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Asian monsoon transport of pollution to the stratosphere

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Transport of air from the troposphere to the stratosphere occurs primarily in the tropics, associated with the ascending branch of the Brewer-Dobson circulation. Here we identify the transport of air masses from the surface, through the Asian monsoon, and deep into the stratosphere, using satellite observations of hydrogen cyanide (HCN), a tropospheric pollutant produced in biomass burning. A key factor in this identification is that HCN has a strong sink from contact with the ocean; much of the air in the tropical upper troposphere is relatively depleted in HCN, and hence broad tropical upwelling cannot be the main source for the stratosphere. The monsoon circulation provides an effective pathway for pollution from Asia, India and Indonesia to enter the global stratosphere.

The Asian summer monsoon circulation contains a strong anticyclonic vortex in the upper troposphere and lower stratosphere (UTLS), spanning Asia to the Middle East. The anticyclone is a region of persistent enhanced pollution in the upper troposphere during boreal summer, linked to rapid vertical transport of surface air from Asia, India and Indonesia in deep convection, and confinement by the strong anticyclonic circulation (1-6). A mean upward circulation on the eastern side of the anticyclone extends the transport into the lower stratosphere, as evidenced by satellite observations of water vapor (7) and ozone (8), plus carbon monoxide and other pollution tracers (1,4,5). Model calculations have suggested that transport from the monsoon region could contribute significantly to the budget of stratospheric water vapor (8,9), but this effect has not been isolated from broader-scale tropical upwelling in observational data.

Hydrogen cyanide (HCN) is produced primarily as a result of biomass and biofuel burning, and is often used as a tracer of pollution originating from fires (10-12). In the free atmosphere HCN has a long photochemical lifetime of over 4 years (12,13), but it has a strong sink resulting from contact with the ocean surface (11,12). In the tropics this behavior results in relatively low values of HCN in the troposphere apart from seasons with local biomass burning (10,14). Global satellite observations of HCN in the upper troposphere from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) satellite instrument (15,16) (Fig. 1a) reveals the signature of air depleted in HCN over the tropical oceans, together with enhanced values isolated within the Asian monsoon anticyclone during boreal summer (June-August). The tropical minimum for HCN is a distinctive signature which is very different from most other tropospheric pollution tracers, such as carbon monoxide (17). The overall structure of HCN is accurately simulated by a three-dimensional chemical transport model (Fig. 1b),

incorporating HCN sources from wildfires and biofuel combustion, plus an imposed sink from contact with the ocean surface (18). The realistic structure in this simulation suggests a reasonable understanding of the processes leading to the observed global-scale HCN behavior, especially the role of the oceanic regions in depleting HCN, and the Asian monsoon circulation in transporting HCN from the surface to the upper troposphere.

The relative minimum in HCN over the tropical Pacific ocean is a feature that is observed throughout the year (Fig. S1). In addition to the maximum associated with the Asian monsoon during boreal summer, seasonally-varying sources of HCN include burning over Indonesia and Africa during boreal spring (March-May), and burning over Africa and South America during austral spring (September-November), with these emissions transported to the upper troposphere by deep convection. However, the upper tropospheric circulation associated with the Asian summer monsoon is more coherent and vigorous than the monsoonal circulations in these other regions and seasons, with a vertical extent that reaches across the tropopause into the lower stratosphere. A longitudinally averaged cross section of the satellite measurements during boreal summer (Fig. 2) shows high HCN values throughout the extratropical Northern Hemisphere, extending across the tropopause into the lower stratosphere; the pronounced cross-tropopause maximum near 30° N is associated with the monsoon anticyclone shown in Fig. 1. The high HCN values in the stratosphere extend to low latitudes and vertically over the equator, and are transported into the middle stratosphere in the upward Brewer-Dobson circulation, within the so-called tropical pipe (19). The enhanced summer HCN values are observed to persist in the Northern Hemisphere lower stratosphere through the following seasons (Fig. S2).

Further evidence of the Asian monsoon-stratosphere coupling comes from examining interannual variations of HCN in the satellite record. Measurements of HCN from the Aura

Microwave Limb Sounder (MLS) satellite instrument (20) complement the ACE-FTS observations, providing continuous space-time coverage for ~7 km thick layers covering the lower to middle stratosphere (but not below the tropopause). Time series of the MLS data in the lower stratosphere (~16-23 km) from late 2004 to the end of 2009 (Fig. 3) show HCN maxima in the NH subtropics during each boreal summer (~June-October); this is a clear fingerprint of the Asian monsoon influence (consistent with the ACE-FTS observations in Figs. 2 and S2). In this short time record, the HCN maxima extend most strongly to near-equatorial latitudes during 2005 and 2007, and less-so in the other years. Previous analyses (21) have demonstrated that these 2005 and 2007 equatorial HCN maxima propagate coherently upwards into the stratosphere with the tropical Brewer-Dobson circulation; this so-called ‘tape-recorder’ effect is evident in other stratospheric trace constituents (e.g. H₂O) which have seasonal or interannual anomalies originating near the tropical tropopause (22). Figure 3 demonstrates that these stratospheric anomalies are linked to enhanced boreal summer (Asian monsoon) maxima during 2005 and 2007. Figure 3 also shown isolated HCN maxima in the Southern Hemisphere subtropics (~0° – 20° S) during late 2004 and late 2006, which result from enhanced austral spring burning over Indonesia during these years (23). However, direct transport to the stratosphere from these episodes appears smaller than the boreal summer sources linked to the Asian monsoon.

The exact causes of the enhanced tropical lower stratospheric HCN during 2005 and 2007 seen in Fig. 3 are difficult to determine from the limited sampling of the satellite observations. We have searched for systematic changes in transport or circulation of the Asian monsoon anticyclone during these years (or links to the stratospheric quasi-biennial oscillation, QBO), but do not find obvious links to the enhanced HCN anomalies. Rather, it is likely that these patterns reflect variations in tropospheric sources, subsequently transported through the monsoon

circulation; we note that the detailed attribution of such tropospheric sources is difficult based on the sparsely sampled ACE-FTS measurements. Recent model simulations of global HCN variability (24) suggest enhanced sources linked to the Indonesian fires in late 2004 and 2006, and the persistence into the following years and entrainment into the Asian monsoon circulation is reasonable given the long HCN photochemical lifetime in the free atmosphere.

These HCN observations demonstrate a large discernable chemical influence on the stratosphere from the Asian monsoon circulation. This pathway complements the large-scale troposphere to stratosphere transport that occurs in the deep tropics throughout the year (25), and there are likely distinct source regions for air within each pathway. Upwelling over the deep tropics primarily transports air with recent contact with the ocean surface, and less concentrated anthropogenic influences. In contrast, transport in the monsoon region connects surface air with enhanced pollution (biomass and biofuel burning, plus urban and industrial emissions) to the lower stratosphere. Model calculations (6) suggest that surface emissions over a broad region covering India to eastern Asia are entrained into the monsoon circulation and transported to the lower stratosphere. This air will have enhanced black and organic carbon, sulfur dioxide (SO₂), reactive nitrogen species (NO_x), and possibly short-lived halogen compounds from Asian industrial emissions, which have the potential to influence stratospheric ozone chemistry, aerosol behavior and associated radiative balances. For example, a recent increase in background stratospheric aerosol concentrations has been observed, possibly linked to growth in sulfur dioxide emissions over China since 2002 (26), and the monsoon is an effective pathway for such transport. The monsoon influence on the stratosphere is expected to become increasingly important given the ongoing growth of Asian emissions (27), with large continued increases over the next decades expected for SO₂ and NO_x. Furthermore, potential changes in the strength and

variability of the Asian monsoon circulation in an evolving climate (linked to increased convection and rainfall (28)) could modify this transport pathway, with potential influence on composition and climate of the stratosphere.

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18. Figure 1b shows results from the NCAR 3D Whole Atmosphere Community Climate Model (WACCM). This version of WACCM is relaxed to observed meteorological fields from the Goddard Modeling and Assimilation Office (GMAO) GEOS5.1 data assimilation system. HCN has been added to the standard chemical mechanism with a chemical loss by reactions with OH (with a corresponding lifetime of 4.3 years (13)) and with O(¹D). The model also includes wet deposition through washout (which is weak as HCN is insoluble), and parameterized dry deposition over open-ocean (with a corresponding lifetime of 3 months (12)). HCN emissions were determined by scaling CO emissions (using 0.012 HCN/CO molar ratio) for biomass burning and anthropogenic biofuel combustion.
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29. The ACE mission is funded primarily by the Canadian Space Agency. Some funding was also provided by the UK Natural Environment Research Council, NERC. The US National Center for Atmospheric Research is sponsored by the National Science Foundation. We also acknowledge support from the US National Aeronautics and Space Administration.

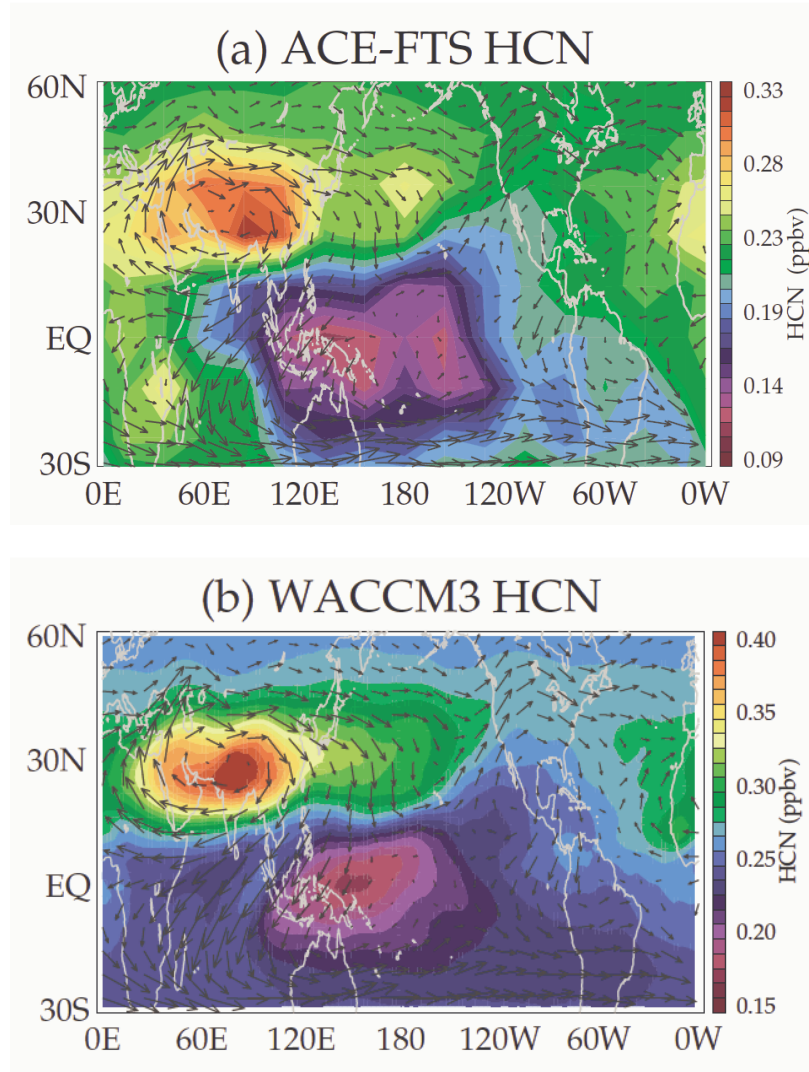


Figure 1. Time average mixing ratio (ppbv) of hydrogen cyanide (HCN) near 13.5 km during boreal summer (June-August) derived from (a) ACE-FTS observations and (b) WACCM chemical transport model calculations. Arrows in both panels denote winds at this level derived from meteorological analysis, showing that the HCN maximum is linked with the upper tropospheric Asian monsoon anticyclone.

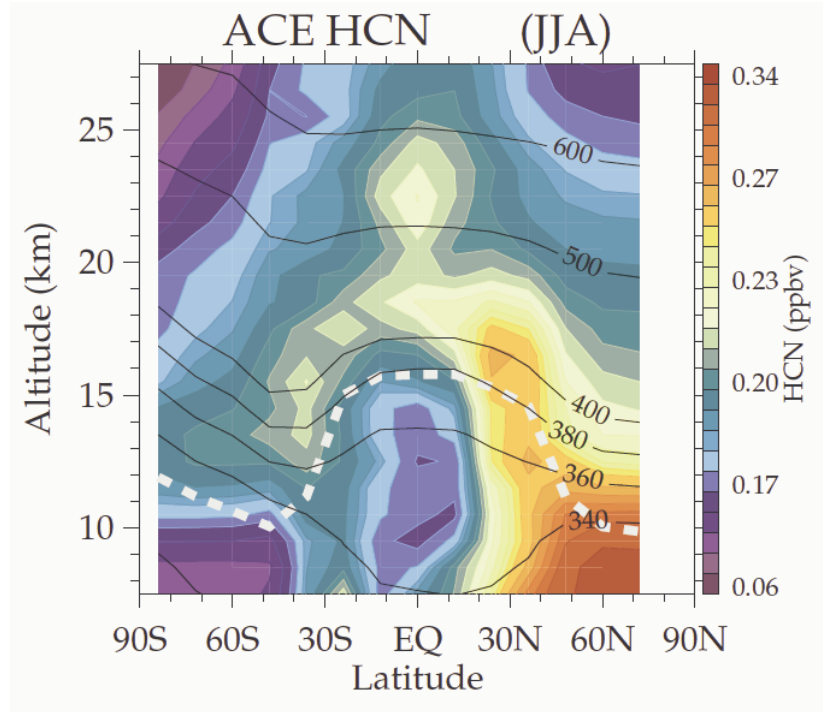


Figure 2. Time and zonal average mixing ratio (ppbv) of hydrogen cyanide (HCN) during boreal summer (June-August) derived from ACE-FTS satellite measurements. The white dashed line denotes the tropopause, and black lines denote isentropic levels.

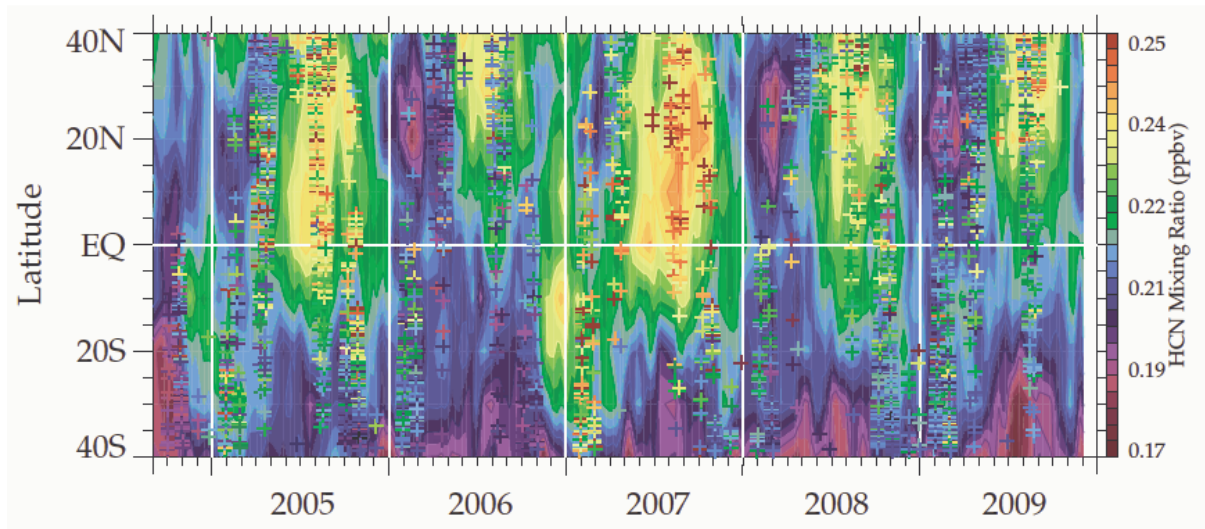


Figure 3. (a) Color contours show latitude-time variations of HCN mixing ratio (ppbv) for the lower stratosphere layer 16-23 km, measured by the Aura MLS satellite during September 2004 – November 2009. These MLS data are zonal mean values averaged over individual week periods, as described in (21). Colored crosses indicate HCN for the 16-23 km layer derived from the ACE satellite measurements, with each cross indicating an individual profile measurement. Comparison of the MLS and ACE-FTS data are shown in Fig. S3.

Supporting Online Material (SOM) for Randel et al, (“Asian monsoon transport of pollution to the stratosphere”).

1. Seasonal ACE-FTS HCN observations

Seasonally averaged ACE-FTS measurements reveal climatological structure of HCN at different altitudes in the upper troposphere and lower stratosphere (Fig. S1). In the upper troposphere (13.5 km) relative minimum values are observed over the tropical Pacific ocean during all seasons, resulting from the upwards convective transport of air depleted in HCN via contact with the ocean surface. These minima extend to the lower stratosphere (17.5 km) during boreal winter and spring, reflecting the stronger mean upward Brewer-Dobson circulation during these seasons. Enhanced HCN values are observed at 13.5 km during boreal spring over Indonesia and Africa, during boreal summer in the Asian monsoon, and during austral spring in the SH subtropics over Indonesia, Africa and South America; these latter are the strongest seasonal HCN sources in the troposphere. In the lower stratosphere (17.5 km) the maximum values are associated with the Asian monsoon; the tropospheric maxima from other seasons do not extend into the lower stratosphere. Note that the enhanced stratospheric HCN during boreal summer over $\sim 20^{\circ} - 40^{\circ}$ N persists in the same latitude band throughout autumn (as also seen in Fig. 3).

Zonally averaged seasonal ACE-FTS observations of HCN over 7.5 – 27.5 km (Fig. S2) reveal further details of the chemical coupling between the troposphere and stratosphere, and highlight the importance of the Asian monsoon circulation. The enhanced HCN in the lower stratosphere during boreal summer persists through the following seasons, with transport into the tropics subsequently entrained into the upward Brewer-Dobson circulation. Note the upward propagation of the near-equatorial maximum throughout the year, so that by the following boreal summer there is a secondary tropical maximum near 22.5 km. Figure S2 also highlights that the large HCN sources during austral spring (over $\sim 10^{\circ}$ -40 S) are mainly confined to the troposphere, with little direct extension above the tropopause. There is evidence for transport of these HCN emissions into the extratropical lowermost stratosphere, with weak maxima observed during the following seasons, and possible links to the tropics. However, the overall patterns clearly highlight the dominant role of transport to the stratosphere from the Asian summer monsoon.

2. Comparison of ACE-FTS and MLS observations

Remote sensing measurements of HCN place stringent requirements on satellite observations and retrieval methodologies, because of the extremely low ambient mixing ratios (in the range of 0.1 – 0.3 ppbv). The ACE-FTS and MLS retrievals of HCN are preliminary science products, with less emphasis to date on validation (although the approximate values in the upper troposphere and lower stratosphere are in reasonable agreement with previous ground-based and aircraft observations^{12,13}). The ACE-FTS and MLS observations in the lower stratosphere (16-23 km, shown in Fig. 3) provide an opportunity for direct comparison between these data (Fig. S3), insofar as the measurements overlap in space and time. We note that there are substantial differences in space-time sampling between the ACE-FTS data (which represent individual profile measurements) and the MLS data (which are weekly zonal average amounts). In spite of these differences, the comparisons (Figs. 3 and S3) show reasonable overall agreement for the magnitude and variability of HCN in the lower stratosphere, providing enhanced confidence in these observations.

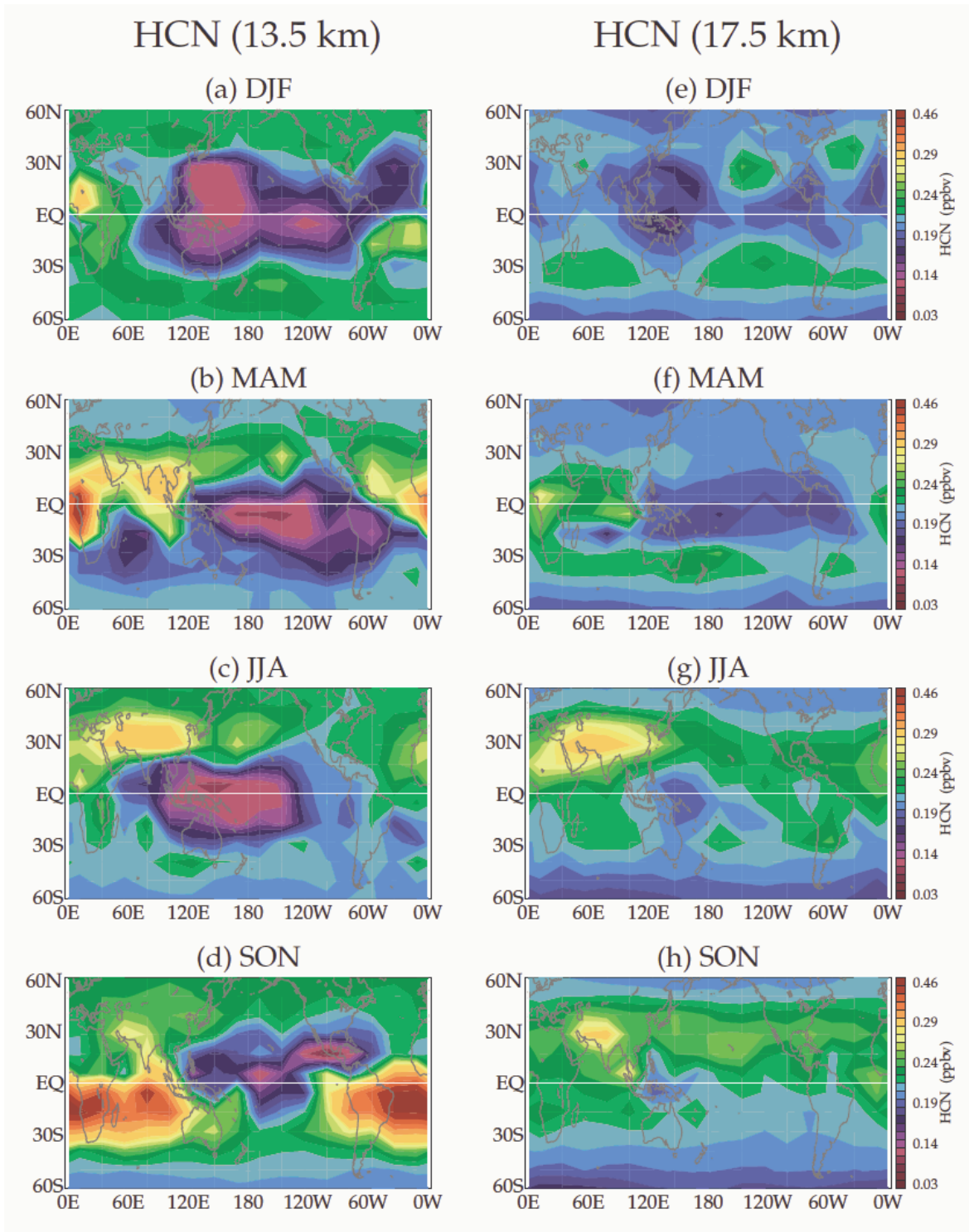


Figure S1. Climatological seasonal structure of HCN (ppbv) derived from ACE-FTS data, for measurements near 13.5 km (left) and 17.5 km (right).

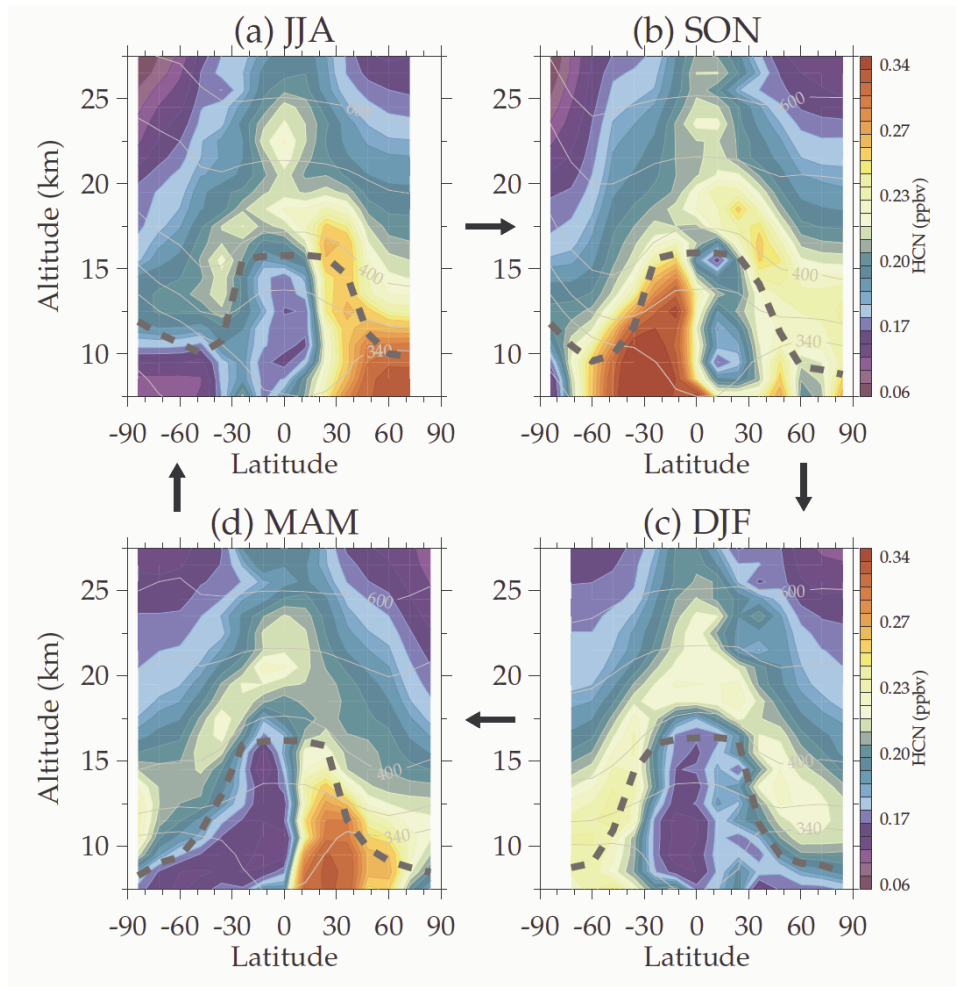


Figure S2. Climatological seasonal average zonal mean cross sections of HCN mixing ratio (ppbv) derived from ACE-FTS data. The grey lines denote isentropic levels, and the thick dashed lines show the seasonally varying tropopause.

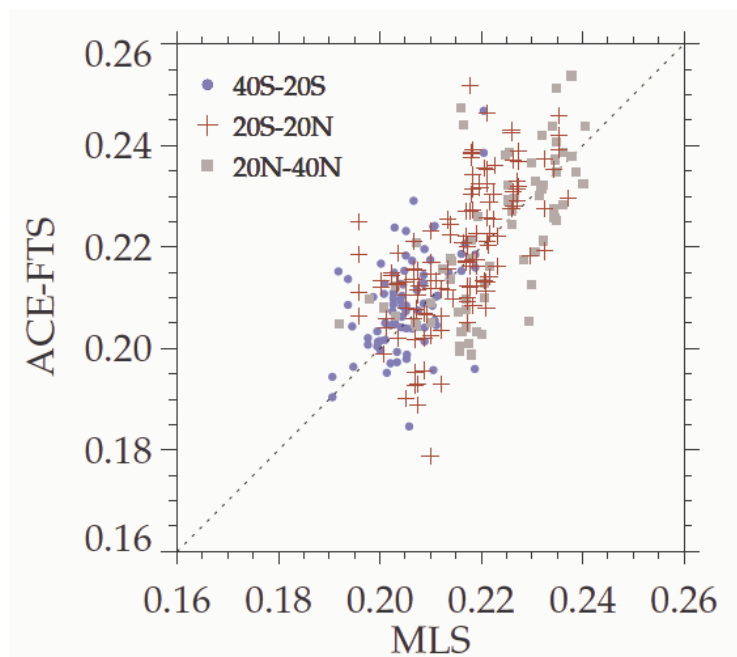


Figure S3. Comparison of ACE-FTS and MLS measurements of HCN (ppbv) in the lower stratosphere (16-23 km) shown in Fig. 3. The ACE-FTS data are individual profile measurements (with sampling shown in Fig. 3), while the MLS data are weekly zonal average values, sampled at the latitude of the corresponding ACE-FTS measurements. The correlation coefficient is $r=0.68$.