Asian Monsoon Transport of Pollution to the Stratosphere

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curred when the large DOC pool had been reduced in size enough to no longer represent a negative feedback to global climatic cooling.

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Supporting Online Material

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Materials and Methods
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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 substantially 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,

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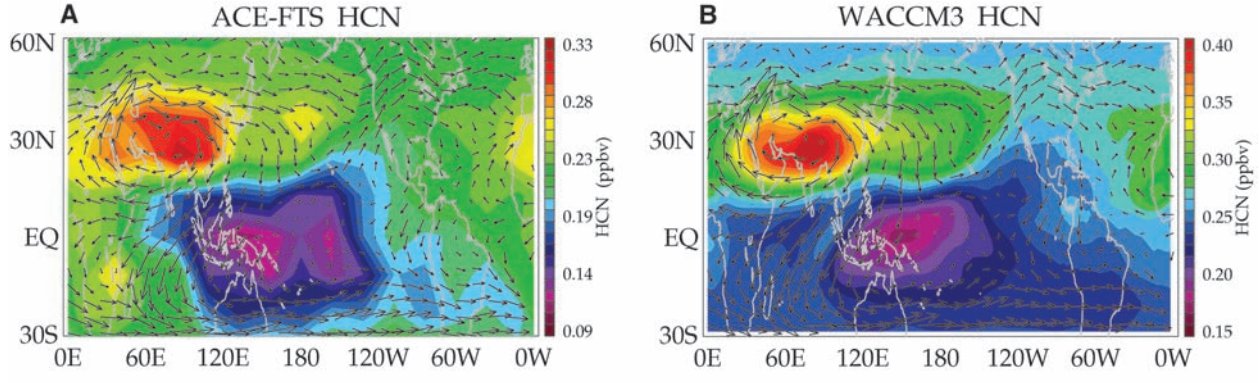

Fig. 1. Time average mixing ratio [parts per billion by volume (ppbv)] of HCN near 13.5 km during boreal summer (June to August) derived from (**A**) ACE-FTS observations and (**B**) WACCM chemical transport model calculations. Arrows in both panels denote winds at this level derived from meteorological analysis, showing that the HCN maximum is linked with the upper tropospheric Asian monsoon anticyclone.

HCN has a long photochemical lifetime of more than 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*–*17*) (Fig. 1A) reveal 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 to August). The tropical minimum for HCN is a distinctive signature that is very different from most other tropospheric pollution tracers, such as carbon monoxide (*18*). The overall structure of HCN is accurately simulated by a three-dimensional (3D) chemical transport model (Fig. 1B) incorporating HCN sources from wildfires and biofuel combustion, plus an imposed sink from contact with the ocean surface (*19*). 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 to May), and burning over Africa and South America during austral spring (September to 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 (*20*). 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 (*21*) 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 to 23 km) from late 2004 to the end of 2009 (Fig. 3) show HCN maxima in the Northern Hemisphere subtropics during each boreal summer (~June to October); this is a clear fingerprint of the Asian monsoon influence (consistent with the ACE-FTS observations in Fig. 2 and fig. 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 (*22*) have demonstrated that these 2005 and 2007 equatorial HCN maxima propagate coherently upward into the stratosphere with the tropical Brewer-Dobson circulation; this so-called tape-recorder effect is evident in other stratospheric trace constituents (e.g., H_2O) that have seasonal or interannual anomalies originating near the tropical tropopause (*23*). Figure 3 demonstrates that these stratospheric anomalies are linked to enhanced boreal summer (Asian monsoon) maxima during 2005 and 2007. Figure 3 also shows isolated HCN maxima in the Southern Hemisphere

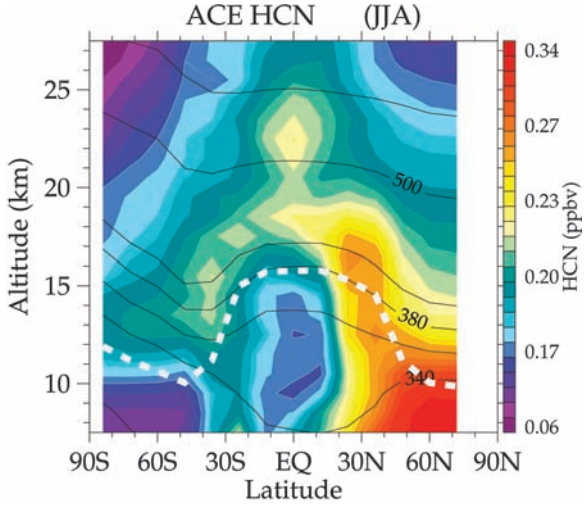

Fig. 2. Time and zonal average mixing ratio (ppbv) of HCN during boreal summer (June to August) derived from ACE-FTS satellite measurements. The white dashed line denotes the tropopause, and black lines denote isentropic levels.

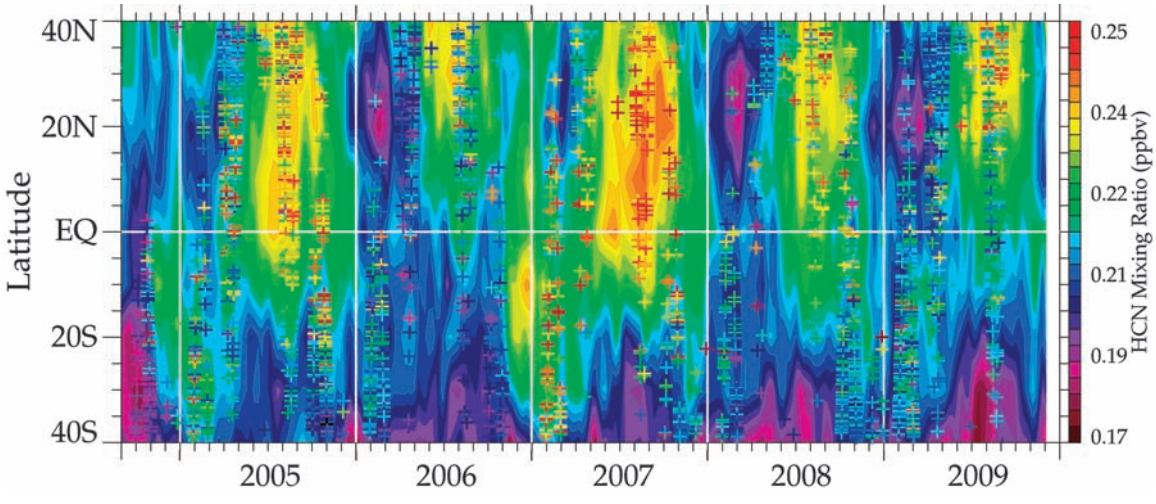

Fig. 3. Color contours show latitude-time variations of HCN mixing ratio (ppbv) for the lower stratosphere layer, 16 to 23 km, measured by the Aura MLS satellite during September 2004 to November 2009. These MLS data are zonal mean values averaged over individual week periods, as described in (*22*). Colored crosses indicate HCN for the 16- to 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.

subtropics (~0° to 20°S) during late 2004 and late 2006, which result from enhanced austral spring burning over Indonesia during these years (*24*). 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 quasibiennial oscillation (QBO)], but we 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 (*25*) 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 discernible 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 (*26*), 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 (SO_2), reactive nitrogen species (NOx), 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 SO_2 emissions over China since 2002 (*27*), 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 (*28*), with large continued increases over the next decades expected for SO_2 and NOx. Furthermore, potential changes in the strength and variability of the Asian monsoon circulation in an evolving climate [linked to increased convection and rainfall (*29*)] could modify this transport pathway, with potential influence on composition and climate of the stratosphere.

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19. Figure 1B shows results from the National Center for Atmospheric Research 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 because 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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Lab Experiments for the Study of Social-Ecological Systems

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Governance of social-ecological systems is a major policy problem of the contemporary era. Field studies of fisheries, forests, and pastoral and water resources have identified many variables that influence the outcomes of governance efforts. We introduce an experimental environment that involves spatial and temporal resource dynamics in order to capture these two critical variables identified in field research. Previous behavioral experiments of commons dilemmas have found that people are willing to engage in costly punishment, frequently generating increases in gross benefits, contrary to game-theoretical predictions based on a static pay-off function. Results in our experimental environment find that costly punishment is again used but lacks a gross positive effect on resource harvesting unless combined with communication. These findings illustrate the importance of careful generalization from the laboratory to the world of policy.

Designing and conducting laboratory experiments in the social sciences enables the unpacking of complex problems to examine the effects of different components on outcomes and to replicate results with diverse participants (*1*). In this report, we discuss an experimental research program on the study of social-ecological systems, especially the governance of common-pool resources (CPRs). CPRs are resource systems where the harvesting of resource units by one user subtracts units from a pool potentially available to others. Examples in the field include forests, pastures, fisheries, and water systems.

The widely accepted economics textbook model of CPRs (*2*, *3*) is a simple, static production function that is used to conclude that the users of a