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## **UNCNET**

# Urban nitrogen cycles: new economy thinking to master the challenges of climate change

D7/1: Conceptual Nitrogen budget

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Authors: Wilfried Winiwarter

Dissemination Level		
PU	Public	
PP	Restricted to other programme participants (including funding agencies)	$\boxtimes$
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#### **Executive Summary**

Establishing Urban Nitrogen Budgets requires to view cities and their material supply just as a metabolism. Flows of reactive nitrogen compounds, sources (formation of reactive nitrogen, i.e. fixation from molecular/gaseous dinitrogen) and sinks (denitrification, formation of dinitrogen) are processes to be explored. Here we performed a comprehensive literature survey over available approaches and their interpretation to learn about methodologies and best practice applications. It turns out that sectoral budgets and budgets related to environmental media alone may provide relevant information. Media of interest are those providing transport (air and water) and storage media (soil in particular). The food sector as well as waste/wastewater have been identified as sectors of particular interest. Also complete budgets are available (attempting to cover all flows of reactive nitrogen), but the numbers of cities for which such efforts have been made public is limited, out of seven cities in total there are four in China, and five in Asia. In all cases, temporal trends that allow to assess developments have proven very useful. Many aspects can be viewed in their development over time, and such development is very visible under Asian (Chinese) conditions. Moreover, a spatial indicator (mass of N per area, e.g. kg N ha<sup>-1</sup>) proves useful in comparing across different approaches. Also an indicator based on population (mass of N per inhabitant) has been used to render results comparable, but provide less information about potential impacts.

The nitrogen cascade, with its multiple pools and flows, conceptually allows for a number of levels of validation. The available scientific literature tends to use this potential to recover missing information rather than to verify the information provided. The budget approach has been used to develop a coarse understanding also in situations where little environmental information can be otherwise obtained. Further use of results may be possible when collecting additional data on N flows and pool changes. In this manner, diverging assumptions can be flagged and possibly errors identified and corrected, or flows can be reconciled and uncertainties quantified.

The set of existing approaches also allow to develop descriptive parameters for describing similarities and differences between patterns of urban nitrogen flows. The first is a degree of separation – to which remain different input pathways separated even when leaving the city? When separately looking at food processing and combustion, N flow patterns often may remain very independent and just reflecting parallel nitrogen cascades rather than an interacting web of connections. Separate processing may simplify the evaluation, and also directs potential abatement measures more directly to the immediate source of a potential environmental damage. Secondly, the level of recycling vs. the level of pollution per input of reactive N can be quantified. A way of reducing is denitrification as in wastewater treatment – reactive N becomes harmless, yet valuable resources may go lost: again fixing fertilizer for nitrogen requires substantial amounts of energy. Recycling, by contrast, is N that finds good use even after being released to the environment – e.g. as a nutrient in compost. Hence, the rate of recycling becomes a meaningful parameter, especially in comparisons. Finally, nitrogen stocks and stock changes in cities are fully underresearched. With nutrient abundancy in cities, yet in the wrong places,

Obtaining better understanding of these three items (or parameters) will be of use also for policy makers and/or other stakeholders in the city. First of all, clarifying the source apportionment may direct regulation to where it is needed, and avoid misallocation. Furthermore, understanding sites (and pools) of N accumulation may avoid hazards of pollution, even before they become leaky and impact the environment. Finally, proper benchmarking using parameters such as recycling rate or loss rate may guide towards the potentials achieved elsewhere, in order to develop ambitions to reduce on nitrogen pollution (affecting air and water quality, biodiversity and the climate) at home.



## **ANNEX 1**

Urban nitrogen budgets: flows and stock changes of potentially polluting nitrogen compounds in cities and their surroundings – a review

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Urban nitrogen budgets: flows and stock changes of potentially polluting nitrogen compounds in cities and their surroundings – a review

Wilfried Winiwarter<sup>1,2</sup>, Barbara Amon<sup>2,3</sup>, Zhaohai Bai<sup>4</sup>, Andrzej Greinert<sup>2</sup>, Katrin Kaltenegger<sup>1</sup>, Lin Ma<sup>4</sup>, Sylwia Myszograj<sup>2</sup>, Markus Schneidergruber<sup>5</sup>, Monika Suchowski-Kisielewicz<sup>2</sup>, Lisa Wolf<sup>6</sup>, Lin Zhang<sup>7</sup>, Feng Zhou<sup>8</sup>

#### **Abstract**

Concepts of material flow and mass consistency of nitrogen compounds have been used to elucidate nitrogen's fate in an urban environment. While reactive nitrogen commonly is associated to agriculture and hence to large areas, here we have compiled scientific literature on nitrogen budget approaches in cities, following the central role cities have in anthropogenic activities generally. This included studies that specifically dealt with individual sectors as well as budgets covering all inputs and outputs to and from a city across all sectors and media. In the available data set, a clear focus on Asian cities was noted, making full use of limited information and thus enable to quantitatively describe a local pollution situation. Time series comparisons helped to identify trends, but comparison between cities was hampered by a lack of harmonized methodologies. Some standardization, or at least improved reference to relevant standardized data collection along international norms was considered helpful. Analysis of results available pointed to the following aspects that would reveal additional benchmarks for urban nitrogen budgets: analyzing the share of nitrogen that is recycled or reused, separating largely independent sets of nitrogen flows specifically between food nitrogen streams and fossil fuel combustion related flows, and estimating the stock changes for the whole domain or within individual pools.

#### **Keywords**

Nitrogen budget; urban metabolism; agro-food chain; nitrogen cascade; material flow analysis

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<sup>&</sup>lt;sup>1</sup> International Institute for Applied Systems Analysis (IIASA), A-2361 Laxenburg, Austria

<sup>&</sup>lt;sup>2</sup> Institute of Environmental Engineering, University of Zielona Góra, PL 65-417 Zielona Góra, Poland

<sup>&</sup>lt;sup>3</sup> Leibniz Institute for Agricultural Engineering and Bioeconomy (ATB), 14469 Potsdam, Germany

<sup>&</sup>lt;sup>4</sup>CARR, CAS, Shijiazhuang 050021, Hebei, China

<sup>&</sup>lt;sup>5</sup> brainbows, 1010 Vienna, Austria

<sup>&</sup>lt;sup>6</sup> E.C.O. Institute of Ecology, 9020 Klagenfurt, Austria

<sup>&</sup>lt;sup>7</sup> Laboratory for Climate and Ocean–Atmosphere Studies, Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing 100871, China

<sup>&</sup>lt;sup>8</sup> College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

#### 1. Introduction

On a global scale, human activities greatly accelerated the cycle of reactive nitrogen compounds – reactive nitrogen (Nr) representing all chemical forms of N except for the dominant stable form of molecular N<sub>2</sub>. At currently about twice its natural level (Fowler et al., 2013), environmental impacts of Nr to air, water, soils, climate and biodiversity have been observed and investigated at different scales (Sutton et al., 2011; Erisman et al., 2013). As nutrients to boost plant production for human consumption are a key element to the increase observed, analysis logically focused on agriculture as one key sector, hence predominantly covering area sources. Administrative regions such as countries that provide plentiful statistical data therefore are considered adequate to investigate the fate of nitrogen compounds, e.g. in the form of national nitrogen budgets (Leip et al., 2011). On a global scale, the nitrogen cycle is believed to be one of the very few confirmed examples that human activities already now have exceeded the "planetary boundary" of a safe and sustainable earth (Rockström et al., 2009; Steffen et al., 2015).

Anthropogenic activities focus on cities. With, globally, 55% of the population living in cities, a number that is expected to increase to 68% by 2050 (UN, 2018), cities provide an immense opportunity for interventions in malfunctioning systems on a global scale. Already now, about 80% of GDP is generated in cities (Grübler and Fisk, 2012). Interventions on an urban scale have proven to be more efficient and less time consuming than similar action on a national or country level, which often require legislative procedures and involvement of a large number of stakeholders. Hence also several policy processes that previously had been under the authority of national governments, have started their urban initiatives and show potential towards significant progress (Kuramochi et al., 2019).

Here we hypothesize that not only their high population density make cities a clear target for successful environmental remedies (in a combination of enhanced impacts and the associated political strength) also in relation to Nr. We argue that, further to that, urban lifestyle, urban practices and production patterns directly influence the local nitrogen cycle, so that action in cities and around cities, in the peri-urban space, can lead to immediate benefits that a large share of the population profit from. While Nr and the "nitrogen cascade" (Galloway et al., 2003) typically are associated with agricultural activity (e.g., Galloway et al., 2008), reports occasionally point to predominant impacts from urban sources, e.g. on urban air pollution related to Nr (Pan et al., 2016). Moreover, urban and peri-urban agriculture may specifically affect the environment (Zhao et al., 2017). The role of cities for a future global food system (and their relationship to greenhouse gases) has just recently been emphasized (Pradhan et al., 2020).

In order to understand the level of support given by science to tackle potential impacts related to urban Nr flows, we investigated the scientific literature for their use of nitrogen budget approaches to cities. Specifically, we searched for the terms "urban" in combination with either "nitrogen budgets" or "nitrogen balance". Both Google Scholar (titles only) and Scopus (title, keywords and abstract) were used. From the resulting overview of papers we selected those that specifically attempted to quantify physical flows of Nr compounds in a clearly urban setting (areas within city administrative boundaries or peri-urban regions with high population density). Only physical flows were covered, indirect effects or footprints were not part of this analysis. We noted that a sizable number of studies focused on specific sectors or environmental media only so that we had to analyze them separately from the investigations of full budgets that addressed interactions beween all sectors. The set of puclications was extended by relevant articles cited in these papers, as well as by articles referencing them. Specific attention was given to identifying issues that could characterize Nr flows or make them comparable between individual studies. All of this material allowed us to assess which approaches these studies have in common, what results deserve particular attention and in which areas further specific development of urban nitrogen budgets are needed.

This paper is structured as follows. Evidence and examples of the importance of sectoral nitrogen budgets on urban scales are shown in section 2, while section 3 reports on full urban nitrogen budgets. These two sections provide an overview of the breadth of approaches and the diversity of concepts. In section 4 we identify common elements from these studies and conclude in section 5.

#### 2. Sectoral views on nitrogen inputs and outputs

Nitrogen budgets use material flow approaches to constrain the amount of Nr by quantifying flows in and out of a system boundary, the sources and sinks within the boundary and stock changes. The method allows to accommodate analysis of individual sectors, or of environmental media that commonly offer storage and transport functions for Nr. For the latter, observations along homogeneous physical media (soil, water, atmosphere) are available, for the former budget studies on specific sectors of anthropogenic activities have been made (food system or waste and wastewater). Differences in system boundaries, substrates considered, and sectors covered provide considerable challenges to compare results of published studies.

Soil: Soil nitrogen budgets have been a classical instrument in agronomy, hence their application on urban agricultural areas was merely a logical extension. Hou et al. (2012) studied smallholder farms in peri-urban Beijing, and they observed N surplus about three times as high in vegetable systems (nearly 1600 kg N ha<sup>-1</sup> yr<sup>-1</sup>) compared to orchards or cereal production. While reported variability between farmers was high and quantity clearly depended on practices, the relevance of cropping type and especially of quantity of vegetable production was shown clearly. A totally different system, application of mineral fertilizers on private gardens and lawns, was investigated by Law et al. (2004) for Baltimore. Again characterized by huge variability, average application rates of almost 100 kg N ha<sup>-1</sup> yr<sup>-1</sup> were determined for systems that would not target for production. Analyses also were available under N deficit conditions (Tadesse et al., 2019) in two peri-urban regions in Ethiopia. Livestock-oriented mixed farms had higher income, obtained external sources of N and hence 4-5 times higher N input, but a poor nitrogen use efficiency (NUE) compared to crop-oriented mixed farms. High-input systems, at least in relative terms, have been observed in urban agriculture in Ghana (Werner et al., 2019), where the widespread use of wastewater for irrigation led to nitrate leaching losses as high as 200 kg N ha<sup>-1</sup> yr<sup>-1</sup>.

Water: With water being an essential transport medium for Nr, budgets on riverine N were available also on an urban scale. Barles and Lestel (2007) focused on a historical situation, nineteenth-century Paris, and early quantification attempts of nitrogen losses along rivers. These authors also described efforts to reclaim urban nitrogen for agronomic purposes, in a situation where nitrogen was a scarce resource. Differences between measured upstream and downstream concentrations of nitrogen compounds have been used to identify the urban contributions to the pollution of the Po river near the city of Turin, Italy, and to estimate the phenomena responsible (Genon and Marchese, 2007). Extending such an approach to the total upstream area led to consideration of watersheds, a logical spatial delineation when working with rivers and water bodies as transport media. Watersheds provided the basis for inverse modelling by Divers et al. (2013) to quantify sewage leakage in an urban watershed of Pittsburgh, PA, at rates between 6 and 14 kg N ha<sup>-1</sup> yr<sup>-1</sup>, based on biweekly water sampling and measurements. Urban farming impacts on water quality were studied by Cameira et al. (2014) for vegetable gardens in Lisbon (Portugal), with mean observed drainage concentrations of almost 300 mg L<sup>-1</sup> NO<sub>3</sub> and N accumulation rates in lower soil depth found close to 100 kg ha<sup>-1</sup> yr<sup>-1</sup>. A combination of dedicated measurements and an inventory approach was taken to assess the N budget of a small watershed in the city of Changchun (Jilin province, North-Eastern China). The study area, agricultural land constituting a major source of drinking water for the city, received close to 190 kg N ha<sup>-1</sup> yr<sup>-1</sup> as inputs, and outputs were about 100 kg N ha<sup>-1</sup> yr<sup>-1</sup> (including N contained in product, leaching and denitrification), indicating a considerable N accumulation in

soil due to human activities (Song and Liu, 2013). Human impacts were also quantified to a large water body next to a city, the Lake Pontchartrain next to New Orleans (LA), by Turner et al. (2002). While still nitrogen limited, the authors find a 10-fold increase in nitrogen loading due to anthropogenic activities, most of all by water advection. N loads could triple easily when flood protection gates were being opened to divert Mississippi river water into the lake, or water diversion projects for wetland protection were implemented, demonstrating the environmental trade-offs encountered in urban environments. In a statistical analysis of measurements of ionic component (ammonia, nitrate, nitrite) from rivers in Moscow, Russia, Laryushkin-Zheleznyi and Novikov (2005) identified unimodal distributions for nitrite, but often bimodal distribution for other N forms. They used correlations between the respective components to develop a kinetic model describing nitrification and denitrification processes, suggesting maximum admissible levels of total ionic N of 3.5 mg L<sup>-1</sup> and NH<sub>4</sub> of 1.3 mg L<sup>-1</sup> to prevent assimilation of N species by aquatic organisms. Atmosphere: Nitrogen budgets seemed to play much less of a role for the other important transport medium, the atmosphere. Urban airshed and air quality modelling provided a detailed account of a number of trace constituents, their distribution, conversion, and deposition to the surface, but the atmospheric reality was considered much less appropriate to take advantage of specifically limiting nitrogen. Instead, other elements of conversion took a role, like the occurrences of urban ozone pollution formed by photochemical reactions of nitrogen monoxide and nitrogen dioxide with volatile organic compounds (Lu et al., 2018), or the formation of particulate matter (secondary inorganic aerosols) by combination of ammonia, sulfur dioxide and nitrogen oxides, or their reaction products (Stokstad, 2014). The lifetimes of Nr and its products in the atmosphere are about a few weeks (Fowler et al., 2013), and thus urban or peri-urban Nr emissions can be transported outside and affect air quality over a broader region. Source apportionment studies (see the overview by Viana et al., 2008) allowed to obtain a more direct link between the release of gaseous compounds and observed atmospheric concentrations of particulate matter. Using a source attribution method, Zhang et al. (2015) found that reduced and oxidized nitrogen emissions together contribute about 22% of the heavy wintertime particulate matter at Beijing. Food: With reactive nitrogen being a key element of protein, and protein being essential in human food, observing the fate of reactive nitrogen along the food chain seemed plausible. Forkes (2007) investigated the urban flow of food and N contained within for the city of Toronto, Canada. Quantifying flows for three years within a 15 year period, the author found improvements on a very low level in reclamation of N due to organic waste diversion efforts (from below one permille in 1990 to 4.7 and 2.3 % in the 2000's). N in the food system behaved basically very linearly. More decisive changes observed were improvements of the wastewater treatment system, which effectively removed N from waste streams and converted a growing fraction (more than 40% of inputs in the food system in the last year observed, 2004) into molecular N<sub>2</sub>. A much more dynamic situation than for Toronto was described by Ma et al. (2014), who analyzed impacts of urban expansion on nitrogen and phosphorus flows in the food system of Beijing over a 30-year period (1978 – 2008). Using a combination of statistical data bases, surveys and the NUFER model (nutrient flow in the food system, environment and resource), these authors captured the greatly increased food imports to Beijing metropolitan area as a consequence of a rapid increase in the number of temporary migrants. While input of N to the Beijing food system increased from 180 to 281 Gg during the observation period, the share of N in food and feed imports doubled from 31 to 63%, and losses of ammonia and N<sub>2</sub>O to air and of N to groundwater and surface waters increased by a factor of about 3. Overproportionate increase in pollution was interpreted as lack of treatment of waste streams – about 52% of N input accumulated as wastes (in crop residues, animal excreta, and human excreta and household wastes). This pointed to abatement potential for N pollution, e.g. from livestock production in high intensive peri-urban agriculture. Wei et al. (2018) showed for Beijing that the success of existing and future polices relied on optimizing spatial management of

new livestock production systems. They concluded to focus on optimizing livestock diet and onfarm manure management in industrial livestock production systems typical for peri-urban regions. Waste and Wastewater: Due to its function of N burying and removal (conversion to molecular N<sub>2</sub> by nitrification), the waste and wastewater sector takes a central role in the urban N cycle. Municipal waste treatment plants have a great impact on the reduction of Nr and the control of direct emission to air, soil and water. Yet, little research was found that would comprehensively allow to trace the fate of N compounds. Instead, just the emissions of N<sub>2</sub>O and NH<sub>3</sub> during waste treatment and storage were commonly reported. NH<sub>3</sub> emissions occured mainly in the composting and storage process. N2O was formed at every stage of waste management, from treatment to landfilling. According to Beck-Friis et al. (2001) about 98% of N emissions from waste were NH<sub>3</sub> and 2% were N<sub>2</sub>O. Emissions can vary widely (two orders of magnitude), in general increasing with longer treatment times (Clemens and Cuhls, 2008). In landfills, leachate was considered a source of N<sub>2</sub>O when recirculated (Lee et al., 2002). As a source of N<sub>2</sub>O emissions to the atmosphere and nitrate release to water bodies, wastewater treatment plants were more relevant than landfills (Svoboda et al., 2006), even while highly dynamic and depending on operational conditions (Arnell, 2016). Plants that achieved high levels of nitrogen removal generally emitted less N<sub>2</sub>O (Law et al., 2012). Both the nitrification step (oxidation of ammonium/ammonia to nitrite) and denitrification of nitrite or nitrate to molecular N<sub>2</sub> contributed to N<sub>2</sub>O formation. Removal of N compounds was never complete (Wen et al., 2018). Due to its relatively high solubility in water, N<sub>2</sub>O could be retained in the aqueous phase and denitrified downstream of a wastewater treatment plant, unless aeration caused N<sub>2</sub>O to be stripped to the atmosphere (Kampschreur et al., 2009). Remaining N from waste water treatment plants generally was first treated (composting, liming, and/or anaerobic digestion) before being recycled back to agricultural soils via direct land application. Recovery to ammonium sulfate or ammonium nitrate guided towards production of nitrogen fertilizer with positive environmental impacts (Shaddel et al., 2019).

#### 3. Evaluating complete urban flows

Bringing together information from individual sectors or media, a city can be investigated as a whole. Urban systems have features that resemble an organism: materials enter, they are converted, and again leave the city boundary. Conceptualizing a city as a metabolism (Wolman, 1965; Kennedy et al., 2007) facilitates material flow analysis to be performed. The approach allows adopting biological principles and denominating the investigated situation an "ecosystem". A simple example for such an approach has been presented by Svirejeva-Hopkins et al. (2011), who developed an urban N budget for the city of Paris and its urbanized surroundings for the year 2006. Largely independently, fossil fuel related emissions to the atmosphere, and urban food consumption including wastewater treatment formed two separate chains of N flows. While food imports into the city constituted the largest N flow, just over half of that amount was being converted into molecular  $N_2$  in wastewater treatment plants. The largest impact on the environment, by quantity, came from gaseous emissions from fossil fuel combustion. Hence, reducing road traffic NO<sub>x</sub> emissions was identified having the largest potential to reduce negative impacts of Nr. Enabling recycling of N in wastewater treatment would have the largest overall impact towards circular economy. Recycling of N (and P) also was the focus of a study on Bangkok Province, Thailand (Faerge et al., 2001), purely based on aggregated statistics and literature values for N contents of goods. Huge quantities of N discharged into the nearby river (and the sea), constituting 92% of inputs, kept recycling rates at a low 7%, for an overall balanced situation of N flows in and out of the city in the year 1996. According to the authors, improving the ecological nutrient cycle could be possible by operating wastewater and night soil (human excreta) treatment plants, which would allow to return

more N to food production, but even with 50% of households connected the resulting recycling rates would remain at a meager 11%.

A comprehensive accounting of flows of N in and out of city boundaries, largely based on locally collected data, was performed for the larger Phoenix, AZ, area (Baker et al., 2001), an arid region that prevented intensive agricultural activities and hence the exact choice of the geographical boundaries was not decisive. These authors differentiated nine "subsystems" and estimated flows individually, taking advantage of, either, studies available for the local situation, or downscaling from a U.S. dataset. While flows between the subsystems were assessed, results would not allow for an additional validation level. Inputs into the system were considered much larger than outputs, and despite of providing just one-term data (authors seemed to implicitly assume constant conditions over time, using data from the 1990's), considerable amounts of accumulation of N (for a single year, 44% of inputs in agricultural area, and 25% in urban area) have been assessed. Further studies on urban N budgets have been performed for Chinese cities. Gu et al. (2009, 2012) investigated N budgets of Hangzhou and Shanghai, respectively, both cities in the vicinity of the Yangtze river delta. Using concepts of a "greater urban area", these authors attempted to cover both the built-up city itself as well as the peri-urban agricultural belt. In the earlier work on Hangzhou (Gu et al., 2009), the concept very much drew from that on the Phoenix area, with some adaptations to the now 10 "subsystems". River systems contributed distinctively to N flows in this humid environment. Accumulation, as the difference between inputs and outputs, here was found somewhat lower at about 17.5%. Balances were calculated for two distinctive years, 1980 and 2004, with roughly a doubling of N flows in that period. For the analysis of Shanghai (Gu et al., 2012), the approach was refined in both dimensions. Largely the same subsystems (now 13) were used to create a model framework ("CHANS"), which here allowed for the development of a whole time series extending over 53 years, based on the coupling of specific information for any given year (from statistical yearbooks) and time-invariant coefficients describing the transfers. During that period, a nine-fold increase of Nr inputs was observed. However, no accumulation was identified, by contrast, throughout the whole time series outputs exceeded inputs.

The urban ecosystem of Zhengzhou (Henan province, China) was chosen as investigation area by Zhao et al. (2018). For the base year 2014, their approach investigated nine subsystems, with fossil fuel combustion related NOx emissions in industry and the "human" subsystem (including transportation) covering 60% of all N inputs. With inputs about 10% larger than outputs (but quite variable within subsystems), an accumulation was hypothesized.

Zhang et al. (2016) analyzed in detail the flows entering and leaving the city of Beijing, China, and they further pushed on improving the methodology towards applying a consistent and proven modelling framework. Exclusively focusing on inputs and outputs, these authors performed network modeling of N flows to and from the external environment and between 16 source sectors (in this approach termed "nodes"). Transfer between nodes used statistical information as well as transfer coefficients. These coefficients, compiled from available literature and presented in full detail, were maintained over time, assuming processes remained unchanged. The approach exposed significant differences and strong temporal trends over the observation period 1996–2012. It distinguished direct and integral (consisting of direct and indirect) flows, with integral flows having clearly higher values. Over the 16-year period, N flows changed markedly, and while farming and farmland played a central role in the early phase, this was largely replaced by transport as a new central node. All direct flows together increased by 24% over time (or 1.33% per year; input flows even less, 18% over the entire period – roughly 1% per year), as fertilizer input to farmland decreased massively compensating much of the increase in the transport sector. Data presented did not allow to analyze accumulation of Nr. A follow-up analysis of the Beijing situation (Zhang et al., 2018) identified structural dependencies between the network's nodes in order to devise paths of impacts, but still did not further resolve their quantities. Partly the same author team performed a factor decomposition analysis for Beijing N flows (Zhang et al., 2020) and detected increases over time of

energy and food related N, while N in fertilizer and animal feed decreased. Specifically, the increase in  $NO_x$  emissions from transport to almost 80% of energy related N input in 2015 (or about 40% of total N input into the city) represented a most decisive change. These authors demonstrated some key achievements that urban nitrogen budgets were able to provide: trends and temporal development of Nr flows over time, also relative to each other, were isolated and priorities (or change in priorities) of mitigation were identified. Factor decomposition helped trace reasons for change. The full potential of such budgets could be further extended from here, once having evaluated the potential offered by the available literature.

## 4. Discussion: From individual case studies to a generalized description of urban nitrogen flows

The individual studies discussed above, implemented for city areas and their surroundings, demonstrated the importance given and the wealth of results available, also for urban situations that were typically not characterized by the prime source of N release, agriculture. Specifically considering urban situations was deemed possible, and pinpointing the flows individually allowed to focus attention to areas of relevance and to sectors undergoing changes. The original hypothesis underlying this study, that cities are relevant entities for nitrogen pollution as well as mitigation efforts, has proven valid.

Available literature primarily focused on the unique situation of individual cities. Approaches have been developed according to the respective requirements of a specific city and possibly also a particular sector. Hence results were often not easily comparable. However, such a comparison is essential to identify common features and to validate observations.

Hence, a few of the studies devised ways that allowed at least on a limited level to find similarities. For those approaches that used the identical method for two or more base years, temporal trends were used as indicators whether or not environmental relevance of Nr was increasing. Both sectoral as well as complete budgets delivered such trends. Dynamics in some cases, specifically for East Asian cities, were so strong that, beyond a signal of the general economic development, making further conclusions became difficult. Another way of normalizing the results from different cities took advantage of the population numbers. Specific flows per capita were derived by Zhang et al. (2016; 2020) and compared to the results of other available literature. Interpretation, however, was limited due to methodological differences. Here harmonization of approaches should be able to provide additional benefits to understand and interpret results.

Normalization of results also has proven effective on an area scale. Several of the sectoral balances (see above) provided results as loss (or accumulation) per area. In agronomy, the unit of "kg N/ha" is widely used, therefore a comparison between N surplus due to irrigation water of 200 kg N/ha (Werner et al., 2019) and loss as a consequence of leaking sewage (Divers et al., 2013) became meaningful despite of describing very different sectors and activities.

It is worth noting that several approaches made use of a rather limited dataset. Quantification of an urban budget based alone on flows in and out of an urban domain required rather little information from the city itself (see e.g. Faerge et al., 2001). Such approaches left outcomes less robust, especially when it comes to comparing cities. Full input/output matrices for subsectors, as implemented in several of the urban N budgets presented, clearly was an advantage, as also flows between these sectors could be evaluated. As e.g. shown by Zhao et al. (2018), accumulation in every single sector could be derived. If, however, accumulation merely was assessed as the difference between inflows and outflows, it remained a mathematical entity. Stock changes are physically possible, both accumulation and depletion of material. Understanding difficulties in data acquistions, quantifying such material effects is deemed extremely valuable in this context, helping validating results. In a situation of outputs continuing to be larger than inputs over a full 50-year

time series (as reported by Gu et al., 2012), data consistency is at stake. It does not need measuring pool changes to pinpoint a potential data problem.

Closing the balances is a central purpose of any material flow analysis. A stock-flow approach not only allows to assess an overall N balance, but balances for each pool individually. What is true for the overall balance also counts on the level of each pool ("sector", "subsystem" or "node"): accounting for all flows, sources, sinks and stock changes in a pool should total zero. In practice, considerable variations in such accounts will provide hints to processes not so well understood. Here biophysical explanations of these processes, collecting and using parameters specific for the respective situation rather than default values will be beneficial. Also, interpretation of results from a mechanistic view, i.e. including the possible underlying mechanisms, can be very helpful. Environmental concentrations of nitrogen loads (air pollutant or water pollutant concentrations) are frequently measured and collected, and such results are available for comparison with statistical inputs and nitrogen contents. Comparison is possible, but rarely has been done in the literature investigated, and should include considerations of flows as much as of stock changes and retention of compounds in environmental media.

Filling data gaps and investigating variations in sectoral analyses can strongly benefit from interaction with stakeholders. Expertise on the respective processes, as indicated above, will be needed and that may require the input of experts beyond scientists. Including policy makers, NGOs, industry and other interest groups may provide such expertise. Using "round table discussions" – a method widely used in urban planning (Sperling, 1999 and Wiese-von Ofen, 2001) – these stakeholders may reflect and discuss the impact of an N budget along the given content, substance and range. In environmental policy and the management of protected areas, "Good Governance" also means developing solutions that involve those people that are affected. Practice proves such approaches to be useful (Cash et al., 2003; Jungmeier et al., 2019; Borrini et al., 2013, Amon et al., 2014), as stakeholders were able to take joint ownership of the results achieved.

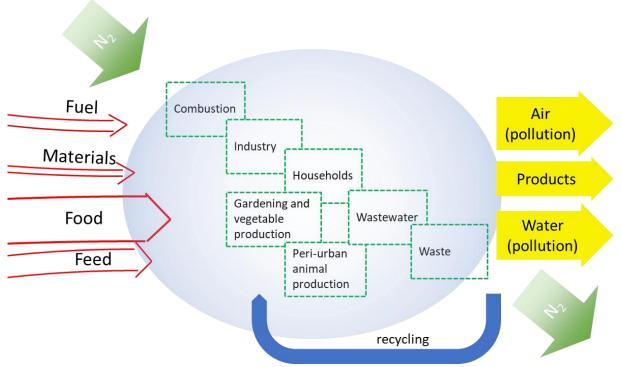


Figure 1: Concept of Urban N budget. Key N flows are denoted as arrows, open arrows are inputs, solid arrows represent outputs (products or pollutants). The rectangular boxes enclosed in dashed lines represent some important pools containing Nr in a city, which itself is here represented as the elliptical shape. Relevant for detailed evaluation are (see text) the extent of recycling, the

stratification of flows (extent to which output flows can be directly traced back to inputs), and the sub-budgets within each of the pools.

In order to further analyze urban nitrogen budgets, from the available material we noted specifically three aspects of general relevance. They may have been identified and studied previously, but not systematically or consistently and hence results are available only in part of the studies. These aspects (see Fig. 1) may be considered an essential set of parameters to be developed in such approaches. The first is the "linearity" of N throughput in cities – or, if put inversely, the recycling rate. Even with processes only partially understood, that parameter may guide to minimize N waste in cities, and to optimize their use (see Forkes, 2007). Also denitrification (as in wastewater treatment) is a way to reduce pollution, nevertheless it needs to be seen as the destruction of Nr as an otherwise valuable resource. Next, any possibility to identify stratified flows (disentangle flows that are unconnected) needs to be pursued. As evident in the analysis by Svirejeva-Hopkins et al. (2011), fuel combustion related nitrogen oxide formation in cities may be largely independent of the food chain extending from production to the waste streams. In environmental consequences, like soil acidification or even atmospheric particle formation, interactions will certainly occur, but any possible separation should be identified and investigated, as also intercomparisons may then be simplified and differentiated mitigation measures can be taken. The notion of a "nitrogen cascade" reflects such a straightforward flow pattern in urban surroundings better than it represents the interfacing web of nitrogen transformations it is usually taken for (Galloway et al., 2003). Finally, the question of accumulation of matter (in a respective pool) becomes relevant when describing the situation of Nr. Beyond the question discussed above whether stock changes in the respective pools may also indicate data problems, excess Nr or depletion of Nr both will affect environmental performance of pools and have consequences on flows. While it may be difficult to quantify pools, assessing pool changes can provide otherwise missing information, valuable when comparing between different situations (cities, years).

There is a long way from conceptual statements of intervention possibilities (see e.g. Sutton et al., 2013) to quantifiable and robust methods to implement measures that minimize the losses of nitrogen in practice on an urban level. A few relevant successful approaches have been made available in the scientific literature already in the past. A focus on urban areas seems to provide very useful and practical results. As is quite typical for N related issues, sectoral exclusive views too often take the risk of identifying one-sided solutions considering benefits for a specific sector only. This becomes evident from some of the trends identified, e.g. by the observed decrease in animal feed (Zhang et al., 2020), which just pushes the problem outside city regions. Hence, further coupling of this tool with other scales (province or country) may introduce new and necessary perspectives – implying also to consider "footprints" or effects a city may have on areas outside.

#### 5. Conclusions and way forward

With N compounds in the center of many environmental hazards, sustainable development and the achievement of the United Nations Sustainable Development Goals can be greatly advanced by improving and harmonizing the understanding of the environmental behavior of N compounds. The concept of urban nitrogen cycles to describe the environmental situation in city environments has not been used extensively in the scientific literature, but a few very relevant studies are available, which each refer to one specific city only. Such work has been focusing in part on specific sectors, but we also noticed seven cities for which full budgets are available, five of which are in Asia. The availability of these studies confirms our hypothesis of urban activities being relevant also for the N cycle. Moreover, as urban nitrogen budgets can be derived also with

incomplete data sets and complementing from budget considerations, this approach seems useful also when data access is limited.

Results found in the literature, whether sectoral or full budgets, are promising as they identify key aspects on priorities, trends and future directions of N related environmental issues, whether this is air pollution, water pollution or impacts on biodiversity. They are useful for the respective application, but do not allow for easy comparison due to differences in the system boundaries set, the sectoral pools used to classify N flows to, and the chosen methodologies. Comparibility between cities of different makeup needs standardization. Flows (and stock changes) per area or per population may serve as first proxies. More thorough development of indicators is possible from the experience presented with the selected studies. Specifically we identify the following promising approaches to characterize urban N budgets: (1) the rate of N recycling indicates the extent of N wasted, (2) in an urban situation, N flows may be stratified in part and dealt with independently to simplify balancing, and (3) pool changes and especially accumulations pointing to potential future release sites.

Taking up from international programs originally devised on a national scale (e.g. greenhouse gas inventories) and implementing existing information on the processes of N release and its environmental conversions can push further to harmonize available results. Directly comparing cities that differ in their urban fabric using a common method can be a good testing ground to develop benchmarks. Resulting information to stakeholders and to urban environmental policy will support abatement efforts for improved air and water quality, reduce climate impacts and ultimately prevent shifts of N compounds between environmental compartments.

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#### **ORCID**

Wilfried Winiwarter: 0000-0001-7131-1496 Katrin Kaltenegger: 0000-0001-7751-7794

Lin Zhang: 0000-0003-2383-8431

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