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Review article

Reactive nitrogen: A perspective on its global impact and prospects for its sustainable production

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ABSTRACT

It is necessary to review the reactive nitrogen cycle, from its generation to its emission to the environment, because it is intimately connected, as either a problem or a solution, to food security, environmental degradation, climate change and alternative energy. These connections are not often appreciated because the researchers in these disparate fields of scientific investigation do not interact extensively. This paper intends to fill that gap by presenting a broad overview and offering an important perspective on this important substance.

It is well known that the global food supply depends on anthropogenic nitrogen fertilizer and that its production and overuse has contributed to the deterioration of the environment. This paper points out that biofuels production and carbon capture schemes may also increase demand for reactive nitrogen. In addition, ammonia is being proposed as an alternative fuel. Although the exact magnitude of future requirements is uncertain, a large demand for reactive nitrogen may be inevitable and recovery from waste must be pursued. However, the recovery of reactive nitrogen from waste streams is imperfect because natural processes tend to return reactive nitrogen to the more stable state, N_2 . Thus, new technologies to replace the Haber–Bosch process must be developed. This paper reviews these new technologies and shows that almost all of the alternative means for producing reactive nitrogen are in early development. Intensified efforts to develop scalable alternatives are recommended.

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1. Introduction

The vast need for reactive or “fixed” nitrogen for the global food supply and its grave effects on the environment is well known (Eickhout et al., 2006; Galloway et al., 2003; Smil, 2001; Zhang et al., 2015). The association between population, food demand, nitrogen fertilizer and environmental degradation is so strong that the currently accepted paradigm is that practically any amount of nitrogen generation for human use is considered harmful. This is the concept behind the nitrogen “planetary boundary” (Rockström et al., 2009), which was set as 25% of the total amount of newly generated reactive nitrogen at that time. The link between reactive nitrogen and environmental damage is valid at present because 77% of anthropogenic reactive nitrogen consumption in 2015 was for fertilizer (FAO, 2017). Indeed, subsequent refinements of the planetary boundary for nitrogen derive the boundary from fertilizer requirements (De Vries et al., 2013). However, the use of reactive nitrogen has moved beyond the traditional use for food agriculture. The production of biofuels has increased agricultural activity and reactive nitrogen production, with the consequent increase in environmental damage (Erismann et al., 2010). In addition to the increased agricultural activity, reactive nitrogen in the form of ammonia is used for pretreatment of lignocellulosic feedstock prior to fermentation to ethanol also requires ammonia (Tao et al., 2011). Reactive nitrogen is also needed for biotic carbon capture schemes (Coskun et al., 2016). Carbon capture and storage by chemical absorption also requires ammonia (Shakerian et al., 2015). Lastly, ammonia is being proposed to be used directly as an alternative fuel (Zamfirescu and Dincer, 2008)[p]. The nature and prospects for these new applications be discussed more extensively later in the paper,

The reactive nitrogen cycle at present and, prospectively, in the future are diagrammed in Fig. 1. Current roles are shown as solid lines whereas roles that may come in the future are shown as dotted lines. It is important to note that not all of these new applications may be scaled-up to commercial levels in the future. Some applications have a higher probability of widespread adoption than the others but the appreciation of the possible scale of these new applications is vital to decision-making on whether new production technologies are worth pursuing further.

While much of this knowledge about the need for reactive nitrogen is common knowledge within each specialized field of inquiry, the connection between each one is not common knowledge. To acquaint researchers of developments outside their specialized fields of inquiry, I point out these connections and indicate possible directions that the relationship between reactive nitrogen, the environment and energy and food security may take. To do this, the following are reviewed: (a) the impact of nitrogen fixation on the environment and global resources in Section 2; (b) the role of reactive nitrogen on food security, alternative energy and carbon sequestration and how this translates to future demand in Section 3; (c) the prospects for reducing the use of reactive nitrogen in Section 4 and (d) possible new sources of synthetic reactive nitrogen in Section 5. In doing so, we gain the insight that reduction of reactive nitrogen usage is not a realistic option in the next few decades and, therefore, alternatives to the Haber–Bosch process need to be developed.

This paper is an updated and more comprehensive version of a previous Perspective paper (Razon, 2014a). Because of the breadth of the scope, the paper will largely cite review papers. Specific selection criteria for inclusion of other papers are explained in individual sections as needed.

2. The impact of reactive nitrogen on the environment

In this section, the focus is on recent review papers on the environmental impact of reactive nitrogen and its extent. Other papers and reports that have made key insights are also included.

Since the Industrial Revolution, there has been an immense increase in the amount of nitrogen fixation in the world. Fowler et al. (2013) have done an accounting for all sources of reactive nitrogen and estimate that the total amount fixed annually is about 413×10^9 kg. Out of this total, non-anthropogenic sources like lightning and diazotrophic organisms account for about 203×10^9 kg. Fowler et al. (2013) also estimate that human activity is presently responsible for roughly an equal amount as that from natural sources. The agriculture of nitrogen-fixing crops like peanut and soybean account for 60×10^9 kg a year. Unintentional anthropogenic nitrogen fixation comes from the combustion of fuels, which produces nitrogen oxides (NO_x). This is estimated at 30×10^9 kg per year. The intentional production of reactive nitrogen is primarily via the Haber–Bosch process for ammonia, which accounts for about 120×10^9 kg per year.

The impact of the overuse and misuse of reactive nitrogen has been very well studied and reviewed (Erismann et al., 2013). The reactive nitrogen cycle is complex. Once the reactive nitrogen molecule is created, it cascades through the environment creating a variety of effects on a wide range of distance- and time-scales. Short-distance effects include the direct inhalation of nitrogen oxides (NO_x) and NH_3 , which causes a variety of health effects. In addition, NO_x increases particulate matter and induces the formation of photochemical smog and tropospheric ozone. Reactive nitrogen also has longer-range, regional-level polluting effects on aquatic ecosystems through eutrophication and acidification. The longest-range impact of reactive nitrogen is its impact on global climate change but there are complex, counterbalancing interactive effects that seem to cancel each other out. Erismann et al. (2011) have reviewed estimates of the influence of reactive nitrogen on climate radiative forcing and arrived at the figure of -0.24 Wm^{-2} . This suggests that there may be a net cooling effect of reactive nitrogen on global climate change but the range is very large and goes from positive ($+0.2 \text{ Wm}^{-2}$) to negative (-0.5 Wm^{-2}). There is thus some uncertainty in the overall impact of reactive nitrogen on the global climate.

The environmental impact of reactive nitrogen comes not just from its use but also from its commercial production. Fig. 2 shows a simple representation of the Haber–Bosch process as commonly practiced at a commercial scale. Note that the primary raw materials are natural gas and air. CO_2 is the primary direct emission. While the Haber–Bosch process was originally proclaimed to produce “bread from air” (Smil, 1997), in reality, a considerable amount of fossil fuels is consumed during the production of hydrogen. Energy consumption occurs at all stages but it is worth pointing out the very high pressures and temperatures at which the conversion process occurs. Considerable amounts of energy are also spent to purify the feed stream in order to protect the catalyst (Appl, 1999). It has previously been stated that about 1.2% of the world primary energy is consumed for the production of fertilizer (IFA, 2009a), of which 94% is for the production of ammonia. Because of the inherent chemistry of the steam reforming of natural gas or coal, the industrial-scale Haber–Bosch process is also associated with large releases of CO_2 , regardless of the energy source for other activities. An estimate for 2007 stated that fertilizer production

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