The relative roles of dynamics and emissions in determining atmospheric HCN
- reconciling ground-based FTIR and space-borne data by using a 3-D CTM

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1. Introduction

We used the GEOS-Chem global 3-D chemical transport model (CTM) to interpret the tropical atmospheric distributions of HCN observed by NASA Aura Microwave Limb Sounder (MLS) instrument. The main sources of hydrogen cyanide (HCN) are from the biomass burning (BB) and domestic fossil fuel burning while the main sink is ocean uptake, resulting in a tropospheric lifetime of > 5 months. This lifetime is sufficiently long to let the variability in HCN introduced by surface emissions be observed in the tropical upper troposphere and stratosphere. The recent study of MLS data has reported a 2-year cycle in the HCN concentrations in the stratosphere which was partly attributed to the inter-annual variabilities of biomass burning Australia and Indonesia. On the other hand, the intermittent tropospheric HCN column data from ground-based instruments across the tropics show an annual cycle. In this study, we used the model to reconcile the discrepancy between the space-borne and ground-based observations of HCN over the tropics. By comparing the observations and the CTM simulations, the roles of dynamics and surface emissions determining atmospheric HCN will be investigated.

2. GEOS-Chem simulation of HCN

Description

- 3-D global CTM driven by assimilated meteorology from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office
- 2x2.5 degree horizontal resolution and 30 vertical levels up to 80km
- HCN in troposphere: BB and domestic fossil fuel burning sources, ocean uptake sink
- HCN in stratosphere: CH_3CN+OH source, HCN+OH sink
- Monthly mean BB emission from Global Fire Emissions Database Version 2
- Monthly mean OH fields from GEOS-Chem (troposphere) and Aura MLS (stratosphere)

Model Evaluation with ground-based FTIR measurements

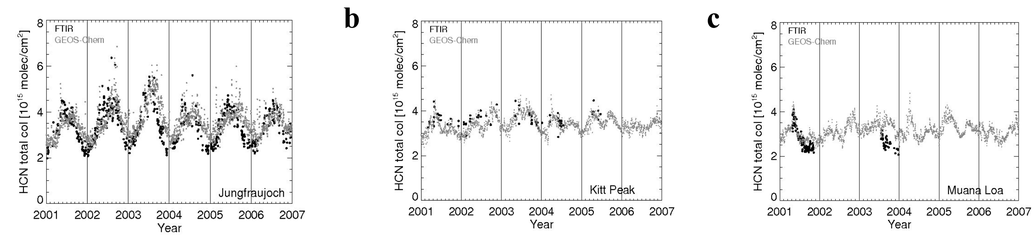


Fig.1 HCN total columns observed by ground-based FTIR (black dots) and simulated by GEOS-Chem (grey dots). a) Jungfraujoch, Switzerland (46.6N, 8.0E). b) Kitt Peak, Arizona (31.9N, 116W); and c) Mauna Loa, Hawaii (19.5N, 155.6W).

3. Aura MLS observation vs GEOS-Chem simulation

Tape recorder – tropical anomalies of HCN

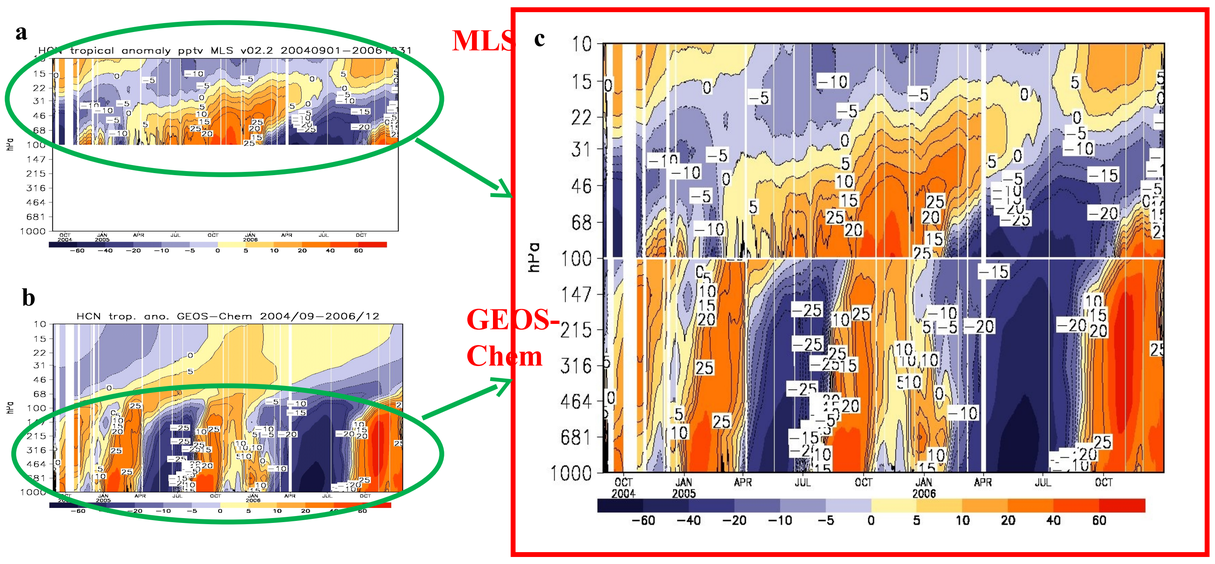


Fig.2 Timeseries of tropical anomalies (2004-09 – 2006-12) of zonal mean of HCN volume mixing ratio (pptv) on meridional plane from; a) Aura MLS observati; b) GEOS-Chem simulation; and c) match of MLS (100-10hPa) and GEOS-Chem (1000-100hPa).

GEOS-Chem long term run

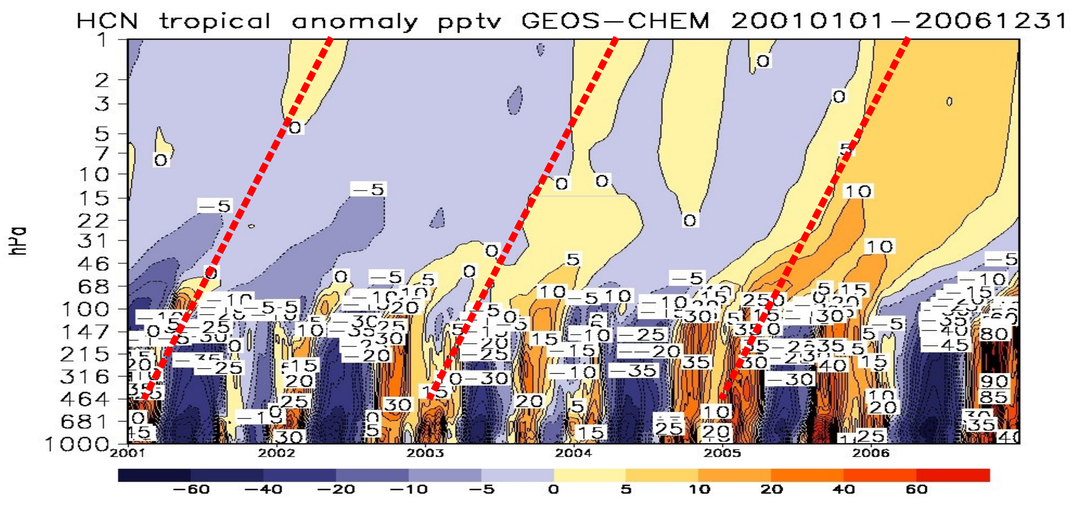


Fig.3 Time series of tropical anomalies of HCN mixing ratio from GEOS-Chem long term (2001 – 2006) run..

4. Roles of surface biomass burning (BB) emissions

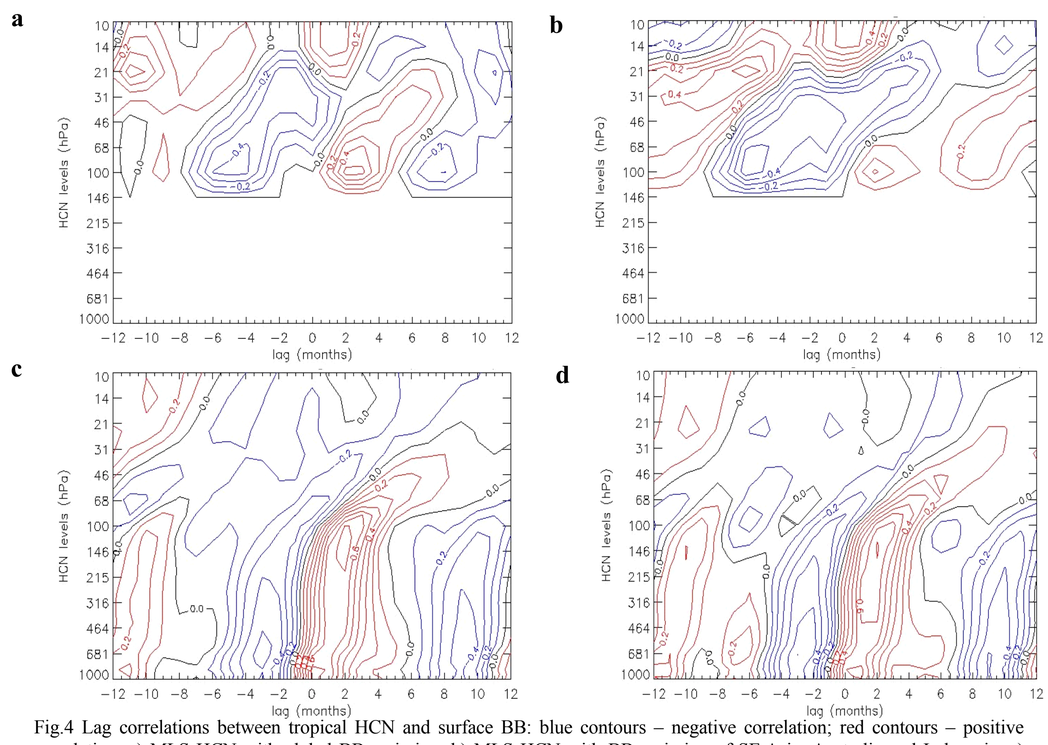


Fig.4 Lag correlations between tropical HCN and surface BB: blue contours – negative correlation; red contours – positive correlation. a) MLS HCN with global BB emission. b) MLS HCN with BB emission of SE.Asia, Australia and Indonesia. c) GEOS-Chem HCN with global BB emission. d) GEOS-Chem HCN of long term run (2001-2006) with global BB emission.

5. Roles of Quasi-Biennial Oscillation (QBO) of tropical stratospheric zonal wind

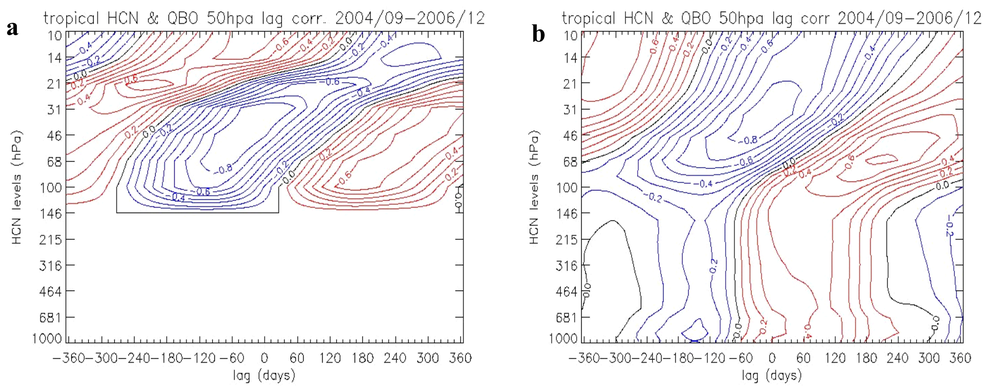
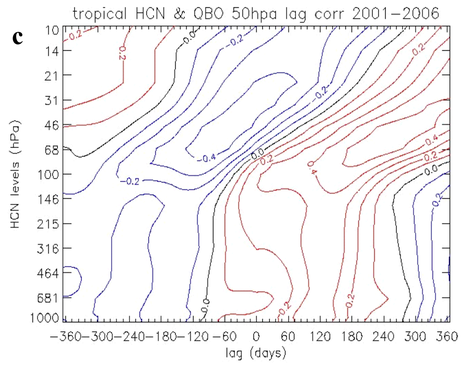


Fig.5 Lag correlations between tropical HCN mixing ratio and the 50hPa QBO index. Color setting same as Fig.4. a) MLS HCN with QBO index. b) GEOS-Chem HCN with QBO index. Timeseries of tropical anomalies of zonal mean HCN vmr (pptv) observed by Aura MLS (2004 – 2006): tape recorder. (white columns represent the gap of data).



6. Conclusions

- **The ground-based FTIR observations show the annual variability of HCN while the satellite Aura MLS data show the 2-year cycle in tropical stratosphere**
- **Simulation of GEOS-Chem shows that the annual cycle and the 2-year cycle in the lower stratosphere are consistent with each other over the Stratosphere-Troposphere Exchange (STE)**
- **The long lifetime of HCN in the stratosphere combined variations of biomass burning induced the 2-year tape recorder observed by MLS**
- **The correlation analysis shows that the variations of surface biomass burning emissions mainly impact on the tropospheric HCN**
- **The high correlations in the stratosphere indicate that the QBO plays an important role on the 2-year cycle of stratospheric HCN**

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