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Atmospheric Nitrogen Oxides Emissions: Major Environmental Challenges of the 21st Century

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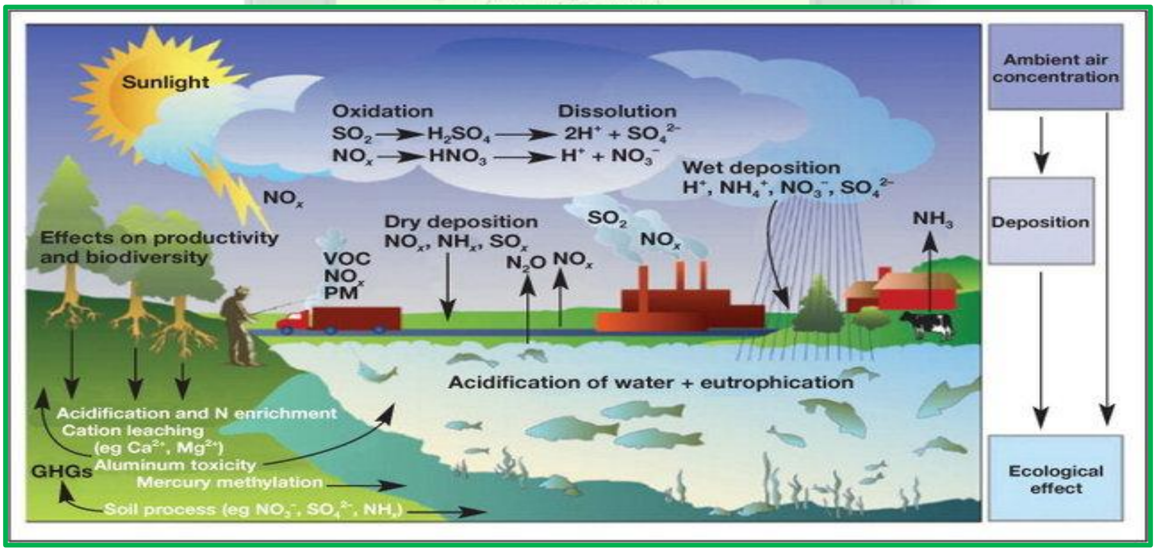
Reactive nitrogen compounds in the atmosphere are defined as nitrogen compounds that are chemically reactive, biologically active, or radioactively active by absorbing infrared radiation or other radiation. These compounds contrast with nitrogen gas (N_2), which is non-reactive. Reactive nitrogen compounds include chemically-oxidized inorganic nitrogen such as oxides of nitrogen (NO_X), nitric acid (HNO_3), the nitrate radical and ion (NO_3 and NO_3^-), and nitrous oxide (N_2O); chemically-reduced inorganic nitrogen such as ammonia (NH_3) and ionic ammonium (NH_4^+); and organic nitrogen such as urea, amino acids, and proteins. Increased reactive nitrogen emissions annually increased costs associated with environmental management, economic losses, substantial health risks for vulnerable human populations, etc. Reactive nitrogen loss to the environment is one of the major environmental challenges of the 21st century impacting climate change; energy and food security; air, water, and soil quality; and human health.

Effect of NO_X Emissions on Ecosystem

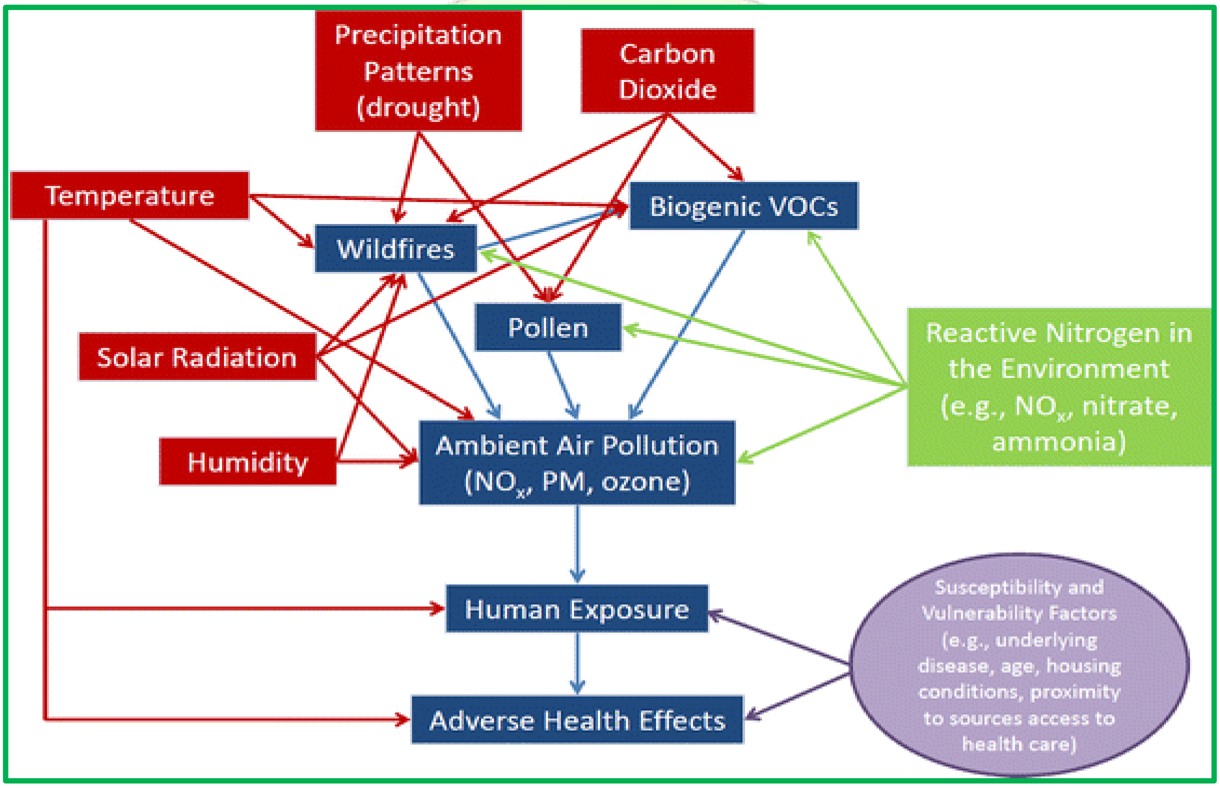
NO_X emissions contribute to many air pollution problems, including smog, tropospheric O_3, acid rain, and elevated levels of fine particulate matter (PM2.5). NO_X comprises nitric oxide (NO) and nitrogen dioxide (NO_2), which are easily interconverted. These are important trace constituents in the troposphere, where they regulate the production and consumption of photochemical oxidants, ozone (O_3), and hydroxyl radicals. Tropospheric O_3 is a significant air pollution problem in the United States, as well as in most developed and developing countries.
It is harmful to both human health and welfare. NO_X is an important precursor to tropospheric ozone. NO_2 reacts in air to produce NO and O_3. When the NO concentration is below 3-8 ppt, NO reacts with



O_3 to produce NO_2 and O_2 thus consuming O3. But when the NO concentration is higher, NO catalyses the oxidation of CH_4, CO, and volatile organic compounds to produce O_3 NO is recycled to NO_2 by free radicals. In rural environments, the reaction of NO with biogenic can be a predominant source of ozone. NO_X in the troposphere can be oxidized and react with water to form nitric acid (HNO_3). This contributes to acid rain, which directly accelerates acidification and eutrophication processes in regional ecosystems

Source of NO_X Emissions

NO_X emissions stem from both anthropogenic and natural sources. Nitrogen gas and oxygen combine to form NO_X in lightning and during combustion processes. Microbes in the soil also produce NO as they metabolize nitrogen compounds, which may be present naturally in the soil, or enhanced by nitrogen fertilizers. Fossil fuel combustion is the largest source of NO_X emissions, contributing more than half of the global NO budget. Soils, especially agricultural soils, are an important source of biogenic NO emissions. NO_X emissions from large combustion sources and industrial sources are regulated in the U.S. and many other countries under air pollution programs designed to ameliorate tropospheric O_3, acid rain, PM2.5, and regional haze. However, emissions of NO from agriculture are unregulated.



Factors Affecting the NO Emission Rate

Many factors affect the NO emission from soil. The availability of nitrogen compounds in the soil is a key factor affecting the NO emission rate. This soil nitrogen can be derived from various sources, including nitrogen-fixing bacteria in the soil, deposition of NH_3, NO_X, acid rain, or other nitrogen compounds from the atmosphere, decay of organic material, or inputs of synthetic nitrogen fertilizer and manure. In agricultural soils, the primary sources of nitrogen are synthetic fertilizers, manure, and nitrogen-fixing crops such as soybeans. In addition to the availability of nitrogen, the NO emission rate is affected by environmental factors, including soil temperature, soil pH, and soil moisture. Soil water content controls the rate of O_2 supply, which directly affects nitrification and denitrification. NO emission decreased after precipitation but increased during drought. In addition, due to the positive effect of soil temperature on microbial processes, NO emission generally increases with soil temperature. There is no direct relationship between the rate of NO emission and soil pH; however, microbial nitrification processes, which convert chemically reduced nitrogen compounds (such as

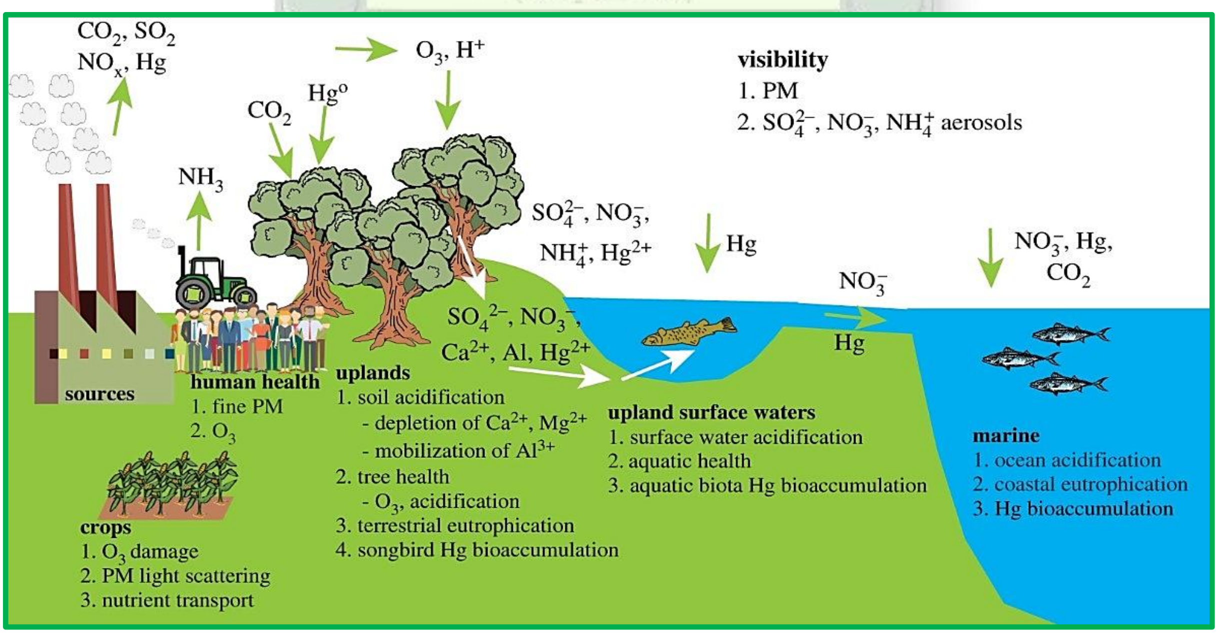
NH_4^+) to chemically oxidized compounds (NO_3^-), are enhanced at higher soil pH. In addition to other oxidized nitrogen compounds, nitrification processes produce some NO, which can be emitted into the atmosphere.

Global and regional estimates for NO emissions are subject to considerable uncertainty, and emissions estimates cover a wide range between the lowest and highest values. Most measurements are short-term and inherently do not represent the spatial and temporal variation of NO emission. Accurate estimates of NO emissions are necessary for global inventories. Global and regional estimates are also important for developing better models to assess the impact of NO emission on the atmosphere and the deposition of reactive nitrogen in terrestrial ecosystems. The emissions factor approach is generally used to estimate local, regional, and global NO and other species emissions. In this approach, emission factors are computed in terms of the mass of NO emissions per mass of nitrogen applied in fertilizer, for a variety of nitrogen fertilizers, and manure. To compute NO emissions, these emission factors are multiplied by the amount of nitrogen-based fertilizer applied over agricultural regions.

Health and Environmental Effects

There is compelling evidence that NO x respiratory contact can cause and aggravate asthma symptoms, and may even contribute to the development of asthma over time. It has also been linked to heart disease, diabetes, birth outcomes, and all-cause mortality, however, the evidence for these non-respiratory consequences is less clear. Nitric acid vapour and associated particles are formed when NO x combines with ammonia, moisture, and other substances. In the presence of sunshine, NO x combines with volatile organic molecules to generate ozone. Ozone can have negative consequences such as lung tissue destruction and a loss in lung function, especially in vulnerable populations (children, elderly, asthmatics). Air currents can transfer ozone and induce health effects far from the sources. According to the American Lung Association, approximately half of the people in the United States reside in regions that are not in ozone conformity. Ground-level ozone pollution is strongest in the countryside and suburbs in South East England, whereas NO emissions can "mop up" ozone to generate NO_2 and oxygen in downtown London and on main roadways. NO x also rapidly interacts with ordinary organic molecules and even ozone to produce a broad range of hazardous products, including nitroarenes, nitrosamines, and the nitrate radical, some of which can induce DNA alterations.

Another route from NO x to ozone has been discovered, which occurs mostly in coastal locations due to the creation of nitryl chloride when NO x comes into contact with salt-mist. The direct effect of NO x emissions contributes positively to the greenhouse effect. In Reaction 3, instead of interacting with ozone, NO can potentially react with HO2 and organic peroxy radicals (RO2), increase in the concentration of ozone. When the concentration of NO x surpasses a particular threshold, atmospheric processes result in the creation of net ozone. Because tropospheric ozone may selectively absorb radiation, NO x's indirect effect is exacerbating global warming.



Technologies for Emission Control and Regulation

SCR and SNCR decrease post-combustion NO_x by reacting exhaust with urea or ammonia to create nitrogen and water. SCR is presently utilized in ships, diesel vehicles, and certain diesel automobiles. The use of exhaust gas recirculation and catalytic converters in automobile engines has resulted in considerable reductions in vehicular emissions. The major emphasis of the Volkswagen emissions issues was NO_x.

Other methods, like flameless oxidation (FLOX) and staged combustion, decrease thermal NO_x greatly in industrial operations. Bowin low NO_x technology is a blend of staged-premixed-radiant combustion technology that includes a major surface burning followed by a modest radiant burning. The Bowin burner premixes air and fuel gas in a ratio larger than or equal to the stoichiometric burning demand. Water injection technology, which introduces water into the combustion chamber, is also becoming a key technique for reducing NO_x by increasing total combustion efficiency. Furthermore, water (often 10 to 50%) is emulsified into the fuel oil before injection and burning. This emulsification can be done throughout (unstabilized) right before insertion, or as a drop-in fuel with added chemicals for long-term emulsification (stabilized).