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What drives the observed variability of HCN in the troposphere and lower stratosphere?

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Abstract. We use the GEOS-Chem global 3-D chemistry transport model to investigate the relative importance of chemical and physical processes that determine observed variability of hydrogen cyanide (HCN) in the troposphere and lower stratosphere. Consequently, we reconcile ground-based FTIR column measurements of HCN, which show annual and semi-annual variations, with recent space-borne measurements of HCN mixing ratio in the tropical lower stratosphere, which show a large two-year variation. We find that the observed column variability over the ground-based stations is determined by a superposition of HCN from several regional burning sources, with GEOS-Chem reproducing these column data with a positive bias of 5%. GEOS-Chem reproduces the observed HCN mixing ratio from the Microwave Limb Sounder and the Atmospheric Chemistry Experiment satellite instruments with a mean negative bias of 20%, and the observed HCN variability with a mean negative bias of 7%. We show that tropical biomass burning emissions explain most of the observed HCN variations in the upper troposphere and lower stratosphere (UTLS), with the remainder due to atmospheric transport and HCN chemistry. In the mid and upper stratosphere, atmospheric dynamics progressively exerts more influence on HCN variations. The extent of temporal overlap between African and other continental burning seasons is key in establishing the apparent biennial cycle in the UTLS. Similar analysis of other, shorter-lived trace gases have not observed the transition between annual and biennial cycles in the UTLS probably because the signal of inter-annual variations from surface emission has been diluted before arriving at the lower stratosphere (LS), due to shorter atmospheric lifetimes.

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