Global impact of fossil fuel combustion on atmospheric NOx

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Abstract

Fossil fuel combustion is the largest global source of NO_x to the troposphere. This source is concentrated in polluted boundary layers, and the extent to which it impacts tropospheric chemistry on a global scale is uncertain. We use a global three-dimensional model of tropospheric chemistry and transport to study the impact of fossil fuel NO_x emissions on the global distribution of NO_x during northern hemisphere summer. In the model, we tag fossil fuel NO_x and the reservoir NO_y species in order to determine the relative contribution of fossil fuel combustion to NO_x concentrations in different regions of the world. Our model includes a detailed representation of NO_x-O_3-NMHC chemistry, which is necessary to properly simulate the export of reactive nitrogen, including organic nitrates such as peroxyacyl nitrates (PANs) from the continental boundary layer. We find that fossil fuel combustion accounts for over 40% of NO_x concentrations in the lower and middle troposphere throughout the extratropical northern hemisphere. PANs are shown to provide an important mechanism for transporting NO_x from source regions to the remote troposphere, accounting for over 80% of the fossil fuel NO_x in the lower troposphere over most of the ocean. Sources in the United States are found to contribute about half of the fossil fuel NO_x over the North Atlantic Ocean. Emissions from China, which are expected to increase rapidly in the coming decades, currently account for about half of the fossil fuel NO_x over the western North Pacific Ocean; the influence of these emissions extends into the tropics. Because of this tropical influence, emissions from China have more potential than emissions from the United States to perturb the global oxidizing power of the atmosphere.

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