



The Diversity of Planetary Atmospheric Chemistry

Lessons and Challenges from Our Solar System and Extrasolar Planets

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Abstract Atmospheres in our solar system range from oxidizing to reducing, transient to dense, veiled by clouds and hazes to transparent. Observations already suggest that exoplanets exhibit an even more diverse range of atmospheric chemistry and composition. Nevertheless, there are commonalities across the atmospheres of our solar system that provide valuable guidance and lessons for observing and interpreting exoplanetary atmospheres. Lessons gleaned from decades of study of planetary atmospheric chemistry are synthesized and explored to understand their implications for exoplanets.

Keywords Atmospheric chemistry · Chemical modeling · Terrestrial planets · Giant planets · Titan · Exoplanets

Understanding the Diversity of Planetary Atmospheres
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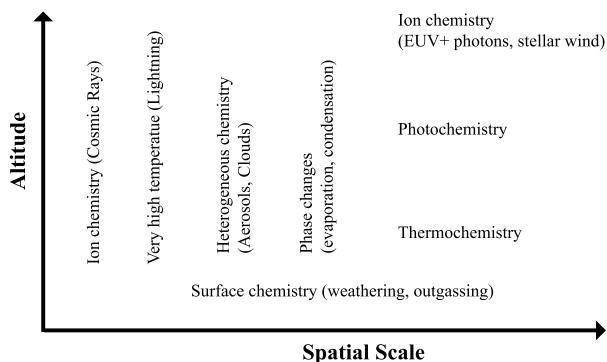
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Fig. 1 Typical altitudes and spatial scales for the types of chemistry occurring in a planetary atmosphere



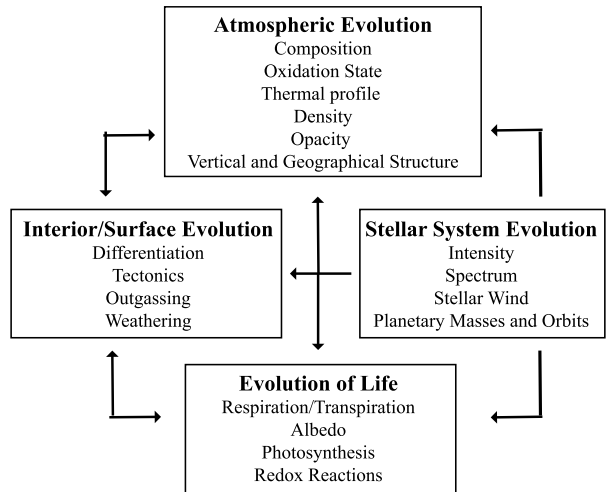
1 Introduction

Atmospheric chemistry is complex due, in part, to its sensitivity to trace species whose relative abundances may be a few parts per billion (ppb) or parts per trillion (ppt) (Finlayson-Pitts and Pitts 2000, Chap. 2). This sensitivity to trace species arises from catalytic cycles in which the reactive radical species may regenerate many times before being lost to slower reactions (Jacob 1999, Chap. 9).

Many types of chemistry can occur in an atmosphere depending on the conditions present, Fig. 1. At the highest altitudes (lowest pressures), ions produced by the interaction of atmospheric constituents with extreme ultraviolet (EUV) and X-ray photons and solar wind particles form the ionosphere (Chamberlain and Hunten 1987, Chap. 5). Reactions between ions and neutrals occur readily (Yung and DeMore 1999, Chap. 3). At lower altitudes, transient positive and negative ions can be produced by galactic and stellar/solar cosmic rays, stellar/solar mass ejection events, and lightning (Dorman 2004, 2009; Nordheim et al. 2015; Yung and DeMore 1999, Chaps. 2, 4, 9). These ions can have a significant local effect on atmospheric chemistry (Rusch et al. 1981; Solomon et al. 1981), while their broader import depends on the interaction of chemistry with mesoscale to synoptic scale circulation. Cosmic rays and negative ions also have been proposed as mechanisms for initiating the production of aerosols (Enghoff and Svensmark 2008). Below the penetration depth of EUV photons, neutral photochemistry dominates. In this region, photons, particularly at far ultraviolet (FUV) to near ultraviolet (NUV) wavelengths but extending into the near infrared (NIR), dissociate molecules and excite atoms/molecules into more energetic, more reactive states (Chamberlain and Hunten 1987, Chap. 3; Yung and DeMore 1999, Chaps. 2, 3; Jacob 1999, Chaps. 9, 10, 11) Photochemistry plays an extremely important role in determining the (observable) composition of an atmosphere because it drives the composition away from the thermochemical equilibrium state (DeMore and Yung 1982).

Aerosols and cloud particles affect composition and chemistry directly through condensation and evaporation. In some atmospheres clouds are globally ubiquitous, while in others they are patchy or dependent on season (de Pater and Lissauer 2010, Chap. 4). Clouds typically form layers near the condensation level for a condensate species, and the composition of the gaseous atmosphere above and below the cloud layer differs significantly. Clouds and hazes can indirectly affect atmospheric chemistry by limiting the depth of penetration of photolyzing radiation within an atmosphere. Aerosols and clouds also affect chemistry by providing a medium within or on which heterogeneous chemistry may occur (Finlayson-Pitts and Pitts 2000, Chaps. 5, 12). Heterogeneous reactions typically have a lower activation barrier so they can catalyze processes that would be much slower in gas-phase. Convection

Fig. 2 Interactions among the components of the planet-star system that influence the mutual evolution of the system's components and determine the observable composition of the atmosphere



and advection of air parcels that entrain particles is one mechanism for generating sufficient static charge separation to induce lightning discharges. A lightning discharge can generate temperatures of 30,000–40,000 K (Orville 1968; Price et al. 1997) but this is followed by rapid cooling as the parcel adiabatically expands and the composition of the air parcel continues to equilibrate until the quenching temperature (~ 1000 K) is reached (Helling and Rimmer 2019; Yung and DeMore 1999, Chap. 2). Lightning effects are local but transport, particularly mesoscale convection, can extend the effects over one to two scale heights. At altitudes below the typical penetration depth for UV-visible photons, increasing atmospheric temperatures enable thermal equilibrium chemistry to become increasingly important (Yung and DeMore 1999, Chap. 3).

At the base of the atmosphere, if there is a solid or liquid surface, longer-time-scale chemistry, such as weathering, occurs via reactions with that surface which will change the composition of both the surface and the atmosphere. Gas exchange between surface liquid and the atmosphere can occur as well as outgassing from and subduction/burial in the interior, and all can change or sustain the bulk composition of the atmosphere (Fegley et al. 1997; Jacob 1999, Chap. 6).

Atmospheric composition evolves over time as the parent star and the planet's interior and surface evolve (Kasting and Ono 2006), Fig. 2. Over time, a star's spectrum and intensity change and, consequently, the energy available at each altitude in a planetary atmosphere changes with time. Outgassing from a planet's interior can change with time and lead to significant changes in the composition of the planet's atmosphere as the planet evolves. Likewise, burial, surface reactions, atmospheric escape, cometary and meteoritic impacts, and the influx of interplanetary dust can alter the composition and redox state of an atmosphere (Catling and Kasting 2017, Chaps. 3, 5) (Grebowsky et al. 2002; Wordsworth et al. 2018). Our initial views of extrasolar planetary atmospheres will be snapshots in time of systems undergoing changes on a broad range of temporal and spatial scales.

Further complexity in interpreting observations arises from the indirect relationship between an atmosphere's remote observables and its bulk atmospheric and planetary compositions. Clouds and refraction are obvious barriers to determining an atmosphere's composition from remote observations. In addition, the dominant chemistry in an atmosphere can change significantly with altitude. One example of compositional change is that above and

below a cloud layer. An example of process change is the transition from ion photochemistry to neutral photochemistry to thermal equilibrium chemistry as the stellar photons available to drive atmospheric chemistry decrease in energy with increasing depth in an atmosphere.

Recognition of the complexity of atmospheric chemistry and the importance of integrating across processes and altitude regions, in combination with increasing computing capability and an absence of constraining data, has shaped ongoing research on the atmospheric chemistry of extrasolar planets. An extremely diverse range of potential conditions has been explored, partially in response to the diversity of astrophysical conditions in which extrasolar planets have been found, including conditions with no analogs in our solar system. Several 3D models integrating atmospheric chemistry, aerosol and cloud microphysics, radiative transfer, and synoptic-scale dynamics have been developed based on terrestrial climate models (e.g., Yamamoto and Takahashi 2008; Lebonnois et al. 2010; Way et al. 2017), and whole planet 1D parameterized models that integrate interior, surface, atmospheric, and exospheric processes are in development (e.g., Barnes et al. 2020). The benchmarks used to assess the accuracy of these models are the planets in our solar system. Temporal variability and evolution in atmospheric conditions are especially important for assessing potential habitability, and paleoclimate simulations, constrained by data, have guided work in this area.

2 Modelling Tools and Their Uses

Most atmospheric chemistry studies have used one-dimensional models intended to represent global-average conditions. Well-constrained, carefully-interpreted one-dimensional simulations can provide significant insight into complex processes, and global-average simulations can shed light on large-scale chemistry, temporal variability, and observational differences (see Fig. 3). However, global-average one-dimensional simulations are biased towards day-side chemistry and do not usually represent the solution one would get from averaging conditions over all longitudes and latitudes around a planet. Figure 4 shows the similarity between calculations for Venus' mesosphere using a diurnally-averaged solar radiation field and calculations using a global-average one ("global-average" = 0.5 times the solar radiation field at solar zenith angle of 45°). Observed nightside abundances and ratios for SO and SO₂ are significantly different from those found on Venus' dayside (e.g., Sandor et al. 2010). Time-variable one-dimensional models can be used to track diurnal or seasonal variations or to predict zonally-averaged photochemical behavior as a function of latitude and season (e.g., Moses and Greathouse 2005; Hue et al. 2015, 2016; Moses et al. 2018) (Fig. 5), but such models neglect the influence of horizontal transport and advection. One-dimensional simulations in general do not adequately capture the significant impacts of circulation on species distributions and chemistry (Forget and Lebonnois 2013).

Three-dimensional general circulation models (GCMs) that incorporate interactive chemistry or effective chemical relaxation times are emerging as computational power has increased (Bougher et al. 1988; Lebonnois et al. 2001; Lefèvre et al. 2004; Cooper and Showman 2006; Drummond et al. 2018a,b, 2020; Mendonça et al. 2018) (Fig. 6). By necessity, the chemistry that can be incorporated into these models is simplified, so it is important to assess the uncertainties in these simulations that arise from the simplified chemical schemes (Dobrijevic et al. 2011; Tsai et al. 2018). This can be done with a lower-dimension chemistry model with more complete chemistry or a combination of a chemical transport model (CTM) and a GCM. Interpretation of atmospheric chemistry simulations using a GCM is further complicated by the need to consider the impacts of imperfect dynamics on the calculated distributions of chemical species (Brasseur et al. 1999; Yung et al. 2009).

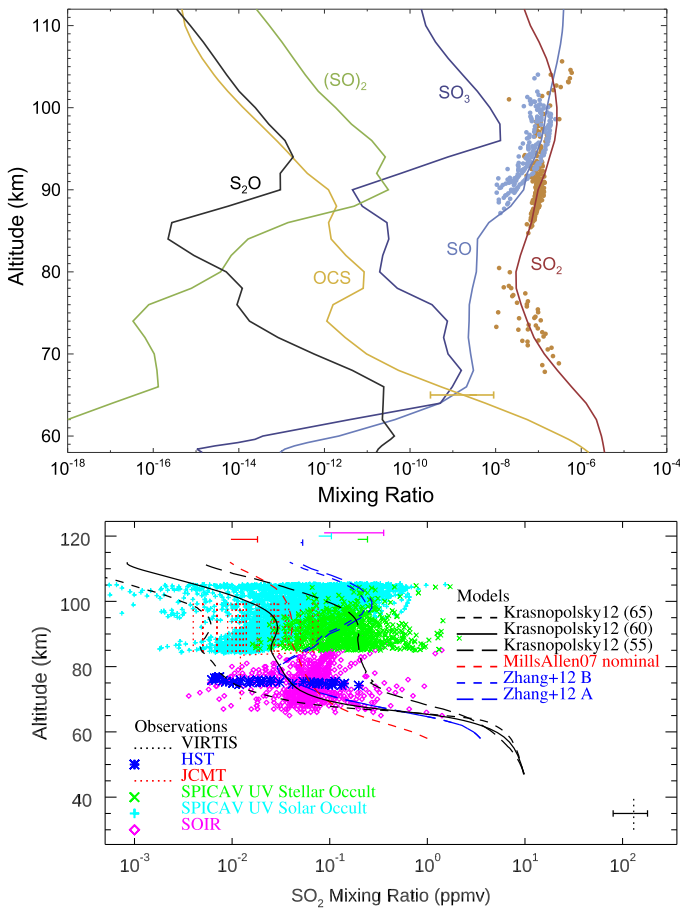


Fig. 3 (a) Calculated volume mixing ratios of sulfur oxides on Venus from a one-dimensional photochemical model, in comparison with observations of SO_2 , SO , and OCS (from Zhang et al. 2012). (b) Observations of and model profiles for SO_2 on Venus (updated from Mills et al. 2019). At least some observed differences are likely to be temporal or spatial variability. Magenta diamonds are IR solar occultation (terminator) observations by Solar Occultation at Infrared (SOIR) on *Venus Express* (Mahieux et al. 2015); cyan plus signs are UV solar occultations (terminator) by Spectroscopy for Investigation of Characteristics of the Atmosphere of Venus (SPICAV) on *Venus Express* (Belyaev et al. 2017); green Xs are UV stellar occultations (nightside) by SPICAV (Evdokimova et al. 2020); blue asterisks are Hubble Space Telescope (HST) UV observations (Jessup et al. 2015); and red vertical dotted lines are James Clerk Maxwell Telescope (JCMT) submm observations above 84 km with an indicative upper limit below 84 km. Tropospheric abundance (black dotted line) is from Visible and Infrared Thermal Imaging Spectrometer (VIRTIS) on *Venus Express* (Marcq et al. 2008). Typical uncertainties are indicated by the half-error bars at the top. Blue long-dash and short-dash lines are models A and B, respectively, from Zhang et al. (2012). Red short-dash line is the nominal model from Mills and Allen (2007). Black long-dash, solid and short-dash lines are models with eddy diffusion break at 55, 60 and 65 km, respectively, from Krasnopolsky (2012)

An intermediate class of models that historically was very important in understanding terrestrial atmospheric chemistry has seen limited application in extraterrestrial studies. Specifically, two-dimensional and three-dimensional CTMs, typically with transport derived from GCM simulations (Yung et al. 2009). For most atmospheres, only a few species' distributions directly influence the thermal structure and dynamics of the atmosphere. Potential

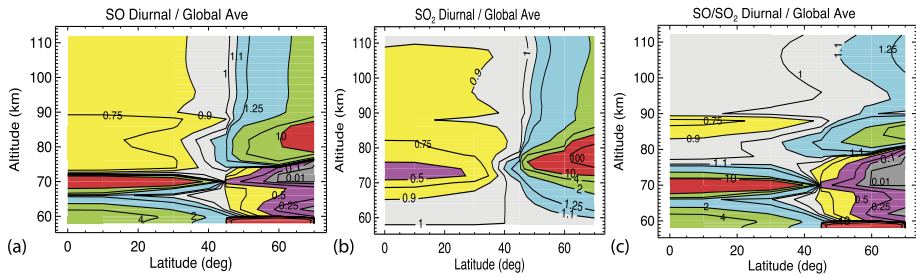
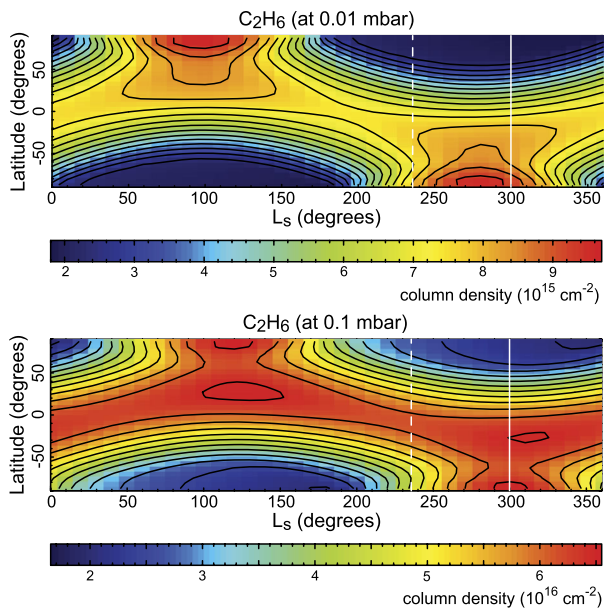


Fig. 4 Comparison of diurnal-average and global-average calculations for **(a)** SO, **(b)** SO₂, and **(c)** SO/SO₂ on Venus as functions of latitude and altitude. The chemistry used is the same as was used for the calculations in Jessup et al. (2015). Contours are at 0.01, 0.1, 0.25, 0.5, 0.75, 0.9, 1, 1.1, 1.25, 2, 4, 10, and 100

Fig. 5 Predicted ethane column density above 0.01 mbar (Top) and 0.1 mbar (Bottom) as a function of latitude and season (represented by solar longitude L_s) on Neptune from a time-variable one-dimensional photochemical model (from Moses et al. 2018)



distributions of other species, including potentially observable species, can be explored accurately in a CTM. For observing scenarios where local time or longitudinal variations in chemistry are important, however, two-dimensional CTMs may not provide appropriate results.

Thermochemical/photochemical kinetics and transport models in which all the chemical reactions are fully reversed have become the de facto standard for simulating giant exoplanet atmospheres (e.g., Moses et al. 2011; Venot et al. 2012; Tsai et al. 2017). These models track atmospheric photochemistry in a traditional way, but they are also capable of reproducing thermochemical equilibrium in regions in which high temperatures allow reactions to proceed efficiently in both directions. Such models also predict the disequilibrium quenched abundances that occur when transport time scales drop below the chemical kinetic conversion time scales between different molecular forms of key elements. Quenching due to vertical transport is expected to be very important on exoplanets with thick, deep, hot atmospheres. Horizontal quenching may also be important on tidally locked transiting ex-

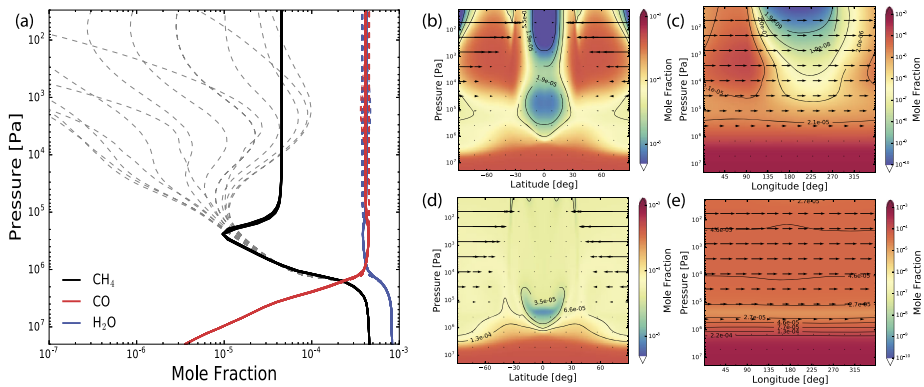


Fig. 6 From Drummond et al. (2018a). **(a)** Vertical profiles of the carbon monoxide (red), water (blue), and methane (black/gray) mole fractions for a number of columns equally spaced in longitude around the equator for chemical equilibrium (dashed) and non-equilibrium (solid) simulations. The meridional flow increases the abundance of methane toward the equator and vertical mixing determines the quenched abundance above $P \sim 105$ Pa **(b–e)** Mole fractions (color scale and black contours) of methane and wind velocity vectors (black arrows) **(b)** for chemical equilibrium at a longitude of 0° , **(c)** for chemical equilibrium as an area-weighted meridional mean ($\pm 20^\circ$ latitude) around the equator, **(d)** for a non-equilibrium simulation at a longitude of 0° , and **(e)** for a non-equilibrium simulation as an area-weighted meridional mean ($\pm 20^\circ$ latitude) around the equator

oplanets, and pseudo-2D models have been developed to track the longitudinal behavior of hot Jupiter exoplanets as a result of both horizontal and vertical quenching (Agúndez et al. 2014; Venot et al. 2020; Moses et al. 2021).

Atmospheric chemistry models are used for many purposes. As a guide to observations and a theoretical assessment of potential planetary diversity, models can be used to explore and speculate on potential compositions, structures, and observable signatures. The range of parameter space that can be explored is extensive, so one-dimensional global-average models that are relatively cheap computationally are by far the most common option. Higher-dimensional models can only be used for highly selective exploration. Chemistry models are commonly used for interpretation of observations, but the number of observables is almost always far fewer than the number of model parameters, so simulations are highly underconstrained. Analysis of species abundance ratios or abundances of multiple species within a chemical family can reduce the complexity of the system and aid in identifying the chemical regime (e.g., Sandor et al. 2010). Other uses of models include comparing observations and laboratory data (e.g., Sander et al. 1989; Nair et al. 1994), identifying gaps in existing laboratory data (e.g., McElroy et al. 1986; Yung et al. 1984), and assessing consistency among observations (e.g., Mills 1999). Simulations of previously untested reactions and pathways have played significant roles in guiding laboratory and now *ab initio* studies (e.g., Yung and DeMore 1982; Mills and Allen 2007). For atmospheres where both remote sensing and *in situ* data are available, chemical models provide checks on the consistency between these types of data. Two final uses are assessing the sensitivity of simulations and observations and assessing uncertainties in simulations (e.g., Stolarski et al. 1978). Terrestrial modeling studies of stratospheric ozone and air pollution sometimes consider the covariance among multiple parameters (e.g., Fleming et al. 2015; Aleksankina et al. 2019). Non-terrestrial studies largely still consider only univariate variance, when they consider uncertainties.

Numerous significant surprises from studies of atmospheric chemistry within our solar system and the large diversity of potential conditions on extrasolar planets make it neces-

sary to consider large regions of parameter space for extrasolar planetary atmospheres, particularly when assessing the prevalence of biosignature false positives. This has prompted innovations in modeling techniques but 2D and 3D CTMs have been under-utilized to date. Fully reversed kinetics has been adopted in simulations of giant exoplanets and hot-Jupiter exoplanets but has not been widely adopted yet for terrestrial-type (e.g., Venus-like) extrasolar planets. Uncertainty, likewise, has been only partially considered in atmospheric chemistry studies for extrasolar planets. The incomplete nature of atmospheric composition observations and observational uncertainties are incorporated in assessments of observability, but laboratory uncertainties and the potential importance of unstudied chemistry have not. The inherently empirical approach of atmospheric chemistry research has meant that interaction/iteration among observations, simulations, and laboratory studies has been critical for understanding the chemical processes occurring in planetary atmospheres in our solar system.

3 Factors Affecting Observability

The extent of the detectable portion of a planetary atmosphere depends on the observing technique, but many factors can cause the composition of the detectable portion of the atmosphere to differ from that in the lower, denser portion of the atmosphere. In addition, gas-phase features in remotely observed spectra of a planetary atmosphere are determined by the product of the column abundance of a gas and its absorption cross section. Consequently, many of the prominent features in planetary atmospheric spectra are produced by the most radiatively active trace gases rather than the most abundant gases. These observational selection effects and the importance of trace catalytic species significantly complicates interpretation of spectra of extrasolar planetary atmospheres, as do degeneracies between the abundance of constituents and atmospheric temperatures or other properties that also affect spectral signatures.

Vertical temperature gradients in an atmosphere accentuate spectral features. Molecules residing within a cold region overlying a warmer continuum region show up in absorption, while temperature inversions can lead to emission features. Extended isothermal regions mute spectral features, complicating compositional analyses.

Clouds and hazes form obvious barriers to photons, but details of their structure and composition strongly influence the resultant spectrum (Liou 1992). Cloud and haze particles can affect the strength of gas-phase spectral features, and the size and shape of the particles can affect the wavelength dependence of the scattering and absorption (Twomey 1977). If the single scattering albedo of the cloud particles is high (the imaginary index of refraction for the particles is small) and gaseous absorption at a wavelength is weak, then nadir-viewing observations can be sensitive to scattered stellar light and thermal radiation emitted from deep in the atmosphere, both of which can provide information on atmospheric composition below or deep within cloud layers with scattering optical depths > 10 (e.g., Allen and Crawford 1984; Mills 1999).

Above the homopause, high altitude haze can form and obscure lower altitudes when the parent gas has a smaller molecular weight than the primary constituent of the atmosphere. On Titan, for example, as illustrated in Fig. 7, methane has a smaller molecular weight than N_2 so the methane abundance increases with altitude above the homopause, which means a significant abundance of methane can react with incident UV photons and ions to form a high-altitude optically thick hydrocarbon haze whose particles settle gravitationally to lower altitudes (e.g., Waite et al. 2007). On the giant planets in our solar system, by contrast,

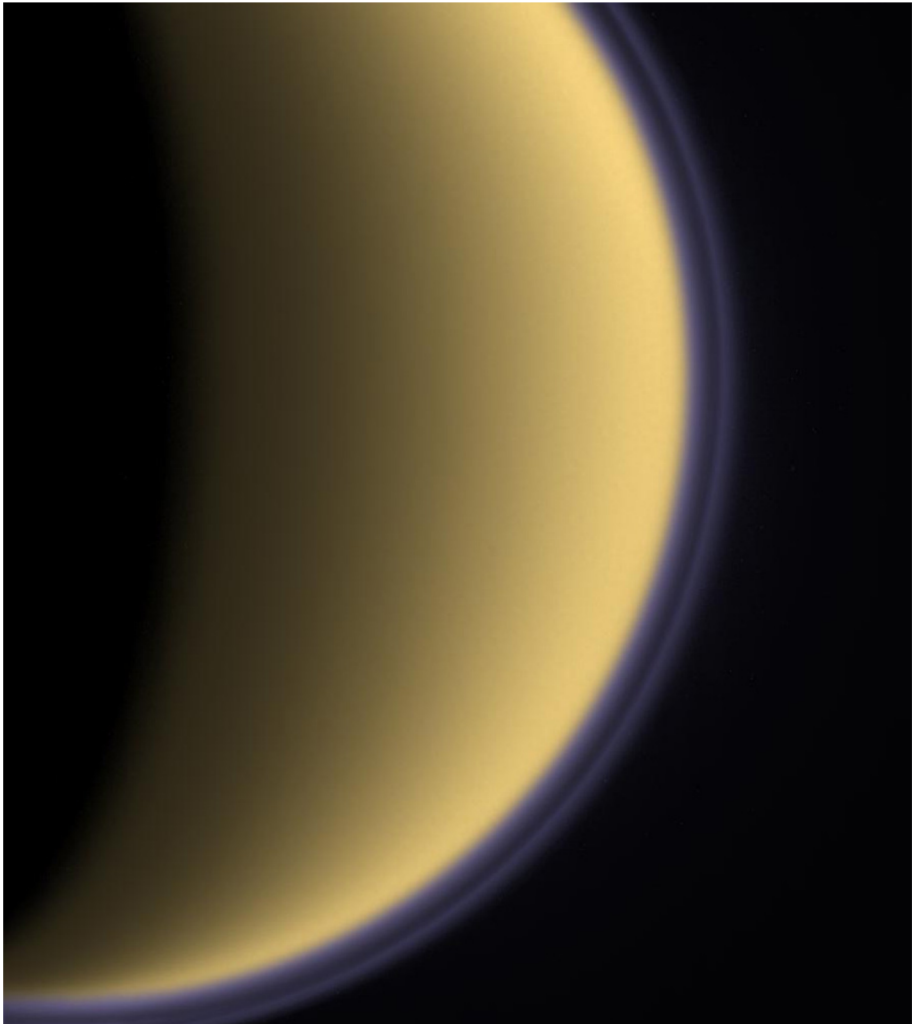


Fig. 7 Cassini narrow-angle camera false color image taken on 3 July 2004 in a filter centered at 338 nm showing Titan's atmosphere shrouded by photochemical hazes. Credit: NASA/JPL-Caltech/Space Science Institute

methane has a larger molecular weight than the primary constituent, H_2 , so the hazes formed from photochemical products of methane occur at lower altitudes, have less dependence on ion chemistry, and tend to be optically thin (e.g., Zhang et al. 2013).

Clouds also mark a significant transition in composition, as the gas-phase concentration of the condensable species decreases by orders of magnitude within the cloud, Fig. 8. The cold trap at Earth's tropopause is crucial for life as it keeps water vapor near the surface where it is protected against photolysis followed by hydrogen escape (Wordsworth and Pierrehumbert 2014). Similarly, condensation on hazes can strongly impact the gas-phase abundances of species. When condensation occurs at depth, it removes species and potentially prevents some chemical elements from appearing in the observable region of a planet's

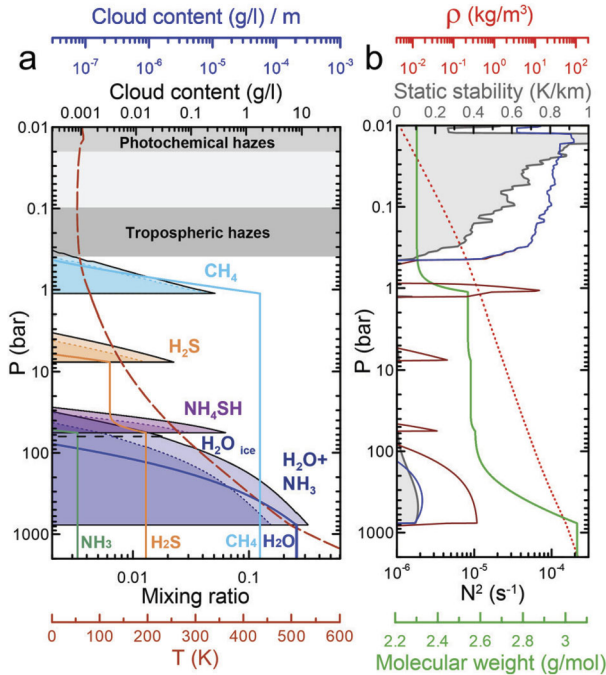
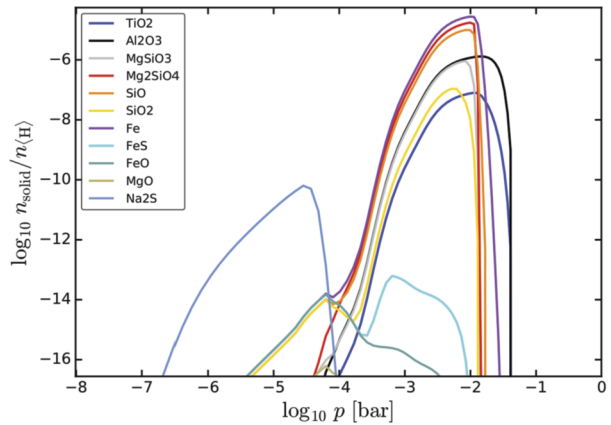


Fig. 8 Illustration of cloud structure and corresponding gas mixing ratios on Uranus from thermochemical equilibrium arguments (from Hueso and Sánchez-Lavega 2019). **(a)** Volatiles mixing ratio (colored lines, lower axis) and maximum cloud density (black lines with shaded regions, upper axis) for 30 times solar abundance for all condensables except for ammonia, which is considered depleted with 3 times solar abundance. A horizontal dashed line shows the transition from the water ice cloud to liquid water with the capability to dissolve ammonia. The red-brown dashed line shows the vertical temperature profile of the atmosphere extrapolated from the Voyager 2 temperature profile following a moist adiabat (bottom axis). Cloud densities (continuous and dotted color lines) are given in g/l per meter (top blue axis). **(b)** Stability of the atmosphere towards convection in terms of the Brunt-Vaisalla frequency, N^2 , and the atmospheric static stability (grey line with shaded region, upper axis). The mean molecular weight of the atmosphere is also shown (green line, bottom axis). The contribution of the thermal structure of the atmosphere to the Brunt-Vaisalla frequency is shown with a blue line. The total Brunt-Vaisalla frequency, incorporating the effects of the vertical gradient of molecular weight, is shown with a red line. The red dotted line shows the atmospheric density

atmosphere (e.g., Marley et al. 2013). Considering the wide range of thermal structures of exoplanets and brown dwarfs, the number of potential condensates rapidly increases. Microphysics models that reveal cloud composition and condensation sequence can provide context for observations, Fig. 9. Condensation also significantly alters relative isotopic abundances, particularly for H and D (e.g., Merlivat and Nief 1967). The D/H ratio in water below and above Venus' cloud layers, for example, is 120–135 and 150–240 times larger than Vienna Standard Mean Ocean Water (VSMOW, D/H = 155.76 ± 0.1 ppm), respectively (Marcq et al. 2018). The isotopic fractionations of water in Earth's upper troposphere and lower stratosphere are $\delta D \sim -400$ – -450 and -600 per mil, respectively (Randel et al. 2012).

Other chemical processes can alter the effects of condensation. An example is water in the Earth's atmosphere. Water's gas-phase abundance in the troposphere, well below the tropopause cold trap, is 0–4%. At the tropical tropopause, its abundance is ~ 3 –4 ppm due to condensation as air is transported upward (e.g., Randel et al. 2012; Oman et al. 2008). In the

Fig. 9 Concentrations of condensed species in a model of a hot-Jupiter with $T_{eff} = 1300$ K and $\log g = 3$, showing a secondary cloud layer almost entirely made of di-sodium sulfide $\text{Na}_2\text{S}(s)$ (from Woitke et al. 2020)



middle to upper stratosphere at midlatitudes, however, the gas-phase abundance of water has increased to ~ 6 ppm as a result of photochemistry in the stratosphere converting methane into carbon dioxide and water after the methane has been transported upward through the tropopause (e.g., Randel et al. 2012; Oman et al. 2008).

A ternary system (binary solution), such as $\text{H}_2\text{O}-\text{SO}_2-\text{H}_2\text{SO}_4$, that includes a readily condensible species can undergo bifurcation. In a sufficiently oxidizing environment, like the middle atmospheres of Venus and Earth, SO_2 and H_2O will relatively quickly react to produce H_2SO_4 . H_2SO_4 acts as a net sink for gas-phase SO_2 and H_2O when it condenses. This can result in the near-complete loss of either H_2O or SO_2 , whichever is less abundant, as seen, for example, in the Earth's stratosphere and mesosphere (e.g., Hamill et al. 1977; Mills et al. 2005) and in the Venus upper cloud (Parkinson et al. 2015; Encrenaz et al. 2016, 2019, 2020; Shao et al. 2020). Consequently, temporal variations in the $\text{H}_2\text{O}/\text{SO}_2$ ratio and mesoscale convection can lead to significant, observable differences in the abundances of H_2O and SO_2 . In Earth's atmosphere some common binary and ternary solutions include $\text{H}_2\text{O}/\text{SO}_2/\text{H}_2\text{SO}_4$, $\text{NH}_3/\text{H}_2\text{SO}_4/(\text{NH}_4)_2\text{SO}_4$, and $\text{HNO}_3/\text{H}_2\text{O}$. On the giant planets, an example is $\text{NH}_3/\text{H}_2\text{S}/\text{NH}_4\text{SH}$.

Vertical mixing can also have a significant impact on both the diversity of species observed and the strength of observed spectral features. For example, the strong vertical atmospheric mixing on Neptune allows methane to be carried to higher altitudes, where it is exposed to UV radiation, leading to the generation of hydrocarbon photochemical products (Moses et al. 2018). The much weaker mixing in the stratosphere of Uranus confines the methane to significantly lower altitudes. Even though photochemical products are transported downward more rapidly when vertical mixing is stronger, the stable photochemical products on Neptune are present over a much greater column of atmosphere than on Uranus, so the resultant planetary spectrum has stronger features. In fact, for our solar-system giant planets, the strength of vertical mixing has a stronger influence on the abundance of their hydrocarbon photochemical products than incident solar flux or any other single factor (Moses et al. 2005, 2018). Similarly, advection and global circulation have significant impacts on the detectable composition of an atmosphere and on chemical processes. Terrestrial stratospheric ozone abundances, for example, are highest at the poles as a result of the Brewer-Dobson circulation even though ozone is primarily produced at equatorial latitudes (e.g., Butchart 2014). A similar pole-to-pole circulation pattern on Mars produces nightglow in the $\text{O}_2(a^1\Delta - X^3\Sigma)$ band in the downward, converging branch near the winter pole (e.g., Fedorova et al. 2012; Bertaux et al. 2012). Intense airglow on Venus is concentrated near

the antisolar point as a result of the thermospheric subsolar to antisolar circulation (e.g., Bougher and Borucki 1994; Soret et al. 2014).

It is also important to consider that atmospheres are inherently three dimensional. Interpretation of disc-averaged spectra typically relies on one-dimensional radiative transfer modeling which implicitly assumes atmospheric conditions are homogeneous across the entire visible hemisphere. A recent study examining both simulated observations derived from a GCM and real observations of WASP-43b found (a) significant differences between standard 1D and new 2.5D retrievals that consider geographic (local time) variations in vertical profiles of temperature and constituent abundances, (b) considerable degeneracy between temperature and gaseous abundances, and (c) significant differences in inferred atmospheric structure, including a potential nightside cloud layer (Irwin et al. 2020). A companion study found geographic (local time) inhomogeneities in atmospheric temperature profiles can lead to significantly biased and/or spurious molecular detections in a 1D retrieval that assumes a geographically homogenized atmosphere (Taylor et al. 2020). The potential for spurious molecular detections increased when retrievals considered larger numbers of species (Taylor et al. 2020). These results suggest that a hierarchy of models will be required for not only simulating exoplanet atmospheres but also for interpreting observations.

As discussed above, studies of atmospheres in our solar system have identified many complex factors that can affect the observable composition of an atmosphere and, consequently, introduce significant ambiguity into interpretation of observations of extrasolar planetary atmospheres. Interpretation, thus, will require as much contextual information as possible and models that incorporate as many of the factors discussed above as possible. Organization and conceptualization of atmospheric chemistry in terms of chemical cycles that operate within specified environmental conditions on any planet will help with interpretation of ongoing simulations and forthcoming observations.

4 Important Chemical Cycles

Analysis of atmospheric chemistry to understand the primary processes occurring in a solar system planet's atmosphere, to assess their potential relevance for exoplanets, and to identify their impact on potential observables requires organizing reactions into chemical cycles that define pathways for production and loss of potential atmospheric observables. Although the radical species that lie at the heart of chemical cycles generally have abundances which are too small to observe remotely, studies of solar system atmospheres suggests the presence or absence of particular chemical cycles can be inferred with reasonable confidence.

Key areas in which chemical cycles are important for understanding planetary atmospheres are their (a) impact on bulk composition, (b) net effect on an atmosphere's remotely observable spectrum, (c) influence on atmospheric evolution and stability, (d) mutual influence and feedbacks on large and mesoscale circulation and thermal structure, and (e) connection with habitability. The importance of each area differs for each type of planet and research objective. For terrestrial-type (rocky) planets, key interests have been clouds, linking upper (observable) and lower (not readily observable) atmospheric compositions, habitability, planetary evolution (and climate change), biosignatures, chemistry-climate couplings, and the chemistry of oxides of nitrogen, hydrogen, chlorine, and sulfur. For CO₂-dominated atmospheres, particular attention has been devoted to the long-term photochemical stability of CO₂, the potential for accumulation of significant abiotic oxygen and ozone abundances, and habitability (past or future). Giant planet studies have focused on clouds and hazes,

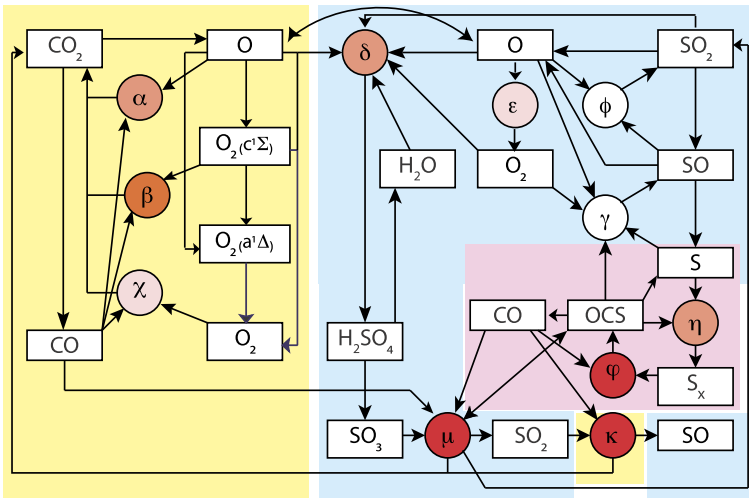


Fig. 10 Schematic diagram for the atmospheric chemistry on Venus (after Mills et al. 2007, 2019). Background shading identifies the carbon dioxide (yellow), sulfur oxidation (cyan), and polysulfur (magenta) cycles. Catalytic schemes are indicated by the Greek letters in circles. The catalytic schemes with white background had been confirmed by laboratory chemical kinetic studies as of 2007. Those in shades of red had not been fully confirmed. The degree of laboratory confirmation is indicated by the lightness of the shade of red: the darkest red had received no confirmation in laboratory studies; those in light red were largely but not completely confirmed

linking upper (observable) and deep (not observable) atmospheric compositions, hydrocarbon chemistry and methane recycling, coupled ammonia-phosphine photochemistry, and the role of vertical mixing. Icy planet studies have focused on clouds and hazes and hydrocarbon and nitrile chemistry. The remainder of this section discusses in more detail some of the important chemical cycles active on solar system planets.

Atmospheric chemistry on Venus appears to be dominated by three large-scale cycles: the carbon dioxide, sulfur oxidation, and polysulfur cycles (e.g., Mills et al. 2007; Marcq et al. 2018; Bierson and Zhang 2020) (see Fig. 10). The carbon dioxide cycle on Venus comprises: photolysis of CO_2 on the day side to produce CO and atomic oxygen, transport of a significant fraction of the CO and O to the night side, reaction of the atomic oxygen to form O_2 as confirmed by observations of intense $\text{O}_2(a^1\Delta - X^3\Sigma)$ airglow on the day and night sides (e.g., Connes et al. 1979; Crisp et al. 1996; Gérard et al. 2012), and conversion of CO and O_2 into CO_2 via catalytic processes to close the cycle. Chlorine catalyzed chemistry is believed to provide the most efficient pathways for producing CO_2 (Krasnopolsky and Parshev 1981; Yung and DeMore 1982; Pernice et al. 2004). The sulfur oxidation cycle comprises upward transport of OCS and SO_2 , oxidation of a significant fraction of the OCS and SO_2 to form H_2SO_4 , condensation of H_2SO_4 and H_2O to form a majority of the mass of the global cloud layers, downward transport of sulfuric acid as cloud droplets, evaporation of these cloud droplets, and thermal decomposition of H_2SO_4 to produce OCS and SO_2 (e.g., Krasnopolsky and Pollack 1994; Zhang et al. 2012; Bierson and Zhang 2020; Shao et al. 2020). The polysulfur cycle involves the upward transport of OCS and SO_2 , photodissociation to produce S , production of polysulfur (S_x) via catalytic processes or association reactions, downward transport of S_x , thermal decomposition of S_x , and reaction with CO or oxidation to produce OCS and SO_2 , respectively (e.g., Mills and Allen 2007; Zhang et al. 2012; Krasnopolsky 2013). Solid observational evidence supports the carbon dioxide

and sulfur oxidation cycles, although many details remain unproven. The more speculative polysulfur cycle is, nonetheless, plausible based on the limited observational and laboratory data that exist.

The carbon dioxide cycle on Mars is similar to that on Venus, but production of CO_2 is believed to be dominated by HO_x -catalyzed reactions (McElroy and Donahue 1972; Nair et al. 1994). NO_x in a CO_2 -dominated atmosphere like that on Mars doesn't catalyze production of CO_2 but it does increase the OH/HO_2 ratio which increases the effectiveness of the HO_x catalytic processes that produce CO_2 (Nair et al. 1994).

The stratospheric ozone cycle on Earth is one of the most important for life, and ozone is often considered a potential biosignature in extrasolar planet studies (e.g., Grenfell 2017; Meadows 2017). Atomic oxygen is produced from photolysis of O_2 , primarily in the O_2 Schumann-Runge bands (175–200 nm) and the Herzberg continuum (< 240 nm) (Minschwaner et al. 1993). The atomic oxygen then undergoes an association reaction with O_2 to form ozone. Ozone is converted back to O_2 via reaction with atomic oxygen (the Chapman process) and via catalytic processes involving ClO_x , HO_x , and NO_x radicals. The primary natural source for stratospheric NO_x is reaction of biogenic N_2O with $\text{O}(^1\text{D})$, with the $\text{O}(^1\text{D})$ coming from O_3 photolysis (e.g., Jacob 1999, Chap. 10). Biogenic CH_3Cl is the primary natural source for ClO_x (WMO 1999), and the primary source for stratospheric HO_x is reaction of $\text{O}(^1\text{D})$ with water (e.g., Jacob 1999, Chap. 10). The importance of these families for ozone destruction varies with altitude and geographic location (e.g., Müller et al. 1999; Ravishankara et al. 1999; Jacob 1999, Chap. 10). Aerosols also play a significant role in ozone loss through heterogeneous chemistry (Solomon et al. 1998; Ravishankara et al. 1999).

In the Earth's troposphere, HO_x and NO_x drive key catalytic cycles. Tropospheric OH, produced when $\text{O}(^1\text{D})$ from O_3 photolysis at 300–320 nm reacts with H_2O , is primarily lost via oxidation of CO and methane, but OH is also a key oxidant for many other species (e.g., Jacob 1999, Chap. 11). Unlike the stratosphere where NO_x destroys ozone, tropospheric NO_x with the much lower concentrations of O_3 and O in the troposphere acts to regenerate both OH and O_3 to maintain the oxidative capacity of the troposphere (e.g., Jacob 1999, Chap. 11). Earth's surface (biomass burning and soils) is the largest natural source for tropospheric NO_x , with natural long-range transport occurring predominantly via peroxyacetylnitrate (PAN) formed from photochemical oxidation of biogenic (and anthropogenic) hydrocarbons (e.g., Jacob 1999, Chap. 11). Lightning is the most important natural atmospheric source of tropospheric NO_x (e.g., Jacob 1999, Chap. 11). NO_x when combined with hydrocarbons (volatile organic compounds) and NUV radiation can produce smog.

A critical issue for understanding atmospheric chemical cycles is that their net effects are context-sensitive (DeMore and Yung 1982). As described above for NO_x , which produces O_3 in Earth's troposphere but destroys O_3 in Earth's stratosphere, the composition of an atmosphere strongly influences which chemical cycles are effective and their impact. ClO_x cycles, for example, destroy odd oxygen and produce O_2 in Earth's stratosphere but the different ClO_x pathways active in Venus' mesosphere and its different chemical state mean Venusian ClO_x destroys O_2 and produces CO_2 (DeMore and Yung 1982).

A methane cycle exists in the atmospheres of Jupiter, Saturn, Uranus, and Neptune. Methane from the deep atmosphere is transported past the tropopause cold trap and up into the upper stratosphere, where it is photolyzed, producing a suite of more complex hydrocarbons, Fig. 11. Some of the more refractory hydrocarbon photochemical products condense in the lower stratosphere, and both the gas-phase hydrocarbon products and haze particles flow downward into the deep troposphere, where high temperatures convert the hydrocarbons back to methane, completing the cycle (e.g., Strobel 1973; Moses et al. 2005).

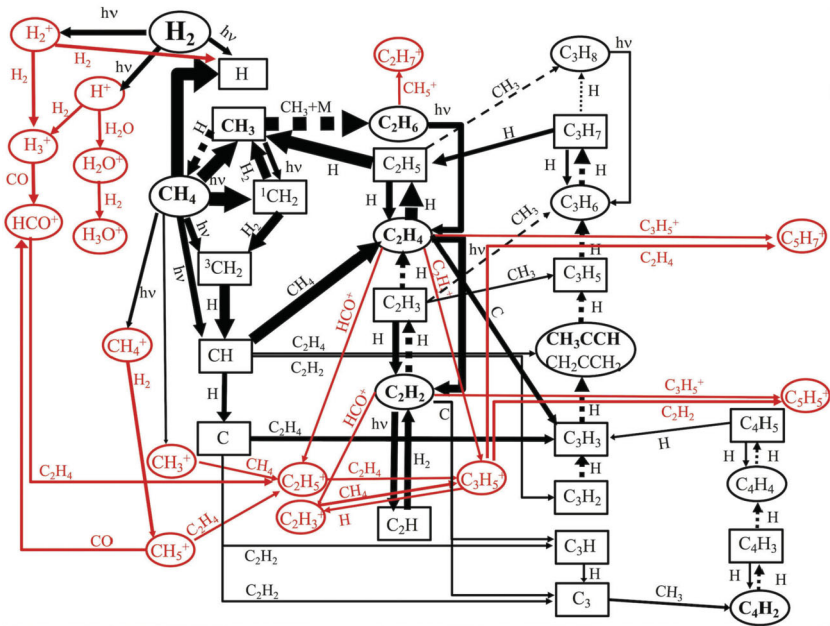


Fig. 11 Schematic diagram illustrating the major neutral (black) and ionic (red) reaction pathways for atmospheric chemistry on Neptune (from Dobrijevic et al. 2020). Radicals are shown in boxes; stable compounds and ions in circles. Dashed lines correspond to three body reactions. Species in bold have been detected

While methane photochemistry on Titan exhibits many similarities to that occurring on the giant planets (e.g., Yung et al. 1984), high-altitude ion chemistry is much more important in producing high-molecular-weight species and hazes, and Titan does not have a deep atmosphere to convert the hydrocarbons back to methane. The fate of the hydrocarbon products at the surface of Titan, as well as the ultimate source of the methane from the surface/interior, is unclear. Nitrogen species also participate in atmospheric photochemistry on Titan, due to the dominant background N_2 gas, and the resulting ion-neutral chemistry is very rich and complex (e.g., Vuitton et al. 2006; Lavvas et al. 2008). Ion chemistry is vital to the formation of high-altitude hazes on Titan, and large negative ions or particles can contribute to haze growth (e.g., Coates et al. 2007).

Similar cycles involving ammonia and phosphine exist in the upper tropospheres of the giant planets. Phosphine photolysis (on Uranus and Neptune) and coupled ammonia-phosphine photochemistry (on Jupiter and Saturn) lead to the photochemical production of P_2H_4 and N_2H_4 , primarily (e.g., Kaye and Strobel 1983, 1984; Teanby et al. 2019). These products condense *in situ* and are transported downward, where the particles evaporate and are eventually converted back to PH_3 and NH_3 .

Phosphine itself, along with CO , AsH_3 , and GeH_4 , are disequilibrium species in the upper tropospheres of the giant planets. The presence of these molecules in greater-than-expected quantities at these altitudes suggests they are “quenched” at thermochemical equilibrium abundances in the deeper troposphere and transported upward from this quench level at a rate faster than they can be chemically converted to the expected equilibrium forms at higher altitudes (e.g., Prinn and Barshay 1977; Fegley and Prinn 1985). This disequilibrium transport-induced quenching process is also important on brown dwarfs, directly imaged giant exoplanets, and hot-Jupiter exoplanets (e.g., Fegley and Lodders 1996; Saumon et al. 2006;

Barman et al. 2011; Venot et al. 2012; Moses et al. 2011, 2016) and might indeed occur on any planet with a thick, deep atmosphere that is hot at depth.

Chemistry in the atmospheres of extrasolar planets has been explored by many groups (see the review of Madhusudhan et al. 2016). The dominant chemical cycles will, of course, depend on the bulk atmospheric composition and temperatures, as well as on planetary and orbital parameters and stellar type. Smaller planets have the potential for having hugely diverse atmospheric composition and properties, due to differences in planetary formation and evolution and atmospheric evolution, as well as current system architecture. However, H₂-dominated giant exoplanets will also have highly variable minor constituent abundances, with atmospheric temperatures strongly controlling what “parent” molecules can be present, what clouds will form at what pressure regions, and what elements will be tied up in condensates at depth, thus being unavailable for further chemistry at observable altitudes. On all types of planets, transient events that loft condensable elements to observable altitudes may offer both puzzling inconsistencies and windows into deeper altitudes.

While studies of atmospheric chemistry on solar-system planets can help inform us of the chemical processes that might be occurring on extrasolar planets, the sheer diversity of exoplanet properties makes it clear that many exoplanets will have exotic atmospheres with no true solar-system analogs. The same basic chemical and physical principles will still control exoplanet atmospheric behavior, however, and the atmospheric chemical cycles identified for our solar system provide a solid foundation for identifying and understanding atmospheric chemistry on exoplanets. The lessons we have garnered from decades of planetary observations and modeling can be adapted to the study of exoplanet atmospheres.

5 Synthesis of Lessons Learned

Atmospheric chemistry on all solar system planets is rich, complex, and varied. Photochemistry produces remotely observable trace disequilibrium products even on planets that are far from a star. Restoration towards thermochemical equilibrium typically proceeds via catalytic cycles involving radicals that usually cannot be observed remotely. One of the major conceptual advances from terrestrial and solar system studies has been organizing reactions into coherent chemical cycles that encapsulate the creation of disequilibrium species and the restoration toward equilibrium. Laboratory studies that confirm (or refute) hypothesized pathways have been and remain a critical part of this process. The strength and net effect of each chemical cycle depends on the local atmospheric context, but reactions within each atmospheric chemical cycle can be categorized into: (1) reactions that produce a specific bond (e.g., the O-O bond in O₂), (2) reactions that inhibit production of that bond, (3) reactions that break that bond, and (4) reactions that suppress catalytic processes (DeMore and Yung 1982). Organizing atmospheric chemistry processes into cycles is one way that knowledge gained from solar system studies has been applied in exoplanet research. A major focus of exoplanet studies has been and remains understanding how known chemical cycles operate under the broader range of exoplanet conditions to predict their implications for exoplanet atmospheric spectra. Another major focus needs to be identifying chemical cycles that may be unique to exoplanets, particularly those with conditions that do not have solar system analogs.

Atmospheres and atmospheric chemistry co-evolve as the planet-star system evolves (e.g., Catling and Kasting 2017). This temporal history will determine what is observable in any epoch and the potential for habitability at the time a planet is observed. A planet’s integrated history then determines the potential for life’s persistence. Factors affecting a

planetary atmosphere's chemistry and chemical evolution are both internal and external to the planet, including: outgassing, weathering, burial, and influx of interplanetary dust and cometary material, stellar particles, and molecules and dust from a planet's satellites. Ions produced by incident stellar radiation and particle flux can affect a planet's remotely observable neutral chemistry. Axial tilt and eccentricity cause time-dependent seasonal radiative forcing and temperatures, and on longer time-scales evolution of axial tilt and eccentricity can dramatically change planetary conditions and habitability. Transient events may mean the observed characteristics of an atmosphere differ from the long-term steady-state. The whole-planet models that are needed and under development to understand exoplanet evolution, habitability through time, and the potential persistence of life are building on similar models developed for planets in our solar system (e.g., Bullock and Grinspoon 2001), and data from solar system research are used to benchmark the exoplanet models.

Nonlinear interactions among radiation, dynamics, and chemistry can significantly influence an atmosphere's composition and chemistry. Trace species produced by photochemistry can affect thermal structure and heating/cooling rates, heating/cooling rates in turn affect circulation patterns, and circulation patterns affect species distributions. Vertical mixing, which solar system research has shown is very difficult to predict from first principles, has a strong influence on species distributions and remotely observable spectral features, and for planets in which some region of the atmosphere has temperatures that exceed ~ 1000 K (e.g., at great depths on giant planets or on the day side of a close-in tidally locked exoplanet), transport-induced quenching can be important. Consequently, the temperature profile, dynamics, and quantum mechanics all influence what species are observable. Such processes are starting to be included in exoplanet models based on their demonstrated importance to solar system planetary atmospheric chemistry, and recent exoplanet models suggest these processes are important for controlling the composition of close-in transiting exoplanets with thick, deep atmospheres (e.g., Moses et al. 2011; Venot et al. 2012; Tsai et al. 2017; Mendonça et al. 2018; Drummond et al. 2018b).

The complexity of atmospheric chemistry and its nonlinear interactions with the climate system noted above mean there is a large range of parameter space to be explored and characterized for exoplanets. This makes a hierarchy of models very useful for predicting or explaining observed behavior. Global-average 1D models are needed for complex chemistry and work well for exploratory simulations, but 3D models that couple chemistry, dynamics, and radiation are needed to predict the full behavior of inherently complex atmospheres. Intermediate complexity models, like 2D and 3D chemical transport models, which have been very useful in terrestrial studies, also have an important role to play but are only beginning to be utilized for exoplanet research.

The remotely observable composition of a planet's atmosphere is not necessarily the same as the composition of its bulk atmosphere. Condensation plays a critical role by sequestering species and elements at deeper levels, limiting the depth of penetration of photons that can initiate chemical reactions, and producing clouds that limit the depth to which remote sensing observations can probe. Even above constituent condensation levels, hazes can form with variable optical depths. Finally, within the remotely observable portion of a planet's atmosphere, the most prominent spectral features are often due to photochemical products with small mixing ratios that may only provide indirect hints about the deeper, bulk composition of the atmosphere. These characteristics and processes are well-known from solar system atmospheres and are among the foremost considerations when translating exoplanet atmospheric simulations into potential observables.

The fundamental tasks for exoplanetary atmospheric chemistry in the next decade are (1) identifying combinations of potentially observable planet-star features (not just the presence and/or absence of species) that constrain bulk atmospheric abundances and provide

high confidence that known chemical cycles are occurring, (2) assessing the viability of alternative pathways and chemical cycles that could plausibly occur on exoplanets but are not important on solar system planets, (3) identifying the most important unknown or poorly constrained laboratory/theoretical data required for quantifying the effects of known and speculative chemical cycles, and (4) developing schemes for efficiently inferring the ranges of parameter space that correspond to forthcoming observations. It is very likely that observations obtained in the next decade will provide information on potentially habitable exoplanets. The challenge will be to interpret exoplanet observations and infer from a significantly underconstrained system what conditions are like on these exoplanets.

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