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Supporting Online Material for

Asian Monsoon Transport of Pollution to the Stratosphere

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This PDF file includes:

SOM Text
Figs. S1 to 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 ~ 20^o – 40^o 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 ~10^o-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.

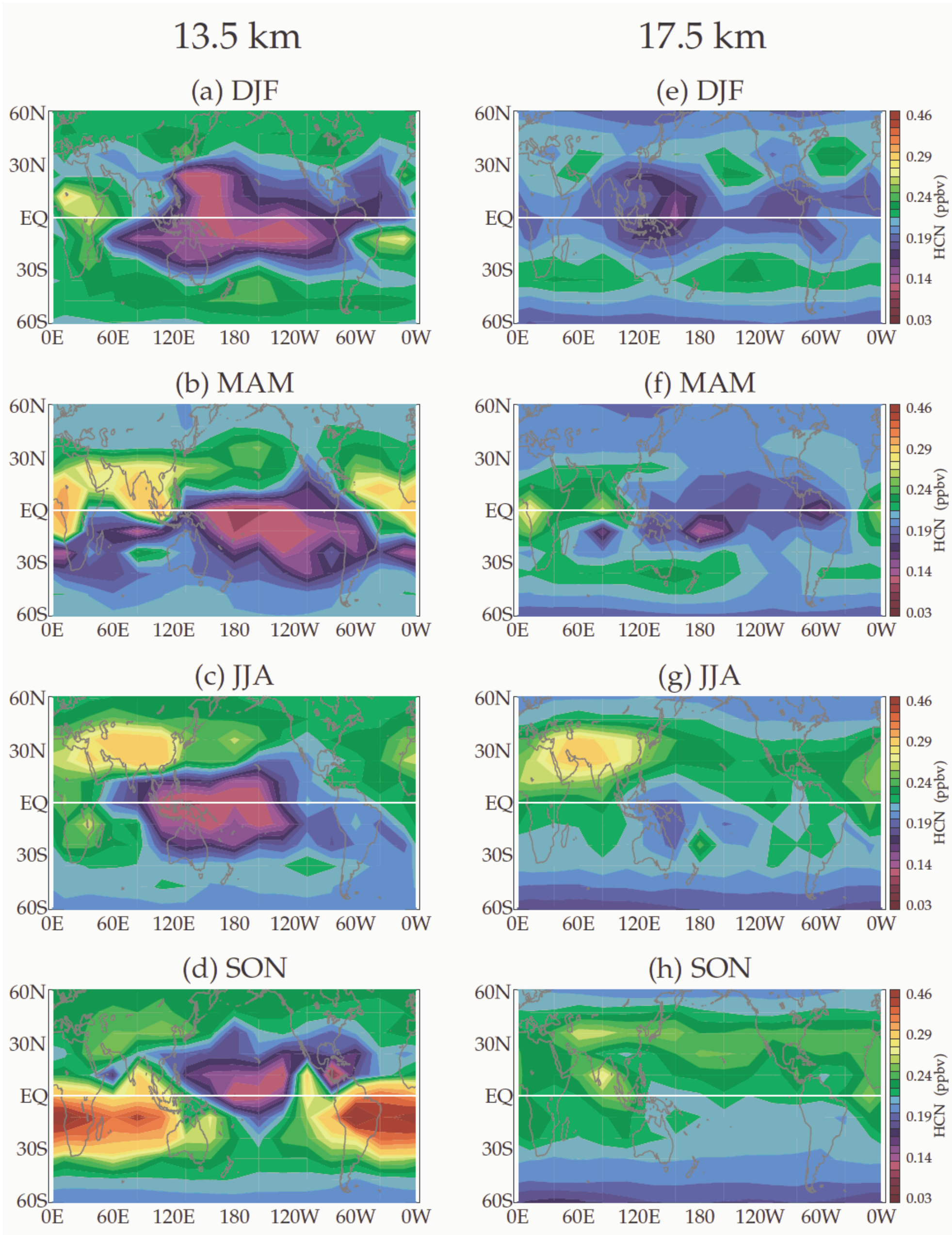

Figure S1. Climatological seasonal structure of HCN (ppbv) derived from ACE-FTS data, for measurements near 13.5 km (left) and 17.5 km (right).

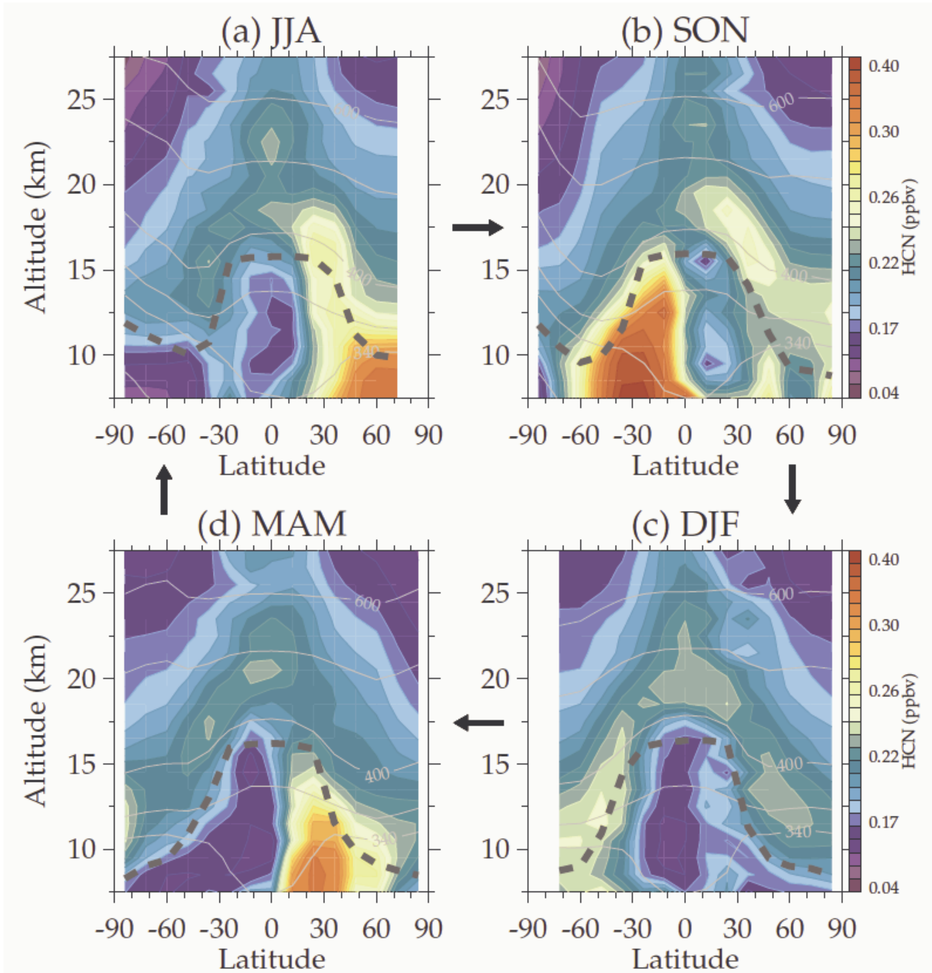

Figure S2. Climatological seasonal average zonal mean cross sections of HCN mixing ratio (ppbv) derived from ACE-FTS data. The grey lines denote isentropic levels, and the thick dashed lines show the seasonally varying tropopause.

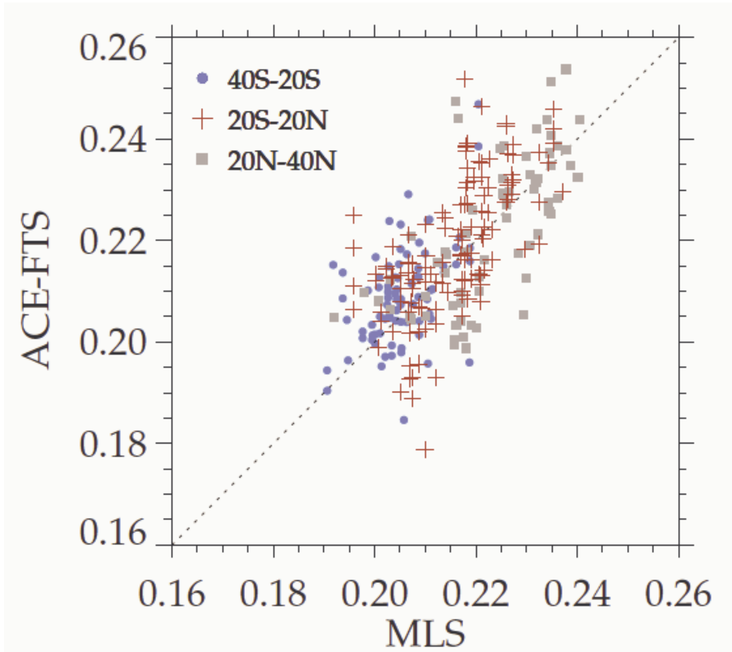

Figure S3. Comparison of ACE-FTS and MLS measurements of HCN (ppbv) in the lower stratosphere (16-23 km) shown in Fig. 3. The ACE-FTS data are individual profile measurements (with sampling shown in Fig. 3), while the MLS data are weekly zonal average values, sampled at the latitude of the corresponding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