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[1] Variations in the mixing ratio of long-lived trace gases entering the stratosphere in the tropics are carried upward with the rising air with the signal being observable throughout the tropical lower stratosphere. This phenomenon, referred to as "atmospheric tape recorder" has previously been observed for water vapor, CO_2, and CO which exhibit an annual cycle. Recently, based on Microwave Limb Sounder (MLS) and the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) satellite measurements, the tape recorder signal has been observed for hydrogen cyanide (HCN) but with an approximately two-year period. Here we report on a model simulation of the HCN tape recorder for the time period 2002–2008 using the Chemical Lagrangian Model of the Stratosphere (CLaMS). The model can reproduce the observed pattern of the HCN tape recorder signal if time-resolved emissions from fires in Indonesia are used as lower boundary condition. This finding indicates that inter-annual variations in biomass burning in Indonesia, which are strongly influenced by El Niño events, control the HCN tape recorder signal. A longer time series of tropical HCN data will probably exhibit an irregular cycle rather than a regular biannual cycle. **Citation:** Pommrich, R., R. Müller, J.-U. Grooß, G. Günther, P. Konopka, M. Riese, A. Heil, M. Schultz, H.-C. Pumphrey, and K. A. Walker (2010), What causes the irregular cycle of the atmospheric tape recorder signal in HCN?, *Geophys. Res. Lett.*, *37*, L16805, doi:10.1029/2010GL044056.

1. Introduction

[2] Tropospheric air enters the stratosphere mainly in the tropics. Although it is known that the air motion in the tropical stratosphere is generally upward and the tropical air is to a large extent separated from extra-tropical air by a transport barrier [e.g., *Plumb*, 2007; *Randel et al.*, 2007; *Konopka et al.*, 2009] large uncertainties remain about the transport pathways for tropospheric pollutants to reach the stratosphere [e.g., *Randel et al.*, 2010] and about the magnitude of upward velocities in the tropical lower stratosphere [e.g., *Fueglistaler et al.*, 2009; *Ploeger et al.*, 2010]. It has been observed that variations in the mixing ratio of sufficiently long-lived trace gases entering the stratosphere are carried upward with the rising air with the signal being observable throughout the tropical lower stratosphere. *Mote et al.* [1995] first observed this phenomenon for water vapor and *Mote et al.* [1996] introduced the term "atmospheric tape recorder". The tape recorder phenomenon has also been observed for CO_2 [*Andrews et al.*, 1999] and for CO [*Schoeberl et al.*, 2006; *Randel et al.*, 2007]. For all three trace gases, an annual cycle is observed in the lowermost tropical stratosphere. In the case of H_2O, the seasonality is driven largely by the seasonal cycle in tropopause temperatures, in case of CO_2 by the seasonal cycle of tropospheric CO_2 [*Keeling et al.*, 2008], and in case of CO it is suggested to be driven by the seasonal cycle in biomass burning [*Schoeberl et al.*, 2006; *Randel et al.*, 2007; *Schoeberl et al.*, 2008]. Recently, a tape recorder signal has also been observed for HCN (an unique tracer for analyzing the transport of tropospherical pollutants into the stratosphere [*Randel et al.*, 2010]), based on Microwave Limb Sounder (MLS) and Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) satellite measurements, but with an approximately two-year period [*Pumphrey et al.*, 2008]. *Pumphrey et al.* [2008] suggested that this signal might be connected to inter-annual variations in biomass burning in Indonesia and the surrounding region. An alternative explanation is offered by *Li et al.* [2009], who suggest a connection between the QBO and the "two-year-cycle" of the HCN tape recorder. Here, we want to answer the question using a different approach.

[3] The questions that we pose are, first, if is it possible to reproduce the pattern of the HCN tape recorder by using HCN emissions only from biomass burning in Indonesia, and, second, what is the cause of the apparent two-year cycle. We performed multi-annual simulations of the HCN tape recorder signal over the time period mid-2005 to end-2007 using the Chemical Lagrangian Model of the Stratosphere (CLaMS) [*McKenna et al.*, 2002a, 2002b; *Konopka et al.*, 2007] and compared the results of our simulation with data from MLS and ACE-FTS.

[4] The HCN boundary condition at the surface is prescribed based only on Indonesian biomass burning emissions. Most of the HCN emissions produced by biomass burning in Indonesia emanate from peat fires, which emit particularly large amounts of HCN per unit mass fuel compared to other fuel types [*Christian et al.*, 2003]. The peat fires in Indonesia are one of the largest HCN sources world-wide [*Schultz et al.*, 2008].

[5] Our simulations demonstrate that the observed *pattern* of the HCN tape recorder signal can be reproduced by the variation of emission fluxes from biomass burning in Indonesia. Our findings confirm the suggestions by *Pumphrey et al.* [2008] and *Randel et al.* [2010] that the

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Table 1. Total Annual Indonesian Fire Emissions of HCN

Year (JUL–JUN)	TgN (as HCN)/year
2001/2002	0.063
2002/2003	0.234
2003/2004	0.043
2004/2005	0.136
2005/2006	0.064
2006/2007	0.246

stratospheric HCN tape recorder signal might be driven by interannual variations in biomass burning in Indonesia which are strongly influenced by El Niño events.

2. Model Description and Simulation Setup

[6] The Chemical Lagrangian Model of the Stratosphere (CLaMS) is a chemical transport model designed to simulate chemistry, advection, and mixing in the stratosphere and upper troposphere [*McKenna et al.*, 2002a, 2002b; *Konopka et al.*, 2007, 2010]. Because of the Lagrangian representation of transport, with the intensity of mixing in model transport being driven by the strength of flow deformation, the model is particularly well suited for the simulation of tracer transport in the vicinity of strong transport barriers and the associated tracer gradients. The specific model setup employed here follows closely the setup described by *Konopka et al.* [2007]. The initial values for HCN are taken from a CLaMS model climatology derived from a perpetual simulation. The vertical velocities are corrected as described by *Konopka et al.* [2009]. The horizontal resolution of our simulation is 100 km and the vertical resolution in the tropopause region is about 400 m. The model domain extends from the ground to 2500 K (≈56 km). The meteorological fields were taken from the operational analyses of the European Centre for Medium-Range Weather Forecasts (ECMWF). As lower boundary condition we used emissions from the region of Indonesia as source of HCN. In order to specifically test the hypothesis that the HCN tape recorder is primarily driven by Indonesian biomass burning emissions, we prescribe only emissions of HCN from the Indonesian region (between 94°E and 141°E and 11°S and 8°N). Global HCN emission inventories are subject to large uncertainties. However, there are no indications of substantial interannual variability of HCN emissions outside of Indonesia [*Schultz et al.*, 2008; *Lupu et al.*, 2009], so they are neglected here.

[7] The only tracer considered here is HCN. Chemical loss of HCN in the atmosphere through the reaction of HCN with OH is taken into account where the OH concentrations are prescribed as a climatology from a two-dimensional model [*Grooß*, 1996]. Other loss mechanisms of HCN, through reactions with $O(^1D)$, $O(^3P)$ and through photolysis, as well as production by reactions of CH_3CN can be neglected.

[8] Monthly HCN emissions from Indonesian biomass burning were estimated using the approach described by *Heil* [2007], in which emissions are calculated separately for fires in grassland, forests, and peat soil. Here, the emission parametrisation for fuel load and combustion efficiency is based on 50 percentile values of the range of values given by *Heil* [2007]. A description of the procedure is also given in the supplementary material of *Schultz et al.* [2008]. For grassland and forests, the fuel-type dependent emission factors used here (0.23 and 0.30 g/kg dry matter burned) refer to the intermediate value of *Andreae and Merlet* [2001] while the emission factor for peat (8.1 g HCN/kg dry matter) is based on the only laboratory measurement of an Indonesian peat sample published so far [*Christian et al.*, 2003]. The Indonesian fire emissions correspond to the annual (JUN–JUL) HCN totals (TgN/year) listed in Table 1.

[9] The monthly HCN emission rates are provided on a 0.5° × 0.5° grid. For the simulation the monthly emission rates are converted into daily rates and it is assumed that the emissions are released into the bottom model layer corresponding to the emission site. The dominant sink for HCN is uptake by the ocean [*Li et al.*, 2000]. We adjusted the loss rate at the ocean surface such that a mean atmospheric residence time of HCN of ≈5 month [*Singh et al.*, 2003] emerges. Over land the uptake of HCN is neglected. *Lupu et al.* [2009] performed a model study of HCN in the upper troposphere and were able to reproduce the observed variability of the volume mixing ratio of HCN in the troposphere and a residence time of ≈5 *month*.

[10] To study the path the air takes from the boundary layer over Indonesia into the region of the tropopause, where the "tape head" is located ($\theta \approx 350$ K), we started daily trajectories in the region between 11° S, 8° N, 94° E, and 141° E from the beginning of 2002 until the end of 2007. The distance between two points is 1°, the release altitude follows the orography. Each trajectory runs for 30 days.

3. Data

[11] We compare our simulation results with HCN research products from the Microwave Limb Sounder (MLS) on the Aura satellite and the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) on SCISAT-1. The HCN data from MLS used for our analysis are described by *Pumphrey et al.* [2006, 2008]. The data between 68 hPa (≈18.7 km) and 0.1 hPa (≈64 km) are of sufficient quality for scientific work. The vertical resolution of the product is reported to be 6 km at an altitude of 32 km, degrading to 8 km at 20 km [*Pumphrey et al.*, 2008].

[12] ACE-FTS is a Fourier transform spectrometer [*Bernath et al.*, 2005; *Boone et al.*, 2005], operating in solar occultation, ACE-FTS features a high signal-to-noise ratio but has limited geographical coverage, measuring up to 32 occultations per day. These usually occur in middle and high latitudes; the measurement latitude passes through the tropics eight times per year. The altitude sampling of the ACE-FTS measurements varies from ≈1.5 to 6 km, but the altitude resolution is 3 km, limited by the instrument's field of view [*Boone et al.*, 2005]. The data from ACE-FTS presented here are a development product for which HCN is retrieved over a larger vertical range than the standard v2.2 product. The retrieval process is described by *Boone et al.* [2005] and the features of the HCN research product are described by *Lupu et al.* [2009]. Near the equator, the majority of retrieved profiles are usable between 68 hPa and 3.8 hPa.

4. Results

[13] Figure 1 shows the simulated HCN mixing ratios in the tropics (±15° latitude) from the ground to the 900 K

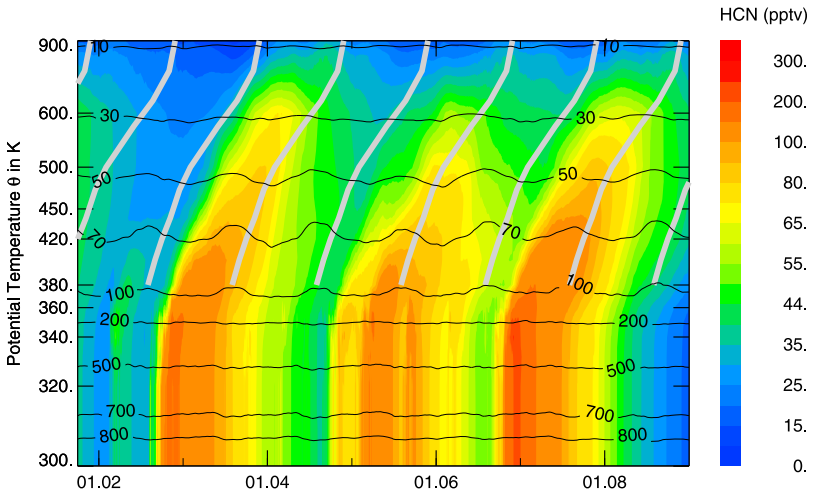

Figure 1. The zonal mean values of the mixing ratio of HCN from the simulation with CLaMS in a latitude window of ±15°. The x-axis is the time, the y-axis reflects the model levels, the black lines are isolines of pressure, and the gray lines show the maximum of the H_2O tape recorder calculated from HALOE measurements [*Grooß and Russell*, 2005]. The mixing ratio in pptv is color coded. The simulated HCN values were interpolated from the hybrid model coordinates [*Konopka et al.*, 2007] to isentropic levels.

potential temperature level. The peak emissions from the biomass burning in Indonesia are clearly visible with rapid vertical transport in the troposphere up to ≈350 K. In the lower tropical stratosphere, vertical velocities are much smaller and a tape recorder pattern emerges. The simulated tropospheric mixing ratios from our simulation are substantially lower than both observations and model simulations with a global emission inventory for HCN indicate [*Lupu et al.*, 2009]. This is expected because we have prescribed only Indonesian emissions (which constitute between ≈5% (2003) and ≈26% (2006) of the global emissions from biomass burning) at the lower boundary.

[14] However, the focus here is on the simulated stratospheric *anomaly pattern* in the tropics. The model results are shown in Figure 2 (bottom); the corresponding MLS observations are shown in Figure 2 (top left). For comparison with MLS, the CLaMS results were smoothed by a 6 km wide vertical filter to approximately take into account the limited vertical resolution of MLS. Overall, the model results agree well with the observations; most importantly the unusual, "two-year" cycle of the HCN tape recorder is reproduced. There is a good correspondence between the simulated and the observed onset of high HCN values at 100 hPa in April 2005 and December 2006. Likewise, the high HCN tape recorder signal that is still noticeable above 30 hPa in late 2004, is reproduced. It is also remarkable that the amplitude of the anomaly is well represented in the model simulation in-spite of the fact that only Indonesian

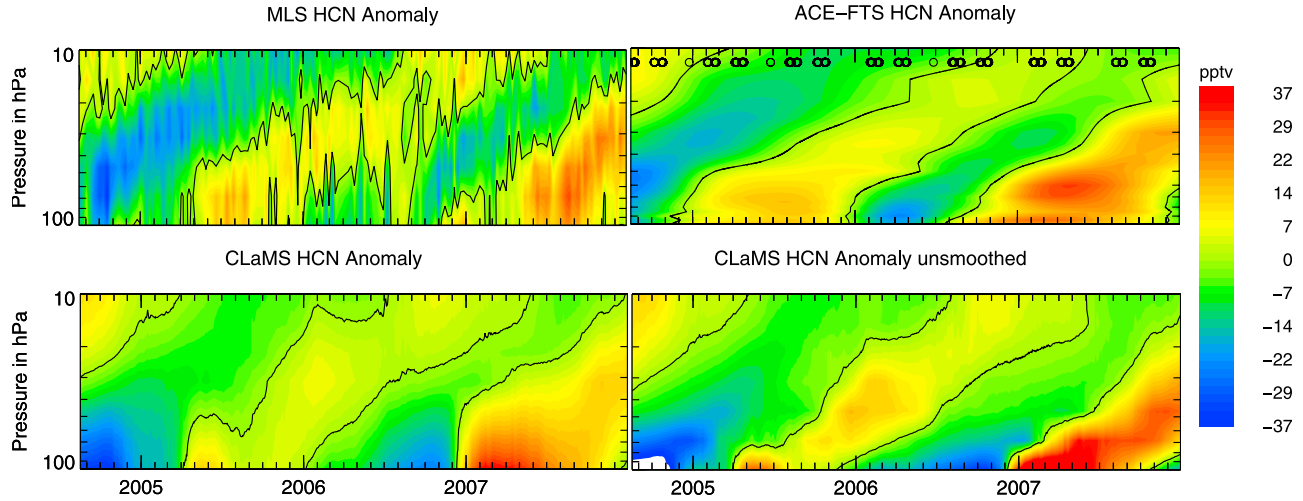

Figure 2. Comparison of the anomaly of the equatorial zonal mean mixing ratio of HCN in a latitude window of ±15° over time. Anomaly of the mean volume mixing ratio of (top left) MLS and (top right) ACE-FTS HCN measurements, and (bottom) the anomaly of the volume mixing ratio of HCN from the CLaMS simulation. The circles show the periods when ACE-FTS made measurements in the shown latitude range. The anomaly is the difference of the zonal mean volume mixing ratio and the mean volume mixing ratio over the shown time.

emissions are prescribed at the lower boundary. This is a strong indication that the HCN anomaly pattern visible in the MLS measurements is driven by Indonesian emissions.

[15] These results are corroborated by the comparison of the simulated HCN pattern with the ACE-FTS measurements (Figure 2, top right). Here, owing to the better vertical resolution of ACE-FTS, the simulated HCN values are taken from the model results without vertical smoothing. As for the comparison with MLS, both the timing and the amplitude of the HCN anomaly pattern agrees well when the CLaMS model results are compared to the ACE-FTS measurements. A model deficiency noticeable in the comparison of the simulation results both with MLS and ACE-FTS is the apparently too rapid ascent in the stratosphere above 30 hPa. The representation of horizontal in-mixing from the mid-latitude stratosphere into the TTL in the model might contribute to this problem, however, a too large vertical velocity in the tropical lower stratosphere is known from other model simulations [e.g., *Fueglistaler et al.*, 2009].

[16] The results of the trajectory study corroborate the link between the observed anomaly patterns and Indonesian emissions. The number of trajectories per month that reached the region of the "tape head" is shown in Figure S1a of the auxiliary material.[1] This shows the well known annual cycle of upward transport of air masses in the tropics [e.g., *Randel et al.*, 2007; *Liu et al.*, 2007]. Figure S2b displays only trajectories that started from areas with emissions of HCN. Figure S2c shows the monthly total mass of emitted HCN. The normalized product of the number of the trajectories that potentially carry HCN and the emitted HCN is shown in Figure S2d. This quantity clearly shows an apparent two-year cycle. The bottom four plots show the signal of the simulated HCN tape recorder in CLaMS in four heights: ≈270 hPa (Figure S2e) ≈160 hPa (Figure S2f) ≈100 hPa, (Figure S2g) ≈80 hPa (Figure S2h) by means to show heights below, midst and above the TTL. The coincidence of the maxima of trajectories reaching the 350 K level and the tape recorder signals is evident. By applying the amount of emitted HCN as a weight to the number of trajectories reaching 350 K, the intensity of the signal changes slightly, but the same conclusion results.

[17] El Niño events occur at irregular intervals ranging from one year to a decade. In the years 2002, 2004, and 2006, however, three El Niño events occurred in a row. The temporal coincidence of strong emissions of HCN and El Niño events implies that the signal will have an irregular cycle rather than a two-year cycle. An interrelation between biomass burning and El Niño, on the example of CO, is described by *Chandra et al.* [2009]. This is consistent with a study of *Duncan et al.* [2007], who report "that the impact of biomass burning pollution on the UT/LS is likely greatest during an El Niño event due to favorable dynamics and historical higher burning rates".

5. Conclusions

[18] The results of the simulations with the chemistry transport model CLaMS show that both the timing and the amplitude of the HCN tape recorder anomaly pattern can be well reproduced when prescribing only HCN emissions from Indonesian biomass burning. Through a trajectory study, we demonstrated that the interplay between HCN emissions in Indonesia and rapid upward transport to the bottom of the TTL is responsible for producing the anomaly pattern. Because of the influence of El Niño events on fires in Indonesia the variability of El Niño will have an impact on the HCN tape recorder cycle. Therefore, we propose that the HCN tape recorder cycle is an irregular cycle rather than a two-year cycle.

[19] **Acknowledgments.** We thank Bill Randel for very helpful discussions. We thank the ECMWF for providing meteorological analyses. The Atmospheric Chemistry Experiment (ACE) is a Canadian-led experiment mainly supported by the Canadian Space Agency.

[1] Auxiliary materials are available in the HTML. doi:10.1029/2010GL044056.

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