Signatures of Nitrogen Chemistry in Hot Jupiter Atmospheres

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Abstract

Inferences of molecular compositions of exoplanetary atmospheres have generally focused on carbon, hydrogen, and oxygen-bearing molecules. Recently, additional absorption in Hubble Space Telescope Wide Field Camera 3 (WFC3) transmission spectra around 1.55 μm has been attributed to nitrogen-bearing chemical species: NH₃ or HCN. These species, if present in significant abundance, would be strong indicators of disequilibrium chemical processes—e.g., vertical mixing and photochemistry. The derived N abundance, in turn, could also place important constraints on planetary formation mechanisms. Here, we examine the detectability of nitrogen chemistry in exoplanetary atmospheres. In addition to the WFC3 bandpass (1.1–1.7 μm), we find that observations in the K-band at ~2.2 μm, achievable with present ground-based telescopes, sample a strong NH₃ feature, while observations at ~3.1 μm and ~4.0 μm sample strong HCN features. In anticipation of such observations, we predict absorption feature amplitudes due to nitrogen chemistry in the 1–5 μm spectral range possible for a typical hot Jupiter. Finally, we conduct atmospheric retrievals of nine hot Jupiter transmission spectra in search of near-infrared absorption features suggestive of nitrogen chemistry. We report weak detections of NH₃ in WASP-31b (2.2σ), HCN in WASP-63b (2.3σ), and excess absorption that could be attributed to NH₃ in HD 209458b. High-precision observations from 1 to 5 μm (e.g., with the James Webb Space Telescope) will enable definitive detections of nitrogen chemistry, in turn serving as powerful diagnostics of disequilibrium atmospheric chemistry and planetary formation processes.

Key words: methods: data analysis – planets and satellites: atmospheres – techniques: spectroscopic

1. Introduction

The chemical characterization of exoplanetary atmospheres is rapidly entering a golden era. Robust detections of carbon, hydrogen, and oxygen-bearing molecules from infrared spectroscopy are now commonplace (e.g., Snellen et al. 2010; Deming et al. 2013; Sing et al. 2016). Optical transmission spectra have offered detections of Na, K, and TiO (e.g., Snellen et al. 2008; Sing et al. 2015; Wilson et al. 2015; Sedaghati et al. 2017), though they are often plagued by clouds or hazes (Ehrenreich et al. 2014; Knutson et al. 2014; Kreidberg et al. 2014). H₂O is the most frequently observed molecule in exoplanetary atmospheres (Madhusudhan et al. 2016), enabled by high-precision spectra from the Hubble Space Telescope’s (HST) Wide Field Camera 3 (WFC3). However, H₂O is not the only molecule with strong features in the ~1.1–1.7 μm WFC3 range. Additional features, due to CH₄, NH₃, and HCN, need to be considered when modeling exoplanetary atmospheres (MacDonald & Madhusudhan 2017).

Nitrogen chemistry is expected to exist in exoplanetary atmospheres (Burrows & Sharp 1999; Lodders & Fegley 2002). However, the anticipated equilibrium abundances of such species in the upper atmospheres of hot Jupiters are small: ~10⁻⁷ and ~10⁻⁸ for NH₃ and HCN, respectively—assuming solar composition, C/O = 0.5, and N/O = 0.2 at ~1500 K (Madhusudhan 2012; Heng & Tsai 2016). Detecting such trace abundances is impractical with current observations, often leading to the exclusion of such molecules from exoplanetary spectral analyses. However, observable nitrogen chemistry may occur under some circumstances. One avenue is enhanced elemental ratios: HCN abundances increase by ~10⁴ for C/O ≥ 1 (Madhusudhan 2012); both NH₃ and HCN weakly increase with N/O (Heng & Tsai 2016). Such enhanced ratios could be remnants of planetary formation (Öberg et al. 2011; Madhusudhan et al. 2014; Mordasini et al. 2016; Piso et al. 2016).

Alternatively, disequilibrium chemistry can enhance NH₃ and HCN abundances by ≥ 2 orders of magnitude over equilibrium expectations at altitudes probed by transmission spectra (Zahnle et al. 2009; Moses et al. 2011, 2013; Venot et al. 2012). There are two principle disequilibrium avenues: transport-induced quenching (e.g., via vertical mixing) and photochemistry (e.g., by UV irradiation). Quenching occurs in atmospheric regions whereby a dynamical transport process is characteristically faster than a certain chemical reaction (e.g., N₂ + H₂ → NH₃). The transport process then fixes the chemical abundances to equilibrium values from those atmospheric regions where local conditions result in a commensurate reaction timescale. For NH₃ and HCN, this occurs in the deep atmosphere (pressures ~1 bar—Moses et al. 2011), where equilibrium abundances are considerably higher. Vertical mixing then dredges up these molecules to the upper atmosphere.

Photochemistry can enhance HCN abundances, at the expense of NH₃, CH₄, and N₂, at pressures < 10⁻⁵ bar (Zahnle et al. 2009; Moses et al. 2011). Photochemical deviations should become more pronounced for lower-temperature planets, due to deeper quench points and slower reaction rates impeding attempts to drive products back toward equilibrium (Moses et al. 2011). These conclusions are relatively insensitive to the C/O ratio (Moses et al. 2013). An atmosphere subject to extreme photochemistry may display abundant HCN and depleted NH₃ in the photosphere, while one with strong vertical mixing and minimal photochemistry could display abundant NH₃ and/or HCN.

The impact of disequilibrium nitrogen chemistry on transmission spectra has been considered before (e.g., Moses...
et al. 2011; Shabram et al. 2011). Shabram et al. (2011) identified HCN absorption features at ∼1.5, 3.3, and 7 μm, suggesting that the James Webb Space Telescope (JWST) NIRSpec prism will be able to observe the former two. Moses et al. (2011) strongly recommended including HCN and NH₃ within spectral analyses. Without including these disequilibrium products, as is somewhat common in atmospheric retrievals, the prospect of detecting nitrogen chemistry has been artificially quenched.

Recently, in MacDonald & Madhusudhan (2017), we reported tentative evidence of nitrogen chemistry in the hot Jupiter HD 209458b. We identified a slope from ∼1.5 to 1.7 μm in the WFC3 transmission spectrum, suggesting NH₃ or HCN as possible contributors. At the precision of the data, either molecule provided reasonable fits. However, qualitatively different WFC3 features become apparent at higher resolution: an “NH₃ shoulder” redward of the 1.4 μm H₂O feature, versus a “HCN peak” around 1.55 μm. The NH₃ feature appears to have been missed in prior studies, possibly due to not including it in models (Deming et al. 2013; Madhusudhan et al. 2014; Barstow et al. 2017) or a priori assumed chemistry (Benneke et al. 2015; Sing et al. 2016). Incomplete opacity data below ∼3 μm (e.g., Shabram et al. 2011, Figure 5) could also contribute, as many studies pre-date the latest NH₃ and HCN line lists (Tennyson et al. 2016). This initial evidence has motivated retrievals to include nitrogen chemistry for other planets. For example, Kilpatrick et al. (2017) observed an apparent absorption feature at 1.55 μm in WASP-63b’s transmission spectrum. Atmospheric retrievals by four different groups identified this as consistent with super-solar HCN.

In this Letter, we identify spectral signatures of nitrogen chemistry in exoplanetary atmospheres that are detectable with present and upcoming instruments. We then examine transmission spectra of nine hot Jupiters for signs of nitrogen chemistry.

2. Atmospheric Modeling and Retrieval Framework

We model transmission spectra in a plane-parallel geometry for planetary atmospheres under hydrostatic equilibrium, using the POSEIDON radiative transfer and retrieval algorithm (MacDonald & Madhusudhan 2017). We assume uniform-in-altitude terminator-averaged mixing ratios, terminator-averaged temperature structure, and allow for inhomogeneous clouds/hazes.

We consider the major sources of opacity expected in H₂-dominated atmospheres: H₂O, CH₄, NH₃, HCN, CO, CO₂, Na, and K (Madhusudhan et al. 2016), along with H₂–H₂ and H₂–He collision-induced absorption. The opacity sources are described in Gandhi & Madhusudhan (2017) and use molecular line lists from the EXOMOL (Tennyson et al. 2016) and HITEMP databases (Rothman et al. 2010).

We use this forward model in two ways. In Section 3, we first generate transmission spectra to investigate signatures of nitrogen chemistry over a range of planetary parameters. Second, in Section 4, we couple the forward model with a Bayesian parameter estimation and model comparison retrieval code. This enables us to derive constraints on nitrogen chemistry from the observed spectra of a sample of hot Jupiters.

Our models have a maximum of 19 free parameters: six for the temperature profile, eight for mixing ratios, four encoding clouds/hazes, and a reference pressure, P ref. For each parameter set, we generate transmission spectra at R = 1000 from 0.2 to 5.2 μm. The model spectra are convolved with the relevant instrument point-spread-functions and binned to the data resolution. The parameter space is mapped via the MultiNest (Feroz & Hobson 2008; Feroz et al. 2009, 2013) multimodal nested sampling algorithm, implemented by PyMultiNest (Buchner et al. 2014).

3. Detectability of Nitrogen Chemistry

We first examine the optimum near-infrared regions to search for nitrogen chemistry. We begin by comparing the cross-sections of NH₃ and HCN to H₂O. We then explore how atmospheric properties alter NH₃ and HCN absorption signatures. Finally, we consider how these findings can be employed by ground- and space-based facilities to uniquely detect NH₃ and HCN in exoplanetary atmospheres.

3.1. NH3/HCN Absorption Features

Figure 1 contrasts the NH₃ and HCN cross-sections to H₂O from 1 to 5 μm at 1000, 1500, and 2000 K. Where the H₂O cross-section possesses local minima, the cross-sections of nitrogen-bearing molecules may exceed H₂O by ∼2 orders of magnitude. The WFC3 bandpass contains NH₃ and HCN features around ∼1.5–1.6 μm (MacDonald & Madhusudhan 2017), along with a
weaker unique NH3 feature at ∼1.2 μm. NH3 possesses a prominent feature at ∼2.2 μm (K-band), while HCN has an especially strong feature at ∼3.1 μm. Both molecules absorb at 4 μm, between the two Spitzer IRAC bands. The K-band NH3 feature is a powerful diagnostic, coinciding with minima for both H2O and HCN. The cross-section contrast between NH3 or HCN and H2O tends to increase at lower temperatures, suggesting that lower-temperature planets may posses amplified nitrogen chemistry features (see Section 3.2.2). HCN peaks become sharper than NH3 features at lower temperatures, which can enable unique identification in regions of overlapping absorption (e.g., the WFC3 bandpass).

### 3.2. Factors Influencing Detectability

The relative strengths of absorption cross-sections are not the only consideration governing the imprint of nitrogen chemistry into transmission spectra. We henceforth illustrate how the transit depth excess—here the difference between a transmission spectrum model with and without enhanced nitrogen chemistry—varies as a function of NH3/HCN abundance, atmospheric temperature, and across the transition from clear to cloudy atmospheres. We perturb a reference hot Jupiter system with the following properties: \( R_p = 1.2 \) \( R_J \), \( g = 10 \) m s\(^{-2} \), \( T = 1500 \) K (isothermal). The volume mixing ratios, with the exception of NH3 and HCN, are representative of chemical equilibrium: log(H2O) = −3.3, log(CH4) = −6.0, log(CO) = −3.3, log(CO2) = −7.0 (Madhusudhan 2012). These “background” abundances are held constant throughout. The reference model considers NH3/H2O or HCN/H2O = 0.1. We take \( P_{\text{cloud}} = 1 \) mbar, \( P_{\text{ref}} = 10 \) mbar, and a terminator cloud fraction of 50%.

#### 3.2.1. Mixing Ratio

Figure 2 (top) demonstrates that the transit depth excess is strongly correlated with the relative mixing ratios of each nitrogen-bearing species to water—dictated by the relative cross-section differences (Figure 1). Since the cross-sections of NH3 and HCN are rarely enhanced by more than 100× over H2O from 1 to 5 μm, it is unsurprising that absorption signatures become negligible for relative mixing ratios below 10\(^{-2} \). However, when nitrogen chemistry abundances become commensurate with H2O, a plethora of features ≥300 ppm emerge throughout the near-infrared.

#### 3.2.2. Temperature

Figure 2 (middle), illustrates two effects that compete as temperatures lower: i) the H2O cross-section minima deepen (Figure 1); ii) the atmospheric scale height decreases proportionally. The combined effect is for many NH3/HCN features to initially intensify from 2000 K → 1500 K, before the stronger features dampen from 1500 K → 1000 K as the atmosphere contracts. Generally, HCN features become sharper for cooler temperatures, as expected from the cross-sections (Figure 1). Overall, nitrogen chemistry absorption features remain potent over a wide range of temperatures expected in hot Jupiter atmospheres (∼1000–2000 K), especially in the WFC3 bandpass for cooler temperatures. K-band is a robust NH3 diagnostic for \( T \gtrsim 1000 \) K, while the ∼3.1 and 4.0 μm HCN features are prominent for \( T \gtrsim 1500 \) K. Thus enhanced nitrogen chemistry, if present, may even be detectable in some of the higher-temperature hot Jupiters.

### 3.3. A Strategy to Uniquely Detect NH3 and HCN

Figure 2 indicates that WFC3 spectra can enable detections of nitrogen chemistry. In particular, absorption at ∼1.2 μm and in K-band uniquely indicates NH3. HCN absorbs strongly around 3.1 and 4.0 μm. We suggest ground-based K-band photometry and/or spectroscopy as a promising avenue to assess the presence of NH3. Null detections in K-band could rule out NH3, while suggesting HCN as a possible cause of 1.55 μm WFC3 absorption. Furthermore, robust detections of HCN via the ∼3.1 and 4.0 μm features will be feasible with JWST.

### 4. Evidence of Nitrogen Chemistry in Known Hot Jupiters

Having identified prominent nitrogen chemistry absorption features, we present tentative signs of these in three hot Jupiter atmospheres. We apply a uniform atmospheric retrieval analysis to nine hot Jupiter spectra, spanning visible to near-infrared wavelengths (∼0.3–5.0 μm). After briefly describing our planet selection, we examine the extent to which current observations can constrain nitrogen chemistry in exoplanetary atmospheres.

#### 4.1. Planet Selection

We focus on the Sing et al. (2016) hot Jupiter transmission spectra with WFC3, STIS, and Spitzer observations: WASP-12b, WASP-17b, WASP-19b, WASP-31b, HAT-P-1b, HAT-P-12b, HD 189733b, and HD 209458b. We also retrieve the WFC3 spectrum of WASP-63b (Kilpatrick et al. 2017), where indications of HCN have been considered. Our goal is to identify planets with plausible suggestions of nitrogen chemistry, such that follow-up observations can robustly establish whether nitrogen chemistry is present in these objects. An initial retrieval was performed for each planet including all model parameters. Candidates were identified wherever the best-fitting retrieved spectrum featured hydrogen absorption features for a WFC3 transit depth excess due to NH3 or HCN >30 ppm. This resulted in three candidates: WASP-31b, WAP-63b, and HD 209458b. We provide the posterior distributions from these retrievals online at doi:10.5281/zenodo.1014847. For each candidate, we
ran three additional retrievals: with NH₃ (no HCN), with HCN (no NH₃), and without nitrogen chemistry.

4.2. Inferences of Nitrogen Chemistry

Figure 3 displays the best-fitting spectra from retrievals with and without nitrogen chemistry from 1 to 5 μm. WASP-31b and WASP-63b feature large nitrogen chemistry transit depth excesses: ∼400 ppm NH₃ (WASP-31b) and ∼200 ppm HCN (WASP-63b) at ∼1.55 μm (Figure 4). HD 209458b has an ∼50 ppm NH₃ transit depth excess.

Uniquely identifying nitrogen-bearing species is challenging at the resolution and precision of present WFC3 observations, given overlapping NH₃ and HCN absorption features. This difficulty is particularly apparent for HD 209458b, as shown in MacDonald & Madhusudhan (2017). Moreover, in the present work we report more conservative estimates of evidence for nitrogen chemistry by marginalizing over the cloud fraction. We also utilize higher-resolution cross-sections (0.1 cm⁻¹) and Spitzer observations. As such, the significance of nitrogen chemistry in HD 209458b is lower than in MacDonald & Madhusudhan (2017). However, for moderate NH₃ or HCN mixing ratios relative to H₂O, nitrogen signatures become sufficiently strong to permit unique detections. This is the case for both WASP-31b and WASP-63b, where strong WFC3
features permit unique identification of signatures attributable respectively to NH\textsubscript{3} and HCN (Figure 4).

We report a weak detection of NH\textsubscript{3} in WASP-31b (2.2\,$\sigma$). A nested model comparison, whereby we computed the Bayesian evidences of retrievals with NH\textsubscript{3} + HCN and without NH\textsubscript{3}, establishes a Bayes factor of 3.8 for NH\textsubscript{3}; this uniquely identifies it as the cause of the $\sim$400 ppm WFC3 feature around 1.5\,$\mu$m. Previous studies of WASP-31b were unable to fit this feature, either due to excluding NH\textsubscript{3} (Sing et al. 2015; Barstow et al. 2017) or assuming chemical equilibrium (Sing et al. 2016). Our retrieval without NH\textsubscript{3} (Figure 3, blue) similarly struggles to fit these elevated points. We predict a $\sim$500 ppm K-band NH\textsubscript{3} feature for this planet (Figure 3). If confirmed, this represents the first inference of ammonia in an exoplanetary atmosphere.

We further reassert a weak detection of HCN in WASP-63b (2.3\,$\sigma$, Bayes factor = 4.7), due to a $\sim$200 ppm peak around 1.55\,$\mu$m. We predict a $\sim$400 ppm feature near 3.1\,$\mu$m and $\sim$200 ppm absorption near 4.0\,$\mu$m (Figure 3). These detection significances include integration over the entire parameter space, including inhomogeneous clouds; thus the transmission spectra of WASP-31b and WASP-63b cannot be adequately fit without disequilibrium nitrogen chemistry.

Derived mixing ratio posteriors for NH\textsubscript{3} and HCN are shown in Figure 5. The maximum a posteriori modes show abundances enhanced by $\sim$3–4 orders of magnitude over equilibrium...
expectations for WASP-31b and WASP-63b, and ∼1 order of magnitude for HD 209458b.

4.3. Resolving Degenerate Solutions

The limited wavelength range of current observations permits a range of possibilities. This is especially true for WASP-63b, where the lack of Spitzer or optical data precludes determining the spectral continuum (Figure 3). With low resolution or limited precision data, retrievals have flexibility in adjusting other parameters to partially compensate for removing NH$_3$ or HCN. For example, molecular abundances can be degenerate with terminator cloud coverage. Such degenerate solutions cause the mixing ratio “tails” in Figure 5. However, present observations are sufficient to distinguish NH$_3$/HCN features from CH$_4$, due to a lack of absorption at ∼1.7 μm.

Despite WFC3 degeneracies, Figure 3 indicates that model differences arise at longer wavelengths. Observing WASP-31b in the K-band and WASP-63b with Spitzer will permit tighter constraints on their NH$_3$ and HCN abundances. HD 209458b is more challenging, as the low inferred NH$_3$ abundance only predicts ∼25 ppm K-band absorption. Ultimately, observations in the K-band and at 3.1 or 4.0 μm are critical to resolving model degeneracies.

5. Summary and Discussion

Nitrogen chemistry will open a new window into disequilibrium atmospheric chemistry and planetary formation mechanisms. High NH$_3$ abundances are indicative of vertical mixing, with abundance measurements constraining the eddy diffusion coefficient (Moses et al. 2011). High HCN abundances can also indicate vertical mixing, enhanced C/O, or, through an absence of CH$_4$ and NH$_3$, photochemistry (Zahnle et al. 2009; Moses et al. 2011; Venot et al. 2012).

We have demonstrated that nitrogen-bearing molecules can be observed in WFC3 spectra. We identified a ∼400 ppm NH$_3$ feature in WASP-31b (2.2σ), and a ∼200 ppm HCN feature in WASP-63b (2.3σ). Nitrogen chemistry is potentially present on HD 209458b; though current WFC3 observations are insufficient to definitively identify a specific species, given overlapping NH$_3$ and HCN features. Ambiguities may be resolved by observing strong NH$_3$ absorption at ∼2.2 μm (K-band) and strong HCN absorption at ∼3.1 and 4.0 μm. JWST
will be ideally suited to observing the plethora of features exceeding the \( \sim 10 \text{ ppm} \) precision expected of NIRISS/ NIRSpec (Beichman et al. 2014). Such observations will enable unique detections of NH\(_3\) and HCN in many exoplanetary atmospheres.

Observable nitrogen chemistry signatures result when NH\(_3\) or HCN exceed \( \sim 10^{-2} \times \) the H\(_2\)O mixing ratio. HCN features at \( \sim 3.1 \) and 4.0 \( \mu \text{m} \) weaken and become sharply peaked for lower temperatures, while most NH\(_3\) features, especially in the WFC3 bandpass, strengthen and remain broad. Extensively cloudy atmospheres have dampened absorption features, though some can exceed \( \sim 100 \text{ ppm} \) even for uniform clouds at 1 mbar.

Our inferred NH\(_3\) and HCN abundances are enhanced over equilibrium values by \( \sim 3 \)–4 orders of magnitude. Such high values suggest that chemical equilibrium is violated in hot Jupiter atmospheres, and should not be imposed a priori in atmospheric retrievals. Though more work is needed to explore scenarios producing enhanced NH\(_3\) or HCN, the unexpected should be embraced, not shunned, as we seek to elucidate the nature of these worlds.

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