Climatologies of NOx and NOy: a comparison of data and models

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Abstract

Climatologies of tropospheric NOx (NO+NO2) and NOy (total reactive nitrogen: NOx + NO3+ 2xN2O5 + HNO2 + HNO3 + HNO4 + ClONO2 + PAN (preroxyacetylnitrate) + other organic nitrates) have been compiled from data previously published and, in most cases, publicly archived. Emphasis has been on non-urban measurements, including rural and remote ground sites, as well as aircraft data, Although the distribution of data is sparse, a compilation in this manner can begin to provide an understanding of the spatial and temporal distributions of these reactive nitrogen spoecies. The cleanest measurements in the boundary layer are in Alaska, northern Canada, and the eastern Pacific, with median NO mixing ratios below 10 pptv, NOx below 50 pptv, and NOy below 300 pptv. The highest NO values (greater than 1 ppbv) were found in eastern North America and Europe, with correspondingly high NOy (5 ppbv). A significantly narrower range of concentrations is seen in the free troposphere, particularly at 3-6 km, with NO typically at 10 pptv in the boreal summer. NO increases with altitude to ~100 pptv at 9-12 km, whereas NOy does not show a trend with altitude, but varies between 100 and 1000 pptv. Decreasing mixing ratios eastward of the Asian and North American continents are seen in all three species at all altitudes. Model-generated climatologies of NOx and NOy from six chemical transport models are also presented and are compared with observations in the boundary layer and the middle troposphere for summer and winter. These comparisons test our understanding of the chemical and transport processes responsible for these species distributions. Although the model results show differences between them, and disagreement with observations, none are systematically different for all seasons and latitudes. Many of the differences between the observations and model results may likely be attributed to the specific meteorological conditions at the time that measurements were made differing from the model meteorology, which is either the climatological flow from GCMs or a several year average of actual meteorology. Particular difficulty lies in the comparison of ground measurements, which are influenced by local sources, which will be smoothed, or possibly ignored, by the models.