A Global 3-D Model Evaluation of the Atmospheric Budgets of HCN and CH_3CN: Constraints From Aircraft Measurements Over the Western Pacific

Qinbin Li, Daniel J. Jacob, Robert M. Yantosca, Colette L. Heald, Hanwant B. Singh, Makoto Koike, Yongjing Zhao, Glen W. Sachse, and David G. Streets

J. Geophys. Res., 108(D21), 8827, doi:10.1029/2002JD003075, 2003.

Abstract

We construct global atmospheric budgets of HCN and CH_3CN through a 3-D simulation of the HCN-CH_3CN-CO system constrained and evaluated with aircraft observations from the TRACE-P mission over the NW Pacific in February-April 2001. Observed background vertical gradients of HCN and CH_3CN imply a dominant ocean sink for both gases, with deposition velocity of 0.13 cm s^{-1} for both and saturation ratios of 0.79 for HCN and 0.88 for CH_3CN. Observations for both gases in the free troposphere implied a dominant source from biomass burning. Enhancement of HCN observed in Chinese urban plumes is attributed tentatively to residential coal burning. Biomass burning and residential coal burning emission ratios relative to CO of 0.27% and 1.6% respectively for HCN, and of 0.20% and 0.25% respectively for CH_3CN, are consistent with observations in biomass burning and Chinese urban plumes, and provide the best fit in the model for simulation of observed TRACE-P vertical profiles, HCN-CH_3CN-CO correlations, as well as long-term records of HCN columns and CH_3CN observations over the northern Indian Ocean. Biomass burning and residential coal burning contribute 0.63 and 0.2 Tg N yr^{-1} respectively to global HCN and 0.47 and 0.03 Tg N yr^{-1} respectively to CH_3CN. Ocean uptake is the dominant sink for both gases, with oxidation by OH representing an additional minor sink. The resulting tropospheric lifetimes are 5.3 months for HCN and 5.8 months for CH_3CN. The model predicts very low HCN and CH_3CN concentrations at high southern latitudes, reflecting the assumption of uniform saturation ratio; observations in that region are needed. In the free troposphere, the dominance of biomass burning sources (70-85% for HCN and 90-95% for CH_3CN) implies that both gases can be used as biomass burning tracers. More work is needed to identify the urban source apparent in the Chinese plume observations.

The full version of this paper is available as a pdf file [Full Text (pdf)].