

## 2.141 The significant role of cloud chemistry in tropospheric NO<sub>x</sub> and oxidant cycles.

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Abstract:

Aqueous cloud chemistry is generally thought to play a minor role in the tropospheric NO<sub>x</sub> cycle, in contrast to aerosol, which is known to be an important NO<sub>x</sub> sink through the hydrolysis of N<sub>2</sub>O<sub>5</sub> and NO<sub>3</sub>. This consensus view seems to originate with model studies in the 1990s reporting that most N<sub>2</sub>O<sub>5</sub> is already consumed by ubiquitous aerosol so clouds have little additional effect. However, those studies assumed reactive uptake coefficients (γ) for N<sub>2</sub>O<sub>5</sub> and NO<sub>3</sub> on tropospheric aerosol that we now know are much too high; they therefore overestimated the importance of aerosol. We reassess the role of cloud droplets as sites for heterogeneous NO<sub>x</sub> loss and its effect on tropospheric O<sub>3</sub> and OH using a global chemical transport model. Simulating heterogeneous cloud reactions in regional and global models that do not resolve individual clouds requires accounting for cloud entrainment in addition to diffusion and reactive uptake constraints on chemical rates. We develop a simple and fast mathematical framework for doing this and show that other commonly used approaches generate large simulation errors. In the improved model, global NO<sub>x</sub> loss through hydrolysis in clouds is about half of that hydrolyzed on non-cloud aerosol and 7% of global tropospheric NO<sub>x</sub> loss. As a result, addition of hydrolysis on clouds lowers simulated tropospheric O<sub>3</sub> by 2.8% (0.5 ppb), OH by 3.3%, and increases the CH<sub>4</sub> lifetime by 3.1%. Despite the greater loss of N<sub>2</sub>O<sub>5</sub> on aerosols, clouds and aerosol have similar impact on global tropospheric O<sub>3</sub> and OH, because clouds have greater influence on the tropics, where O<sub>3</sub> production is NO<sub>x</sub>-limited. The old paradigm of neglecting cloud effects on NO<sub>x</sub> chemistry should be discarded and this work provides efficient numerical methods to treat cloud chemistry in regional and global models.