budgets for the test areas and to facilitate the comparison between the test areas. To evaluate the efficiency and circularity of the system and investigate any potential for improvement, we defined a recycling rate as the ratio of recycled Nr to total Nr input. This can also be described as the inverse of the Nr loss rate, as having a larger recycling rate implies lower Nr losses, which is desirable. Recycled Nr in all test areas was defined as output of one pool that is used as a substitute of an input to another pool. Flows linked to recycling are excreta and sewage sludge (from wastewater) recycled for fertilizing urban plants, waste re-cycled or used as fertilizer and food residues used as animal feed.

To evaluate potential threats to the environment, Nitrogen Use Efficiency (NUE) and Nitrogen surplus (N_{sur}) based on the "soil surface N budgets" following Oenema et al. (2003) were used. These indicators were calculated separately for agricultural, horticultural, and urban green land. N_{sur} was calculated by subtracting the sum of all Nr outputs (N in harvest directed to industry, households, export or waste) from the sum of all Nr inputs (fertilizers from industry and import, wastewater, waste, biological N fixation and Nr deposition) and dividing the resulting values by the respective cultivated area (1). NUE was calculated by dividing the sum of all N in harvested crops by the sum of all inputs (2).

$N_{sur} =$	$\frac{Mineral\ Fertilizer + BNF + N\ Deposition + N\ Excreta + Waste/Wastewater - N\ Ha}{Cultivated\ Area}$	(1)
NUE =	= <u>N Harvest</u> Mineral Fertilizer + BNF + N Deposition + Manure N + Waste/Wastewater	(2)

Nr loss ratio of wastewater (Gu et al., 2015; Ma et al., 2012). Additionally, the Chinese statistical yearbooks (NBSC, 2016) provided information on activity data such as Nr fertilizer application, sown area, urban green area, horticulture area, yield, livestock number, pet number animal weight, population, fossil fuel, industrial product, waste and wastewater.

For the Polish test area, the data for calculating were taken from the databases of the Provincial Statistical Office in Zielona Góra, obtained directly from the local bodies responsible for municipal management. Data on wastewater and waste was obtained from the Spatial Planning and Environmental Protection Departments of Zielona Góra City Hall, the Department of Public Utilities of Zielona Góra City and the citymanaged waste processing plants: sewage treatment plant and collection of municipal waste. In terms of environmental impacts and the characteristics of surface and ground waters, measurement data from the Provincial Inspectorate for Environmental Protection were used. Calculations related to agriculture, including animal husbandry, were made using indicators developed by IUNG (Institute of Soil Science and Plant Cultivation) in Puławy and IMUZ (Institute for Land Reclamation and Grassland Farming) in Falenty, based on the Polish specificity of agricultural production (Bilski, 2008; Czyżyk et al., 2011). Data on pets in the urban area were obtained from the Chief Veterinary of Poland's annual report on visits to animal shelters and general information about dog and cat populations in the EU, shown by country (Sas, 2019).

Other, more general, resources were the "Guidance Document on National Nitrogen Budgets" (Winiwarter & EPNB, 2016) for N content of different food items as well as calculations for pet Nr excretion and volatilization, and the IPCC guidelines (2019) for wastewater calculations. Information on average food intake per capita for each test area was taken from FAOSTAT (2021).

N Harvest ... N in harvest as crops, grass, flowers etc., including fractions going to industry/livestock/export/waste

Mineral Fertilizer ... Mineral Nr fertilizer produced locally or imported that is applied to soils (volatilization was not subtracted)

BNF ... Biological nitrogen fixation

N Deposition ... Nr deposited on soils

N excreta ... Nr excreta from livestock, pets, and humans applied to soils

Waste/Wastewater ... Straw residues, compost and sewage sludge applied to soils

Cultivated Area ... Agricultural, horticultural, or urban green area

3. Results

Calculating the individual flows and bringing all data together for the complete budget enables a clear overview of the Nr flow patterns throughout the urban or peri-urban environments of all test areas. Identifying the pools where Nr enters, accumulates, or exits the system is essential for evaluating the impacts on both the environment and human health.

3.1. Urban Nr budget

A first visual evaluation of each system helps to identify flow patterns and central pools that might need further investigation to evaluate the system's circularity and impact on the environment. Looking at all test areas it becomes apparent that there is a clear distinction between the urban and peri-urban areas of all cities. The household pool with its import and export flows - import from industry and export to wastewater with subsequent Nr conversion to N₂ - plays a central role in all core areas. In the surrounding areas the urban plants pool and to a certain



Fig. 3. Nr budget scheme for the core (top) and surrounding (bottom) area of Vienna, highlighting the largest flows. Figures including all flows for all test areas can be found in the supplementary material (Figs. S3–9).

extent also the urban animals pool play a more central role, with a higher export of goods but also higher Nr losses to water and air, compared with the core areas (see Fig. 3 and Fig. S3-S8).

Looking at potential impacts on the environment through the air and water pools, the importance of identifying the central flows is emphasized. Fig. 4 shows the central pools of the urban and peri-urban area, respectively, make up the largest shares of Nr inflows to air and water. While main inflows to the air and water pool in the core areas come from combustion processes and wastewater treatment respectively, main inflows in the surrounding areas originate from agricultural, horticultural or urban green land. High Nr application on agricultural land drives Nr volatilization and leaching and run-off in the Chinese test areas' surroundings. An exception is the area surrounding Vienna with high emissions from transport and industry. This is possibly driven by





Fig. 4. Shares of inflows by source to the air pool (top) and to the water pool (bottom). Inflows from cultivated land (agricultural land, urban greens and horticulture) represent Nr volatilization (NH₃ and N₂O) from synthetic fertilizers, livestock and pet manure, compost and sewage sludge applied as fertilizers to soils. Inflows from livestock represent NH₃ and N₂O volatilization from manure management. Inflows from waste represent NH₃ and N₂O volatilization from waste treatment such as composting. Inflows from wastewater represent mostly N₂O volatilization from treatment (excluding N₂ emissions). Inflows from combustion represent mostly N₂O volatilization from treatment (excluding N₂ emissions).

commuters (148,000 from the surrounding area to the core area of Vienna - equivalent to around 22 % of total population in the surrounding area – Statistik Austria, 2023) and the oil refinery located in the area surrounding Vienna.

These similar patterns in all cities indicate that the included areas have comparable characteristics when choosing administrative boundaries to define the respective urban and peri-urban regions. However, to fully understand the system and put its flows to water and air in perspective, one needs to look more closely on how and where largest Nr import is converted and whether it is accumulated or exported. This again differs to a great extent between the urban and peri-urban environment but depending on local specificities it also differs between the test areas.

3.1.1. Urban areas

In urban Vienna, Nr entering the area is mainly transformed to N_2 (56% of Nr import) in the wastewater and combustion pool due to denitrification during wastewater treatment and exhaust cleaning

through catalytic converters during combustion processes (Fig. 5). This high ratio can be explained by a high population density (4407 people km⁻²) and low levels of N-relevant industrial and agricultural activities leading to only 1 % of Nr import being transformed to export products (Table S6). 20 % of Nr inflow is converted to Nr emissions to the atmosphere. This relatively high contribution can be explained by waste burning (including sewage sludge) taking place in the Vienna core area, making up over 80 % of Nr input to the combustion pool.

In Zielona Góra core, a wood processing factory situated in this area results in a rather high share (29 %) of Nr inflow being exported as products. The second highest share of imported Nr is converted and eventually emitted as N₂ emissions to the atmosphere with over 90 % of these emissions coming from wastewater treatment. A substantial amount of Nr import is also accumulated in households (10 %) and landfill (7 %). 11 % of Nr import is converted to and exported as waste materials as waste treatment in the urban area of Zielona Góra is limited and there is no capacity for recycling.

The largest share of Nr import is converted to and exported as goods



Fig. 5. Fate of Nr entering the respective test area boundaries. "Products out" include all exported industrial and agricultural products. "Air" indicates the share of emissions from the air pool to the outside of the system (transboundary emissions). "Waste out" encompasses all exported waste. "Stock change water" shows how much Nr accumulates in water. "Stock change urban plant" includes all Nr that remains in the soils of agricultural or horticultural land as well as urban greens. "Stock change household" shows all Nr accumulating in households and "stock change other" includes all Nr accumulating in the waste (landfills) and livestock and pet (N retention) pools.

(22 %) due to local fertilizer, rubber, fiber and plastic production in the core area of Shijiazhuang. Due to higher agricultural activity and excessive use of fertilizers on agricultural land, 20 % of imported Nr import accumulates in managed soil surfaces. Stock accumulation in households as wood furniture, plastic, resin, clothes and shoes is the third most common (16 %) endpoint for Nr. Less efficient wastewater treatment compared to European cities (only 73 % of Nr in wastewater removed), but especially a large share of untreated wastewater (35 %) leads to a lower share of Nr import being converted to N₂. As no information on the fate of this untreated wastewater is available, we could not model its further way through the urban system – marked as 'unknown' in Fig. 5.

In the Beijing core area, the highest share of imported Nr is converted and leaves the system boundaries as N_2 (49%), mostly (around 80%) due to exhaust-gas cleaning during combustion processes. The second highest share does not leave the system but remains within the test area as household stocks (23%) in the form of food, furniture, and clothing.

3.1.2. Peri-urban areas

In the area surrounding Vienna, 57 % of Nr import is converted and exported in the form of products. While agricultural products constitute around one third of this export, industrial products make up the other two thirds. Agricultural goods are exported to a larger extent because crop production exceeds three times the food demand of the local population and the feed needs of livestock. This is due to a low population density (156 persons per square kilometer) and a low livestock density index² (2.3 LSU ha⁻¹), combined with a high share of agricultural land in the total area (50 %). Industrial exports are feed, resins as well as bitumen and diesel from a local oil refinery. The second largest Nr conversion (and sink) is N₂, mainly (80 %) from exhaust cleaning from combustion processes in industry. Nr flow to groundwater also constitutes a larger share of total Nr import conversion of around 13 %. This is due to Nr leaching from fertilized agricultural areas.

In the area surrounding Zielona Góra, exported products constitute the largest share of imported Nr conversion products (55 %). These exported goods, however, are only from agricultural sources (livestock and plant products) as there is no significant industrial production in this area. The second largest part of imported Nr (21 %) is stored as stock in soils. This can be explained by large areas of urban greens (58 % of total area – mostly forest) increasing the significance of Nr input from Nr deposition to the total Nr import, especially in combination with low input to agricultural land from fertilizers (around 60 kg N ha⁻¹), no significant industrial activity and a rather low population density (127 people km⁻²). As there is no significant Nr output from urban green areas, an increased share of Nr import is stored in the soil. This high amount of Nr deposition is in absolute numbers higher than all Nr emissions to air due to low human and industrial or agricultural activity. Therefore, no transboundary emissions to the atmosphere (labelled 'air') are visible in Fig. 6.

In the area surrounding Shijiazhuang, the biggest share (55 %) of Nr import remains as stock in the local agricultural soils due to excessive mineral fertilizer use and double to triple cropping (around 300 kg/ha harvested area and around 1000 kg/ha physical area). The second and third biggest end points of Nr import are water (15 % of Nr import) and air (18 % of Nr import), due to Nr leaching and Nr volatilization from agricultural land.

In the area surrounding Beijing, the largest share of Nr import (30 %) remains in agricultural soils for the same reason as for Shijiazhuang – very high mineral fertilizer input and multi-cropping. The second biggest share of Nr import is converted to N₂ through wastewater treatment. 21 % of Nr import is transformed to emissions to the atmosphere, mainly through Nr volatilization from fertilized agricultural soils (55 % of emissions).

3.2. Indicators

After having identified the largest conversion and exit or accumulation points for Nr as well as the most important inflows to the water and air pool, the use of indicators helps to evaluate the system's overall circularity as well as potential harmful impact on human and ecosystem health.

3.2.1. Recycling rate

With an average of 87 % or 74 % of Nr import being accumulated or being emitted to air and water in the core or peri-urban area, the urgency to increase all system's circularity and reduce Nr losses or accumulation becomes apparent. Nr recycling, as defined in this paper, encompasses manure Nr recycled from livestock to urban plants and waste directed to industry or to urban plants (e.g. in the form of compost). Although these examples represent typical recycling flows that should be included in all calculations, additional flows for each test area's specificities need to be added to account for all recycling activities. In the area surrounding Vienna, sewage sludge being re-used as fertilizer on agricultural land is added to the recycling flows as well as home composting represented by the flows 'wastewater to urban plants'

² LSU (livestock unit) per agricultural area



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Fig. 6. Fate of Nr entering the respective test area boundaries. "Products out" include all exported industrial and agricultural products. "Air" indicates the share of emissions from the air pool to the outside of the system (transboundary emissions). "Waste out" encompasses all exported waste. "Stock change water" shows how much Nr accumulates in water. "Stock change urban plant" includes all Nr that remains in the soils of agricultural or horticultural land as well as urban greens. "Stock change household" shows all Nr accumulating in households and "stock change other" includes all Nr accumulating in the waste (landfills) and livestock and pet (N retention) pools.

and 'household to urban greens', respectively. In the Chinese test areas, the use of kitchen residues as livestock feed was added.

Areas with higher agricultural activity, mostly the peri-urban areas, show a higher recycling rate due to increased recycling of manure N, waste and wastewater/human excreta. The recycling rate is highest for the area surrounding Shijiazhuang, at around 14 %. While the main contributor to recycling flows (87 %) is manure Nr directed to agricultural land at 154 kg N ha⁻¹, the second largest contribution (11 %) comes from the recycling of kitchen residues as feed (0.1 kg N cap $^{-1}$). A relatively high share of Nr recycling from home composting is estimated in the area surrounding Vienna, at 66 kg N ha⁻¹ urban green area, constituting around 50 % of all recycling flows in this area. This estimate should be understood as upper limit and was derived by converting the average Austrian home composting estimate from an Austrian governmental agency of 177 kg per person to applied compost Nr (BMNT, 2017). No recycling takes place in the core area of Zielona Góra because no livestock is kept, and all waste is either exported or treated in a mechanical biological treatment plant (MBT) before it is landfilled (personal communication from the Point of Selective Collection of Municipal Waste in Zielona Góra). Sewage sludge is re-used as fertilizer outside the Zielona Góra core area (personal communication from the Sewage Treatment Plant in Zielona Góra).

On average, recycling rates are quite low (weighted average: 12 %) in all test areas, suggesting room for improvement of circularity, especially in the core areas. Increasing Nr recycling is important from a nitrogen perspective but also in terms of energy use and greenhouse gas emissions. Recycled Nr leads to a decrease in demand for industrial N fixation which accounts for 1-2 % of the global energy consumption and around 1 % of global CO₂ emissions (IEA, 2021; IFA, 2023).

3.2.2. Nitrogen use efficiency and nitrogen surplus to identify challenges

To evaluate Nr surplus on all cultivated areas, the framework of planetary boundaries as described by Steffen et al. (2015) was used. According to the authors, exceeding a threshold value of 55 kg Nr ha⁻¹ of Nr input to erodible soils has a high risk of perturbating the earth system's resilience.

For the evaluation of NUE, suggestions from the EU Nitrogen Expert Panel (2015) were taken into consideration. According to the Panel, a NUE in cropping systems below 50 % implies a high risk of Nr losses such as ammonia (NH₃) and nitrous oxide (N₂O) volatilization or nitrate (NO₃) leaching, with risks of Nr losses already evident below 70 %. NUE above 90 % indicates potential soil mining. Both Nr surplus and NUE are useful indicators, as the thresholds are defined more broadly and do not reflect local characteristics such as soil properties, climate and management practices which would be needed for a more detailed evaluation.

As can be seen in Table 1, the planetary boundary for Nr surplus is

exceeded in all Chinese test areas for all cultivated soils, and NUE remains low, indicating a high risk of losses to soil, water and air.³ As presented in Sections 3.1.1 and 3.1.2, the end point for Nr import to the urban and peri-urban area of Shijiazhuang as well as the peri-urban area of Beijing is soil stocks which, combined with these indicators, points to a high risk for environmental damage, especially a disruption of soil balance, very high Nr emissions to the air, losses the hydrosphere and soil acidification (Velthof et al., 2011). Although leaching and run-off rates are highest on Chinese agricultural land with over 190 kg N ha^{-1} , no increased NO₃ in groundwater was detected which could be explained by this number mostly representing run-off due to the high depth of groundwater bodies. However, as Zhou et al. (2016) show, there is a potential future threat to groundwater from this high Nr accumulation in soils, especially when considering increasing extreme rainfall events due to climate change. Subsequently, measures to reduce Nr input should be considered, especially for agricultural and urban green areas, but also for horticultural areas.

In Zielona Góra, only Nr on horticultural land (a relatively small area - 0.39 ha), exceeds the proposed thresholds, though to the opposite direction to the Chinese areas – towards soil mining. However, this value is very uncertain and may be explained by difficulties in correctly including the whole cultivation cycle, especially for imported flowers. While imported flowers bought as seedlings and grown in a long production cycle are fertilized on the horticultural land, plants imported in pots come with already fertilized soil. In case of a fast turnover with retention up to two weeks, there is no need to additionally fertilize these potted plants. As such detailed data were not available, it was not possible to separate these different types of imported plants, and while the calculations show soil mining, this is most likely not the case.

Although we identified that the largest part of Nr import to Zielona Góra surrounding remains in urban green area soil stocks (Section 3.1), the Nr surplus remains low and does not exceed the threshold value due to the large urban green areas. Hence, on average, no risk for environmental damage is to be expected for the wider area, in terms of this indicator. Conversely, Nr surplus and NUE suggest a high risk for environmental impact on horticultural areas due to high Nr input and little Nr output. Horticulture in Zielona Góra surrounding mainly encompasses extensive orchard cultivation as well as wholesale and retail trade in plant material (ornamental and bedding plants). The area occupied by this activity is very small, but the flow of plant material is relatively large. In the context of bedding plants sale and production, local

 $^{^3}$ The relatively high NUE paired with a high Nr surplus for horticultural areas in Shijiazhuang core and surrounding can be explained by high in- and outputs (over 600 kg/ha) on relatively small areas (222 ha in the core and 16 ha in the surrounding area.

Table 1

Nitrogen surplus (N_{sur}) and Nitrogen Use Efficiency (NUE) for agricultural land, horticultural land, and urban green land for all test areas. Values below/above threshold values in bold print.

	Vienna core	Vienna surrounding	Zielona Gora core	Zielona Gora surrounding	Shijiazhuang core	Shijiazhuang surrounding	Beijing core	Beijing surrounding
Agricultural N _{sur} [kg/ ha]	land 62	46	19	16	638	986	449	890
NUE [%]	55	68	76	82	28	19	2	7
Horticultura	l land							
N _{sur} [kg/ ha]	107	39	(379)	100	116	98	168	168
NUE [%]	33	55	587	7	83	70	52	52
Urban green	land							
N _{sur} [kg/	40	110	34	17	107	121	400	184
NUE [%]	23	47			11	10	3	7
A 11	lond							
N _{sur} [kg/ ha]	48	50	32	17	443	976	334	641
NUE [%]	40	65	22	60	28	19	18	10

gardening companies serve the entire area of municipal green areas and individual buyers.

In the Vienna core area, all cultivated areas show exceedance of the indicator thresholds for either Nr surplus or NUE, indicating a high risk of Nr losses to soil, air and water from horticultural areas and urban greens, and a risk of losses from agricultural lands. These risks were not discovered in the initial analysis of end points of Nr in the total system due to the high Nr values linked to combustion that overshadowed the Nr flows linked to cultivated areas. This emphasizes the importance of separate analysis of relevant indicators.

In the area surrounding Vienna, the risk of environmental impacts as expressed with the two indicators is lower, however still heightened for horticultural areas and urban greens. In the surrounding area, the share of Nr input to water was relatively high (13 %). For both the urban and peri-urban areas, it was possible to partially validate the results by consulting groundwater nitrate (NO₃) monitoring values. This analysis showed that NO₃ levels exceeded the national threshold value of 45 mg/l in two groundwater bodies below Vienna and its surrounding area in 2015 (BMLFUW, n.d.). Exceeding this legal threshold not only affects the environment but also human health (WHO, 2011; Feichtinger, 2013). Therefore, a reduction in Nr input to all cultivated soils, not just agricultural soils, is highly recommended.

Combining the separate indicators and looking at total Nr surplus and NUE for all managed areas reveals that all test areas, except for the areas surrounding Vienna and Zielona Gora, show a NUE below the threshold of 50 %, indicating that Nr input is too high in relation to the Nr output achieved.

4. Discussion

Analyzing largest flows, conversion and exit or accumulation points and using different indicators for evaluation, specific challenges were identified for each test area. In particular, Nr input to urban plant areas needs to be reduced in all Chinese test areas, in the Vienna core area and on urban greens and horticultural areas in peri-urban Vienna as well as on urban greens in the area surrounding Zielona Góra, to avoid soil acidification and groundwater contamination. Additionally, the recycling rates for all test areas have substantial potential for improvement, especially in the core areas and particularly in urban Zielona Góra, where no Nr is recycled.

4.1. Reducing Nr losses

Improved Nr management such as incorporation and deep injection of manures, optimized diet composition and cutting food waste, could improve these situations (Hansen et al., 2017). For China, a combination of dietary change, reduced synthetic fertilizer application and a higher rate of synthetic fertilizer incorporation into soils could decrease Nr losses by up to 63 % compared with Business As Usual (BAU) in 2050 (Zhan et al., 2021; Cui et al., 2022).

4.2. Potentials for improving Nr recycling

To increase Nr recycling rates, the contributing flows need to be assessed to identify potential options. These can differ between test areas, depending on local conditions such as technical feasibilities or legal constraints. Here we focused on (i) the recovery of nutrients from wastewater and sewage sludge flows, (ii) the recycling of nightsoil and/ or urine to agricultural fields, and (iii) the recovery of the organic fraction of municipal solid waste.

4.2.1. Wastewater and sewage sludge streams

For Vienna, a potential solution was found in the recycling of sewage sludge which is currently all burnt and used for district heating (Stadt Wien, n.d.). Substituting mineral fertilizer with sewage sludge would result in a reduction potential of 1-2kt CO₂ as less synthetic Nr fixation is needed. This assumes that 88 % of sewage sludge is composted and around 50 % of this compost can be used on agricultural fields (assumption based on the current situation in the Vienna surrounding area). The Nr in this composted sludge would be sufficient to substitute the total use of mineral Nr fertilizer in the Vienna core area, with two thirds of the compost still available for export. A disadvantage of this substitution would be increased ammonia emissions due to a higher volatilization rate compared to synthetic Nr fertilizers (see supplementary material - AllComp.xlsx). Other promising technologies for Nutrient Recovery and Reuse (NRR) from wastewater streams identified by van der Hoek et al. (2018) include struvite precipitation, the treatment of air from thermal sludge drying facilities, and of digester reject water through membrane filtration devices or air stripping. These technologies can achieve 1.1 %, 2.1 %, 20 %, and 24 % of Nr recovery from a wastewater treatment plant's total Nr inflow, respectively, whereas sludge reuse yields about 11 %. While struvite precipitation and thermal sludge drying are mature technologies already applied in

practice, other better performing methods may require additional development before implementation, such as higher N concentrations in the digester reject water (membranes) or new/improved sanitation infrastructure (separate urine collection).

Maximum increases in the Nr recycling rates from wastewater streams are achieved through the parallel introduction of alternatives throughout the wastewater treatment process, including sludge reuse and Nr recovery from urine as well as from digester reject water. In a best-case scenario, the application of up to 90 % of sewage sludge on agricultural lands (with the highest percentage among the Member States of the European Union in Portugal: Buckwell et al., 2016) would suffice to fully replace the Nr mineral fertilizer requirements in the core areas of Vienna, Beijing, and Zielona Góra, as well as about 12 % and 29 % of the respective needs of the surrounding areas, following Nr surplus redistribution. Due to both smaller quantities of sludge produced, and larger quantities of mineral fertilizer applied on fields, the effect on Nr recycling in the Chinese surrounding areas from the implementation of these technologies remains small (Table 4).

4.2.2. Nightsoil and urine collection

Another way to increase recycling of Nr in wastewater is the direct use of human excreta which is still practiced in some parts of China and South-East Asia (Liu et al., 2014). While comprehensive guidelines now exist for the safe use of human excreta in agriculture, challenges related to social acceptance and higher ammonia volatilization compared to traditional Wastewater Treatment Plants (WWTP) still hinder the technological expansion of this approach (Jönsson et al., 2004; Moya et al., 2019; Spångberg et al., 2014).

In the field of ecological sanitation (ecosan), research is underway to investigate the use of human urine as a fertilizer for crop production to promote nutrient recovery and to close nutrient loops. Urine, which contains the largest proportion of nitrogen (90 %), phosphorus (50-65 %), and potassium (50-80 %) among household blackwater, presents a promising alternative to extracting Human Excreta Derived Fertilizer (HEDF) from traditional WWTP due to its lower microbial content and reduced risk to human health (Hilton et al., 2021; Rose et al., 2015). Urine diversion and source separation, whether through decentralized systems or on-site nutrient recovery within individual toilets, are crucial components of new design concepts aimed at promoting the safe and sustainable use of human urine (Kavvada et al., 2017; Randall and Naidoo, 2018; Wald, 2022). Despite research efforts, industrialized urine treatment reactors are vet to be established (Larsen et al., 2021). Successful implementation of urine recovery technologies requires consideration of local conditions such as street width for urine collection and type of housing. It is also crucial to involve stakeholders such as farmers and government officials, as demonstrated by Magid et al.

(2006) in their study on Nr recovery from urine technologies.

According to Jönsson et al. (2004) and Rose et al. (2015), the urinary Nr excretion rate is estimated to be 4 kg per capita per year. Using urine recovery technologies without considering any losses from the process, it would be possible to fully replace the use of mineral Nr fertilizer in the core areas of Zielona Góra, Vienna, and Beijing. Additionally, by redistributing the Nr surplus from the core areas to the surrounding areas, the recovered Nr could fully replace the mineral fertilizer requirements in the area surrounding Zielona Góra and satisfy about 43 % and 33 % of the surrounding areas' nitrogen mineral fertilizer needs for Vienna and Beijing, respectively (Table 2).

4.2.3. Organic waste composting

The composting of the organic waste fraction from Municipal Solid Waste (MSW) is a third significant pathway for Nr recovery. In the European Union, the organic fraction of MSW, which includes green waste, household waste, food waste from food services (e.g., restaurants, caterers), and retail, amounts to an average of 37 % of total MSW or about 88 Mt. year⁻¹ (Saveyn and Eder, 2013). Although large quantities of organic waste are produced, recovery from such waste streams is typically more challenging due to reduced nutrient concentrations and more heterogeneous waste composition.

In Vienna, the organic waste fractions amount to 37 % and 12 % of total MSW in the core and surrounding areas, respectively. Assuming a composting yield from organic waste of 37 % (Buckwell and Nadeu, 2016) and a best-case scenario where all biodegradable waste is composted or reused, this would cover the mineral fertilizer needs of the agricultural land of the Vienna core area. However, as the organic waste fraction is smaller in the surrounding area of Vienna and agricultural production is larger, only 0.3 % of synthetic Nr fertilizer could be replaced by recycling organic waste to agricultural land outside the core area. In Zielona Góra, the amount of bio-waste that could be recovered from municipal waste in 2015 constituted 29 % of the waste mass, which corresponds to 94,861 kg N year⁻¹. This amount of Nr would meet 100 % of the Nr demand in the core area of Zielona Góra and about 1 % of the fertilizer demand in the surrounding area of Zielona Góra. Additionally, Nr from sewage sludge (amounting to $355,000 \text{ kg N year}^{-1}$) which is currently exported, would cover 99 % of the total Nr fertilizer demand in both the core and surrounding area of Zielona Góra.

Adding all potential alternative Nr fertilizer sources together would lead to a full coverage of the fertilizer needs of the core areas of Vienna and Beijing, and the core and surrounding areas of Zielona Góra. The other more agriculturally active surrounding areas' fertilizer demands and all of Shijiazhuang's fertilizer demand cannot be fully met by making use of all potential alternatives. However, as was shown by Fang et al. (2023), the substitution of even smaller amounts of synthetic Nr

Table	2
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Estimates of recovered amounts of Nr from sewage sludge, urine, and compost of organic waste in all test
areas (kt N year ⁻¹) and their percentages compared to mineral fertilizer requirements before and after Nr
surplus redistribution.

	Vienna Core	Vienna Surrounding	Zielona Góra Core	Zielona Góra Surrounding	Shijiazhuang Core	Shijiazhuang Surrounding	Beijing Core	Beijing Surrounding
Sewage sludge [kt N year ⁻¹]	2	1	0.1	NA	0.2	0.6	1.4	1
Mineral fertilizer use after redistribution [%]	100	12	100	29	2	0	21	1
Urine [kt N year ⁻¹]	7	3	0.5	0.1	5	28	34	21
Mineral fertilizer use after redistribution [%]	100	43	100	125	45	3	100	33
MSW organic fraction [%]	37	12	29	NA	NA	NA	NA	NA
Organic waste composting [kt N year ⁻¹]	0.6	0.06	0.03	NA	NA	NA	NA	NA
Mineral fertilizer use after redistribution [%]	100	1	100	1	NA	NA	NA	NA
Sum of potentials [kt N year ⁻¹]	10	4	0.7	0.08	5	29	35	22
Mineral fertilizer use after redistribution [%]	100	61	100	174	47	3	100	35

fertilizer with recycled food waste can lead to a yield increase which adds further value to this recycling pathway.

5. Conclusions

The new framework for urban nitrogen budgets developed here allows to analyze Nr flows through both urban and peri-urban environments, identifying endpoints of Nr flows as well as Nr accumulation. In combination with given indicators, potential environmental impacts can be pointed out and potentials for increased Nr recycling can be identified. Using the framework for several different test areas allowed consistent comparisons and testing of assumptions based on flow patterns from a single test area. Distinct differences became evident between the urban and peri-urban area for all cities with additional differences between test areas due to local conditions such as industrial or agricultural production levels and population density. The core areas were found to be centers of consumption linked to Nr emissions from human activities (centered around waste, wastewater and combustion), lower Nr recycling rates (0-5 %) and a mostly linear transgression of Nr flows. The surrounding areas were characterized by agricultural production with significant Nr accumulation in soils and emissions to water and air but higher Nr recycling rates (6-14 %) mostly due to manure Nr recycling. While improved Nr management such as incorporation and deep injection of manures and optimized diet will be needed to increase NUE to above 50 % on cultivated areas in all Chinese test areas and both European core areas, recycling urine and sewage sludge as well as organic fractions of waste has the potential to substitute all synthetic Nr fertilizer use in all core areas.

This framework not only enables the comprehensive and comparable modelling of Nr flows through the urban and peri-urban environment but helps identifying potentially harmful flows and exploring options for increased Nr recycling.

CRediT authorship contribution statement

Katrin Kaltenegger: Writing – original draft, Methodology, Software, Investigation, Validation, Data curation. Zhaohai Bai: Methodology, Software. Ulrike Dragosits: Writing – review & editing. Xiangwen Fan: Investigation. Andrzej Greinert: Investigation, Writing – review & editing. Samuel Guéret: Investigation, Writing – original draft, Writing – review & editing. Monika Suchowska-Kisielewicz: Investigation, Writing – review & editing. Wilfried Winiwarter: Project administration, Conceptualization, Methodology, Writing – review & editing, Supervision. Lin Zhang: Project administration, Methodology, Conceptualization. Feng Zhou: Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that support the findings of this study are openly available in the supplementary material. The STAN model for urban nitrogen budgets is openly available on the STAN platform as "Urban Nitrogen Budgets" (https://www.stan2web.net/downloads).

Acknowledgments

This publication contributes to UNCNET, a project funded under the JPI Urban Europe/China collaboration, project numbers UMO-2018/29/Z/ST10/0298 (NCN, Poland), 71961137011 (NSFC, China) and 870234 (FFG, Austria).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.166827.

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