SOLUTIONS TO THE FAINT YOUNG SUN PARADOX SIMULATED BY A GENERAL
CIRCULATION MODEL

by

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Solutions to the faint young Sun paradox simulated by a general circulation model

Thesis directed by Professor Owen Brian Toon

The faint young Sun paradox has dominated our thinking regarding early climate. Geological evidence abounds for warm, possibly hot, seawater temperatures and the proliferation of early life during the Archean period of Earth’s history (3.8–2.5 Ga). However the standard solar model indicates that the Sun was only 75 to 82 percent as bright as today, implying an apparent contradiction between warm surface temperatures and weak solar irradiance. Geological evidence also places constraints on the amount of atmospheric carbon dioxide present early in Earth’s history. Over the past four decades there has been much debate amongst geological, planetary, and climate science communities regarding how to properly resolve the issue of the faint young Sun. Up until very recently, 1-dimensional radiative convective models were the standard tool for deep paleoclimate modeling studies. These studies have notably lacked the ability to treat clouds, surface ice, and meridional energy transport. However, advancements in computing technology now allow us to tackle the faint young Sun paradox using a three-dimensional climate model. Here we use a modified version of the Community Atmosphere Model version 3 from the National Center for Atmospheric Research to study early climate. We find that resolving the faint young Sun paradox becomes less problematic when viewing a full representation of the climate system. Modest amounts of carbon dioxide and methane can provide adequate warming for the
Archean within given constraints. Cooler climates with large ice caps but temperate tropical regions can be supported with even less carbon dioxide. The incorporation of systematic climate system differences expected during the Archean, such as fewer cloud condensation nuclei, reduced land albedos, and increased atmospheric nitrogen, can provide additional non-greenhouse means of warming the early Earth. A warm Archean no longer appears at odds with a faint young Sun. Here, we will also discuss the consequences of the oft-suggested Titan-like photochemical haze that may have enshrouded the early Earth if methane was a significant constituent of the atmosphere. Finally, we briefly consider the inverse problem. What fate may be in store for the Earth as the Sun continues to brighten far past its present level?
## CONTENTS

<table>
<thead>
<tr>
<th>CHAPTER</th>
<th>PAGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. An introduction to the faint young Sun paradox</td>
<td>1</td>
</tr>
<tr>
<td>2. Model description</td>
<td>11</td>
</tr>
<tr>
<td>2.1. Overview</td>
<td>11</td>
</tr>
<tr>
<td>2.2. Treatment of oceans and sea ice</td>
<td>12</td>
</tr>
<tr>
<td>2.3. Treatment of cloud physics</td>
<td>17</td>
</tr>
<tr>
<td>2.4. Treatment of radiative transfer</td>
<td>19</td>
</tr>
<tr>
<td>3. The effects of reduced solar constant and increased greenhouse gases</td>
<td>28</td>
</tr>
<tr>
<td>3.1. Overview</td>
<td>28</td>
</tr>
<tr>
<td>3.2. Standard atmospheres</td>
<td>30</td>
</tr>
<tr>
<td>3.3. Model sensitivity</td>
<td>41</td>
</tr>
<tr>
<td>3.4. Hospitable Archean climates</td>
<td>49</td>
</tr>
<tr>
<td>3.5. Discussion</td>
<td>55</td>
</tr>
<tr>
<td>3.6. Summary</td>
<td>59</td>
</tr>
<tr>
<td>4. Controls on the Archean climate</td>
<td>61</td>
</tr>
<tr>
<td>4.1. Overview</td>
<td>61</td>
</tr>
<tr>
<td>4.2. Increased rotation rate</td>
<td>66</td>
</tr>
<tr>
<td>4.3. Darker surface albedo</td>
<td>71</td>
</tr>
<tr>
<td>4.4. Changes to clouds</td>
<td>76</td>
</tr>
<tr>
<td>4.5. Increased atmospheric nitrogen</td>
<td>87</td>
</tr>
<tr>
<td>4.6. Optimal warming solutions</td>
<td>91</td>
</tr>
<tr>
<td>4.7. Summary</td>
<td>97</td>
</tr>
<tr>
<td>5. A fractal aggregate model of early Earth photochemical hazes</td>
<td>99</td>
</tr>
<tr>
<td>5.1. Overview</td>
<td>99</td>
</tr>
<tr>
<td>5.2. Model</td>
<td>101</td>
</tr>
<tr>
<td>5.3. Fractal microphysics</td>
<td>102</td>
</tr>
<tr>
<td>5.4. Fractal optics</td>
<td>108</td>
</tr>
<tr>
<td>5.5. Results from uncoupled simulations</td>
<td>117</td>
</tr>
<tr>
<td>5.6. Results from radiatively coupled simulations</td>
<td>131</td>
</tr>
<tr>
<td>5.7. Discussion</td>
<td>135</td>
</tr>
<tr>
<td>5.8. Summary</td>
<td>137</td>
</tr>
<tr>
<td>6. Earth's future under the ever brightening Sun</td>
<td>139</td>
</tr>
<tr>
<td>6.1. Overview</td>
<td>139</td>
</tr>
<tr>
<td>6.2. Implications for Earth</td>
<td>143</td>
</tr>
<tr>
<td>6.3. Implications for other planets</td>
<td>151</td>
</tr>
<tr>
<td>6.4. Summary</td>
<td>152</td>
</tr>
<tr>
<td>7. Concluding remarks</td>
<td>153</td>
</tr>
<tr>
<td>8. References</td>
<td>160</td>
</tr>
<tr>
<td>NUMBER</td>
<td>TABLE DESCRIPTION</td>
</tr>
<tr>
<td>--------</td>
<td>------------------</td>
</tr>
<tr>
<td>2.1.</td>
<td>Radiative transfer spectral intervals and absorbing species</td>
</tr>
<tr>
<td>3.1.</td>
<td>Standard atmospheres</td>
</tr>
<tr>
<td>4.1.</td>
<td>Rotation rate sensitivity tests</td>
</tr>
<tr>
<td>4.2.</td>
<td>Surface albedo sensitivity tests</td>
</tr>
<tr>
<td>4.3.</td>
<td>Cloud droplet number concentration sensitivity tests</td>
</tr>
<tr>
<td>4.4.</td>
<td>Nitrogen sensitivity tests</td>
</tr>
<tr>
<td>5.1.</td>
<td>Uncoupled haze simulations</td>
</tr>
</tbody>
</table>
# LIST OF FIGURES

<table>
<thead>
<tr>
<th>NUMBER</th>
<th>PAGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1. The evolution of the solar constant over time</td>
<td>2</td>
</tr>
<tr>
<td>1.2. Paleo-seawater temperature estimates versus time</td>
<td>5</td>
</tr>
<tr>
<td>1.3. Constraints on paleo-atmospheric CO₂</td>
<td>7</td>
</tr>
<tr>
<td>2.1. The evolution of sea ice fraction and atmosphere-to-ocean heat flux</td>
<td>15</td>
</tr>
<tr>
<td>2.2. Radiative transfer flux validations for increasing CO₂ and CH₄</td>
<td>23</td>
</tr>
<tr>
<td>2.3. Vertical profiles of temperature and greenhouse gases used for radiative transfer validation</td>
<td>26</td>
</tr>
<tr>
<td>2.4. Radiative transfer flux and heating rate validations against Archean profiles</td>
<td>27</td>
</tr>
<tr>
<td>3.1. Difference in zonal mean surface forcings and fluxes between Archean and present day atmospheres</td>
<td>34</td>
</tr>
<tr>
<td>3.2. Vertical profiles of temperature, specific humidity, and relative humidity for Archean and modern atmospheres</td>
<td>35</td>
</tr>
<tr>
<td>3.3. Moist convective mass and water fluxes for Archean and modern atmospheres</td>
<td>37</td>
</tr>
<tr>
<td>3.4. Zonal mean cloud fractions and forcings for Archean and modern atmospheres</td>
<td>38</td>
</tr>
<tr>
<td>3.5. Climate sensitivity under changing CO₂ for Archean and modern atmospheres</td>
<td>43</td>
</tr>
<tr>
<td>3.6. Time and temperature evolution of surface albedo, sea ice and snow cover</td>
<td>47</td>
</tr>
<tr>
<td>3.7. Late Archean global mean surface temperature, mean tropical temperature and sea ice margin versus partial pressure of CO₂</td>
<td>50</td>
</tr>
<tr>
<td>3.8. Late Archean global mean albedo, cloud forcing, and greenhouse components versus partial pressure of CO₂</td>
<td>53</td>
</tr>
</tbody>
</table>
3.9. Late Archean global mean and seasonal maximum
seawater temperatures versus partial pressure of CO₂ ............................... 57
4.1. Zonal mean streamfunction for changing rotation rates .......................... 68
4.2. Zonal mean surface temperature for changing rotation rates .................. 69
4.3. Zonal mean surface temperature for changing land albedos ................. 74
4.4. Zonal mean cloud condensate for changing cloud droplet
number concentrations ................................................................................. 81
4.5. Zonal mean cloud fractions and forcings for changing
cloud droplet number concentrations .......................................................... 82
4.6. Zonal mean cloud fraction contour for Archean
and present day atmospheres ..................................................................... 86
4.7. Global mean radiative forcing versus partial pressure of N₂ ................... 89
4.8. CO₂ needed to maintain global mean surface temperatures of 288 K ........ 94
5.1. Early Earth haze production rate global mean vertical profile ............... 103
5.2. Fractal aggregate restructuring parameterization .................................... 107
5.3. The ratio of fractal aggregate optical efficiencies to
mie optical efficiencies versus wavelength for equal
mass particles using Khare’s optical constants ........................................... 111
5.4. Fractal aggregate asymmetry parameters versus wavelength ............... 112
5.5. Fractal aggregate single scattering albedos versus wavelength .............. 113
5.6. Fractal aggregate absorption enhancement sensitivity
for scaled optical constants ........................................................................... 115
5.7. Fractal aggregate absorption enhancement sensitivity
under wavelength independent optical constants ......................................... 116
5.8. Zonal mean haze number density .......................................................... 120
5.9. Zonal mean haze mass concentration ..................................................... 121
5.10. Global mean vertical profile of spherical
and fractal aggregate effective radii ............................................................ 124
5.11. Global mean vertical profile of the mean aggregate monomer number ................................................................. 125

5.12. Sensitivity of fractal effective radii to changing assumed monomer radii ............................................................... 127

5.13. Sensitivity of aggregate monomer number to changing assumed monomer radii ......................................................... 128

5.14. Effective optical depth for spherical and fractal haze layers versus wavelength ...................................................... 129

5.15 Sensitivity of fractal haze effective optical depth to changing assumed monomer radii ..................................................... 130

5.16. Global mean surface temperature and haze effective optical depth as a function of haze production rate ........................ 133

5.17. Zonal mean haze effective optical depth .................................................................................................................. 134

6.1. Surface temperatures and climate sensitivity versus increasing solar constant ............................................................... 145

6.2. Global mean vertical profiles of temperature, water vapor mixing ratio and relative humidity for increasingly hot climates ............................................................................................................................................. 146

6.3. Water vapor column, cloud water column, clear-sky greenhouse forcing and cloud forcing versus increasing solar constant ............................................................................................................................................. 148

6.4. Zonal mean surface temperature, cloud forcing, absorbed solar energy, and outgoing longwave energy for increasingly hot climates .................................................................................................................... 149
Chapter 1

An introduction to the faint young Sun paradox

As main sequence stars age, they gradually increase in luminosity. A star’s energy is produced via the fusion of hydrogen into helium within its core. Since the density of helium is greater than that of hydrogen, over time the core grows denser, contracts, and heats, resulting in increased energy production and subsequently larger stellar luminosities. The gradual brightening of main sequence stars is a rigorous prediction of the standard solar model. If we wind the clock backwards through geologic time, our Sun must have been much less bright in the distant past. Conversely in the distant future Earth’s solar constant will continue to increase. Figure 1.1 illustrates the change in the solar constant over the course of Earth’s history. During the Archean period of Earth’s history, dating 3.8 to 2.5 billion years ago (Ga), the Sun as up to 25 percent less luminous than it is today (Gough, 1981).

However despite weak solar irradiance, geologic evidence indicates that the early Earth had liquid water in the near surface environment along with early life. The so-called “faint young Sun paradox” was first recognized by Sagan and Mullen (1972), noting that the early Earth must have had a significantly larger greenhouse effect or a much lower planetary albedo compared with the present day in order to prevent widespread glaciations.
Figure 1.1. The evolution of the solar constant over time. At the beginning of the Archean, the Sun was 75% as bright at today while at the end of the Archean the Sun was 82% as bright. The Sun will continue to increase in luminosity as time marches into the future.
The earliest evidence for liquid water at Earth’s surface can be traced back to 4.4 Ga via analysis of ancient zircons (Wilde et al., 2001; Mojzsis et al., 2001). However the Hadean Earth (4.6 to 3.8 Ga) was characterized by frequent large meteor impacts and tumultuous geological activity. Thus the presence of surface water deduced from zircons is not a guarantee of a continuously stable climate. Due to intense crustal reworking, little record the Hadean remains left today for study. The Archean period of Earth’s history begins at the termination of the Late Heavy Bombardment; a period of intense meteor impacts spanning 4.0 to 3.8 Ga (Tera et al., 1974). The Archean rock record features widespread evidence of water-lain sediments, indicating the presence of standing bodies of water at the surface (Lowe, 1980; Knauth and Lowe, 2003; Rosing and Frei, 2004). Furthermore, aside from evidence for widespread glaciation between 2.45 and 2.22 Ga (Evans et al., 1997) and a possible regional glaciation at 2.9 Ga (Young et al., 1998), direct evidence for glaciation is conspicuously absent from the Archean geologic record. No evidence is found of dropstones, glacial striations, nor erosional and depositional features characteristic of glacial processes.

There is also continuous evidence for ancient life beginning as early as 3.8 billion years ago, further supporting the conclusion that the Archean was generally hospitable having at least some temperate regions capable of supporting life through the majority of its history. Isotopically light carbon inclusions found in the Isua supracrustal belt, West Greenland dating to 3.8 Ga are best explained by biological processes, perhaps derived from ancient plankton (Mojzis et al., 1996; Rosing, 1999). The 3.43 Ga Strelly Pool Chert, Pilbara, Australia contains a well
preserved stromatolite reef (Allwood et al., 2006). Carbonaceous matter found in the 3.416 Ga Buck Reef Chert, South Africa is conjectured to be evidence of photosynthetic microbial mats (Tice and Lowe, 2004). While the Archean rock record is significantly degraded due to metamorphism, fossil evidence from the Archean period remains remarkably common (Schopf, 2006). All life on Earth is based on the existence of liquid water, thus the widespread presence of life throughout the Archean is strong evidence that the Earth has maintained at least some areas of liquid water.

The isotopic composition of ancient marine cherts serve as a proxy for seawater temperatures during the Archean. Figure 1.2 illustrates several estimates for paleo-ocean temperatures. Earlier works which focused independently on oxygen and silicon isotopic excursions predict exceedingly hot seawater temperatures; as high as 360 K during the early Archean (Knauth and Lowe, 1978; Knauth and Lowe, 2003; Robert and Chaussidon, 2006). This evidence along with the absence of obvious glacial deposits led many researchers to conclude that the early Earth must have been hot and devoid of ice (Kasting, 2010). However, the high seawater temperature interpretation of the isotopic composition of Archean cherts is not universally accepted (Kasting et al., 2006; Jaffres et al., 2007; Shields and Kasting, 2007). More recent attempts to quantify the Archean seawater temperatures have yielded more moderate temperatures, similar to present day ocean temperatures. Hren et al. (2009) use combined analysis of oxygen and hydrogen isotopes and found maximum seawater temperatures of 313 K at 3.42 Ga. Blake et al. (2010) analyze the oxygen isotopic composition of phosphates and
Figure 1.2. Paleo-seawater temperature estimates versus time. Paleo-seawater temperatures have been estimated based on geochemical analysis and oxygen and silicon isotopic ratios in Ancient marine cherts. The Archean is generally believed to have been at least as warm as today despite the faint young Sun.
deduce that seawater temperatures ranged between 299 and 308 K from samples dating from 3.2 and 3.5 Ga. Despite the disagreements amongst temperature estimates, all seawater temperature estimates point towards a warm Archean despite the faint young Sun.

However, the Archean geologic record is both spatially and temporally sparse and the paleolatitudes of samples are unknown (Feulner, 2012). Recent work suggests that by 2.8 Ga nearly 70% of the present day volume of continental crust had been established, however high crustal reworking rates imply that very little unaltered material from this time period remains left today for study (Dhuime et al., 2012). Thus, combined arguments from seawater temperature proxies, fossil evidence for life, and the absence of glacial deposits in Archean samples may not be ironclad evidence for a hot and completely ice-free planet. Meridional gradients of temperature may imply a temperate tropical zone while the poles remained cool. A solution to a weak form of the faint young Sun paradox may only require that liquid water was present on some part of the planet surface.

The most straight forward resolution to the faint young Sun paradox is to invoke an early Earth with elevated greenhouse gases. A CO₂-rich atmosphere could easily keep the early Earth warm despite low solar luminosity (Kasting, 1987), but analysis of paleosols (Rye et al., 1995; Sheldon, 2006; Driese et al., 2011) and banded iron formations (Rosing et al., 2010) impose constraints on its paleoatmospheric concentration (Figure 1.3). Radiative convective models (RCM) infer that 0.03 bar of CO₂ is needed to compensate for a 20% reduction in the solar
Figure 1.3. **Constraints on paleo-atmospheric CO$_2$.** CO$_2$ constraints derived from paleosols place the partial pressure of CO$_2$ between 0.01 and 0.02 bar for the late Archean. Constraints derived from banded-iron formations limit CO$_2$ to 0.001 bar.
constant, marginally preventing runaway glaciation with global mean surface
temperatures hovering at 273 K (Haqq-Misra \textit{et al.}, 2008; von Paris \textit{et al.}, 2008). However, a recent estimate based on the geochemical mass-balance of tonalite
weathering profiles place paleoatmospheric CO$_2$ at the end of the Archean at no
more than 50 times the present atmospheric level (PAL), $\sim$0.018 bar as an upper
limit at 2.69 Ga (Driese \textit{et al.}, 2011). Estimates deduced from the presence of
magnetite within banded iron formations suggest much tighter constraints on CO$_2$,
limiting its paleoatmospheric concentration during the Archean to a mere $\sim$3 PAL
(0.001 bar) severely worsening the apparent paradox (Rosing \textit{et al.}, 2010).
However, the Rosing \textit{et al.} (2010) interpretation is controversial (Reinhard and
Planavsky, 2011; Dauphas and Kasting, 2011).

Additional greenhouse warming may be provided by methane.

Photochemical-ecosystem models infer that up to 0.035 bar of CH$_4$ may have been
maintained in the anoxic Archean atmosphere (Kharecha \textit{et al.}, 2005). However, a
CH$_4$ greenhouse is self-limiting. Titan-like photochemical hazes can form with the
ratio of CH$_4$ to CO$_2$ as low as 0.1 (DeWitt \textit{et al.}, 2009). Thus, too much methane may
result in the formation of optically thick organic hazes that could cool the early
Earth, negating any gained greenhouse warming (Haqq-Misra \textit{et al.}, 2008; Domagal-
Goldman \textit{et al.}, 2008). Nonetheless, using a RCM with interactive haze chemistry,
Haqq-Misra \textit{et al.} (2008) show that suitable combinations of CO$_2$ and CH$_4$ can yield
temperate solutions for the late Archean climate if one adopts the more lenient
constraints on CO$_2$ posed by Driese \textit{et al.} (2011) and Sheldon (2006) as opposed to
sulfide and higher order hydrocarbons have high greenhouse potentials and could
aid in hotter climate solutions, but such trace gas solutions are not unanimously
corroborated by photochemical and climate calculations (Sagan and Mullen, 1972;
Ueno et al., 2009; Haqq-Misra et al., 2008).

For the past two decades paleoclimate modeling studies with relevance to
the faint young Sun paradox have primarily used radiative-convective models
(Kasting et al., 1984; Kasting, 1987; Haqq-Misra et al., 2008; von Paris et al., 2008;
Domagal-Goldman et al., 2008; Goldblatt et al., 2009b; Rosing et al., 2010;
Wordsworth and Pierrehumbert, 2013). Until very recently, only limited research
had been conducted to date on the Archean climate using multi-dimensional models
(Henderson-Sellers and Henderson-Sellers, 1988; Jenkins et al., 1993; Jenkins, 1993;
Jenkins, 2001; Stone and Yao, 2004; Charnay et al., 2013; Kunze et al., 2013; Wolf
and Toon, 2013; Wolf and Toon, 2014b). RCMs are attractive research tools because
they are computationally fast and can incorporate self-consistent atmospheric
chemistry appropriate for reduced atmospheres, a capability not yet included in
three-dimensional models. However RCMs have limitations when it comes to
modeling climate. RCMs represent the entirety of planetary climate with a single
globally averaged vertical column and thus they ignore latitudinally dependent
processes. RCMs often omit clouds and fix the surface albedo to a singular and
constant value. In RCM paleoclimate simulations, the standard procedure is to tune
the surface albedo to duplicate modern climate under clear-sky conditions. The
surface albedo is then held fixed in all subsequent simulations, thus the ice-albedo
feedback and cloud feedbacks are ignored. The absence of realistic longwave cloud
absorption in RCMs also leads to an overestimation of the radiative forcing from CO₂ by up to 25% when pCO₂ = 0.1 bar (Goldblatt and Zahnle, 2011). The difficulty achieving hot Archean climates with RCMs and the stringent constraints on CO₂ posed by Rosing et al. (2010) have led to alternative theories for warming the Archean that invoke changes to the planetary albedo and meridional distribution of energy (Rossow et al., 1982; Henderson-Sellers and Henderson-Sellers, 1988; Jenkins et al., 1993; Jenkins, 1993; Jenkins, 2001; Rosing et al., 2010; Rondanelli and Lindzen, 2010).

In this thesis, the faint young Sun paradox is reexamined using an atmospheric general circulation model coupled to a mixed layer ocean model. While factors of solar physics and geochemistry frame the problem, ultimately the faint young Sun paradox is a question of climate. This work represents the most comprehensive climate modeling study of the faint young Sun paradox to date. Chapter 2 discusses the technical aspects of the model. Chapter 3 explores greenhouse solutions, varying the solar constant, CO₂ and CH₄ only. Chapter 4 explores systematic controls to the Archean climate system that may help warm the early Earth above and beyond that which can be gained from greenhouse gases alone. Chapter 5 explores the microphysical and radiative properties of Titan-like photochemical hazes along with their consequences on climate. Chapter 6 briefly discusses the natural inverse of the faint young Sun paradox; what will happen to climate as the Sun continues to brighten above its current level. The work is summarized in the larger context of the faint young Sun paradox in chapter 7.
Chapter 2

Model description

2.1. Overview

In this study we use a general circulation model (GCM) to simulate Archean climate. We use the Community Atmospheric Model version 3.0 (CAM3) from the National Center for Atmospheric Research (Collins et al., 2004). CAM3 is a widely used GCM and has been extensively validated against modern climate. Regional and seasonal biases exist but global and annual mean climatological statistics agree well with modern climate (Collins et al., 2006; Hurrel et al., 2006). Refer to chapter 3 for statistics for baseline climate simulations of present day Earth conditions. CAM3 consists of component atmosphere, ocean, land, and sea ice models. The model uses a finite volume dynamical core (Lin and Rood, 1996). Simulations were conducted with 4°x 5° horizontal resolution with 66 vertical levels extending up to $5 \times 10^{-6}$ mb. This pressure equates to a model top of ~130 km in simulations of present day and ~90 km for Archean simulations. The difference in model top height stems from the lack of ozone in Archean simulations which results in a cold upper atmosphere with compressed scale heights. Throughout this work, all simulations are initiated from present day surface temperatures. Equilibrium climate is typically reached within ~50 model years, however to ensure that our results are robust against slow adjustment, simulations are run an additional 50–100 years after equilibration. For
all simulations presented, equilibrium conditions with no noticeable systematic drift are reached for the quantities of global mean surface temperature, sea ice fraction, sea ice thickness, surface albedo, top-of-atmosphere albedo and snow areal coverage. For simulations that include hazes, CAM3 has been linked to the Community Aerosol and Radiation Model for Atmospheres (CARMA) (Bardeen et al. 2008). The addition of CARMA allows for sectional microphysical modeling. We reserve an in depth discussion of CARMA haze modeling for Chapter 5. In the following subsections we discuss the CAM3 model components that are most pertinent in evaluating the faint young Sun paradox; oceans, sea ice, clouds, and radiative transfer.

### 2.2. Treatment of oceans and ice

CAM3 utilizes a mixed layer ocean model coupled to a thermodynamic sea ice model (Collins et al., 2004; Collins et al., 2006). We have chosen to use a mixed layer ocean model primarily due to the prohibitive computational expense of running a dynamic ocean model. Though the added computational overhead of coupling a dynamic ocean model to an atmospheric GCM is quite reasonable, spin-up times can be an order of magnitude longer than for GCMs run with a mixed layer ocean model (Hansen et al., 2005). In this study our goal is to probe a wide range of atmospheric conditions for the Archean, a task that would not be practical with a dynamic ocean model given our finite computational resources and time constraints. Nonetheless the utilization of a mixed layer ocean and thermodynamic sea ice model represents
a significant improvement upon previous climate studies of the faint young Sun paradox.

The mixed layer ocean model has seasonally and geographically varying depths between 10 and 200 m. A single temperature, characteristic of the entire mixed layer, is calculated via bulk energy balance equations between ocean, ice and atmosphere at each ocean grid cell. The primary equation controlling the mixed layer temperature is given by

\[
\rho_0 C_o h_0 \frac{\partial T_0}{\partial t} = (1 - A) F + Q_{\text{flux}} + A F_{\text{at}} + (1 - A) F_{\text{frz}} \tag{eq. 2.1}
\]

where \( \rho_0 \) is the density of ocean water, \( C_o \) is the heat capacity of ocean water, \( h_0 \) is the annual mean ocean mixed layer depth, \( A \) is the fraction of the ocean covered by sea ice, \( F \) is the net atmosphere to ocean heat flux, \( Q_{\text{flux}} \) is an internal ocean heat flux convergence term, \( F_{\text{at}} \) is the net ocean to sea ice heat flux, and \( F_{\text{frz}} \) is the heat gained when sea ice grows over open waters (Collins et al. 2004). Sea ice begins to form on the surface when the mixed layer temperature falls below the freezing point, taken to be \(-1.8 \, ^\circ\text{C}\) for seawater. While the mixed layer ocean model is inherently motionless, deep ocean and horizontal transport is parameterized through the specification of a seasonally varying ocean internal heat flux convergence term (\( Q_{\text{flux}} \)). The initial set of \( Q_{\text{flux}} \) values are tuned to reproduce present day ocean-to-atmosphere heat exchange and implicitly incorporate the effects of ocean circulations. As the planet cools, oceans tend to transport heat to the sea ice margin through convective mixing at the ice boundary and through wind driven circulations (Poulsen et al., 2001). Qualitatively similar behavior is observed in our mixed layer
ocean model which utilizes flux adjustments made to the initial set of $Q_{\text{flux}}$ values as a function of changing sea ice cover (Figure 2.1). The primary role of ocean heat transport is to remove heat from the tropics and deposit it at mid and high latitudes. Thus the mean meridional structure of $Q_{\text{flux}}$ is negative (divergence) in the tropics and positive (convergence) in the extra-tropics. Where $Q_{\text{flux}} > 0$, heat is transferred from ocean to atmosphere. The locations with relatively stronger $Q_{\text{flux}}$ convergence correlate to the leading edge of the sea ice as can be observed in figure 2.1. Thus variations to the meridional structure of $Q_{\text{flux}}$, which in part is dictated by adjustments, can affect the location of the sea ice line which in turn affects the global mean surface temperature. However, the stabilization of large polar cap climate states with sea ice extending into mid and low latitudes appears to be a robust feature of climate models, found in energy-balance models and GCMs utilizing both mixed layer and dynamical ocean-sea ice models (Ferreira et al., 2011; Abbot et al., 2011; Yang et al., 2012a; Yang et al., 2012b; Voigt and Abbot, 2012). Configured with a mixed layer ocean model, CAM3 is suited to model climates centered about present day surface temperatures that maintain large areas of open ocean. However, the simulation of a transition into a “snowball” climate state requires the use of dynamical ocean and sea ice models (Voigt and Abbot, 2012). See the subsection on “Model Sensitivity” in Chapter 3 for additional discussion of the model behavior as a runaway glaciation is approached.

The thermodynamic sea ice model is based on the Community Sea Ice Model (Briegleb et al., 2004). The sea ice model calculates ice fractional coverage, ice
Figure 2.1. The evolution of sea ice fraction and atmosphere-to-ocean heat flux. Simulations are color coded by mean surface temperature from warm (red, $T_s = 296.3$) to cold (purple, $T_s = 230.2$) and correspond with equilibrated Archean climate simulations presented in the figure 3.5. As sea ice expands (a), the ocean heat flux convergence ($Q_{\text{flux}}$) (b) is reduced in the tropics while increasing at mid and high latitudes. Positive values for $Q_{\text{flux}}$ indicate a net transfer of heat from oceans to the atmosphere. Adjustments cause $Q_{\text{flux}}$ to increase preferentially near the sea ice boundary, qualitatively similar to predictions made by dynamic ocean and ice models. Asymmetries in the zonal mean sea ice distribution are caused by the asymmetric distribution of continents about the equator.
thickness, surface temperature, snow depth and an internal energy profile of the ice across four discrete layers. The energy balance equation at the top of the sea ice pack is given by

$$F_{\text{TOP}} = F_{SW} - I_{SW} + F_{LW} + F_{SH} + F_{LH} + k \frac{dT}{dz}$$  (eq. 2.2)

where $F_{\text{TOP}}$ is the net energy at the surface of the sea ice, $F_{SW}$ is the absorbed solar flux, $I_{SW}$ is the solar flux that penetrates the ice interior, $F_{LW}$ is the net longwave flux, $F_{SH}$ is the sensible heat flux, $F_{LH}$ is the latent heat flux, $k$ is the thermal conductivity and $dT/dZ$ describes the vertical gradient of temperature within the ice pack. Fluxes are taken to positive downward. When $F_{\text{TOP}} < 0$ sea ice will grows in thickness. Conversely when $F_{\text{TOP}} > 0$ melting occurs. While the atmosphere adjusts quickly, the ocean-sea ice system can take several decades or more to reach thermal equilibrium. The contribution of snow to the total surface albedo can take up to $\sim 100$ years to reach equilibrium (Mengel, 1988).

Ice and snow albedo parameterizations include simple dependencies on wavelength and surface temperature. The wavelength dependency is split into visible ($< 0.7 \mu$m) and near-infrared ($> 0.7 \mu$m) bands. For “cold” snow (sea ice) at surface temperatures below $-1^\circ$ C, the visible albedo is 0.96 (0.68) and the near-infrared albedo is 0.68 (0.30). As the surface temperature rises from $-1^\circ$ C to the melting point ($0^\circ$ C), snow and sea ice albedos decrease following a linear temperature dependency to simulate the darkening of snow and sea ice surfaces during the melt process (Collins et al., 2004; Yang et al., 2012a). For “warm” snow (sea ice) at surfaces temperatures of $0^\circ$ C, the visible albedo is 0.86 (0.61) and the
near-infrared albedo is 0.53 (0.23). Note that the albedo of land glaciers depends only on wavelength and is 0.80 in the visible and 0.55 in the near-infrared. Land glaciers are treated as fixed surface features and do not respond to temperature (i.e. no growth or melting). The albedo of snow covered surfaces is calculated as a weighted average between the snow covered and bare surface fractions. Note that land glaciers and sea ice are often subject to persistent snow cover. The albedo of open-ocean is 0.06 across all wavelengths.

2.3. Treatment of cloud physics

Over the past two decades cloud modeling has advanced tremendously such that realistic three dimensional cloud fields can be prognostically calculated in GCMs. Non-convective cloud processes are treated via the methods of Rasch and Kristjánsson (1998) and Zhang et al. (2003). Deep convection is treated following Zhang and McFarlane (1995) while shallow convection is handled by Hack (1994). There are two primary components of the CAM3 cloud parameterization; a macroscale component that determines the exchange of water between the vapor and condensate phases and a bulk microphysical component that controls the conversion of condensate to precipitate. The macroscale component controls the temperature, water vapor mixing ratio and total cloud condensate mixing ratio in each grid cell based on large-scale tendencies due to advection, expansion, radiative heating, condensation, evaporation and convective processes. The total cloud condensate is then partitioned into liquid and ice phases based on a temperature dependent parameterization. Water vapor and condensate that is detrained by
convective processes is added to the appropriate model layers before computation of cloud physics at each timestep.

The bulk microphysical model is primarily concerned with the conversion of cloud condensate to precipitation and subsequent evaporation of precipitation back into vapor form. The scheme follows closely that of Sundqvist (1988). Supersaturations are not allowed, the cloud number droplet concentrations are specified, and cloud particle sizes are specified based on temperature dependent parameterizations. In default settings, liquid cloud number droplet concentrations are set to 400 cm$^{-3}$ over land, 150 cm$^{-3}$ over ocean, and 75 cm$^{-3}$ over ice. Liquid cloud drop effective radii vary from 8 to 14 µm over land dependent on temperature, while effective radii are fixed at 14 µm over oceans and ice covered regions. Ice particle effective radii follow a strictly temperature dependent parameterization and can vary in size from a few tenths to a few hundred microns. (Collins et al., 2004). Typical cirrus clouds in the model have ice particle effective radii of ~50 µm at air temperatures of 240 K and ~20 µm at air temperatures of 200 K.

Cloud models used in GCMs universally suffer from the issue of sub-grid scale parameterizations. Even at the highest of resolutions, GCMs cannot capture the fine structure of cloud fields and thus appropriate parameterizations must be made. In CAM3 sub-grid scale processes are handled via determining a cloud fraction for each grid box. Cloud fraction is determined for three distinct types of clouds. Marine stratus clouds depend on the stratification of the atmosphere between the surface and the 700 mb level (Klein and Hartmann, 1993). Convective clouds
depend on the stability of the atmosphere and the upward convective mass flux (Xu and Krueger, 1991). All other clouds are determined based on relative humidity thresholds. Typical relative humidity thresholds vary between 0.8 and 0.9 and depend on the altitude regime and model resolution.

Note that the CAM3 cloud physics scheme has numerous tunable parameters; all of which have been adjusted to yield good results for the present day Earth (Collins et al., 2006). In this study tunable cloud parameters have been left as is, except where discussed in chapter 4. Thus we are making the implicit assumption that the cloud fields for the early Earth would have been “like” that of the present day. This assumption is not wholly inappropriate because many of fundamental influencing factors on clouds would have remained similar. The Archean, like the present day Earth, would have large reservoir of water (i.e. the oceans) in constant contact with the surface. As previously discussed, ocean temperatures were likely at least as warm as today, therefore tropospheric temperatures also likely remained in a similar temperature regime. Solar heating, though weaker, would have heated the Archean surface providing convective motion.

2.4. Treatment of radiative transfer

A new radiative transfer model has been implemented into CAM3 that is specifically tuned for anoxic, high-CO$_2$, and high-CH$_4$ atmospheres expected during the Archean period. The model uses a two-stream radiative transfer solver with multiple scattering (Toon et al., 1989). Molecular absorption by H$_2$O, CO$_2$, and CH$_4$ gases are treated using correlated $k$-distribution absorption coefficients that were
calculated using the Atmosphere Environment Research Inc. line-by-line model (LBLRTM) with access to the HITRAN 2004 spectroscopic database (Mlawer et al., 1997; Clough et al., 2005, Rothman et al., 2005). Overlapping molecular absorption is handled using an amount weighted scheme (Shi et al., 2009). Correlated k-distributions are calculated on 56 pressure levels ranging from 3.162 bar to 0.01 bar with successive pressure levels following \( \log_{10}(P_1/P_2) = 0.1 \). Eight equally spaced temperature levels are utilized from 80 to 360 K. A wide range in pressure and temperature space allows for accurate radiative transfer calculations for atmospheres vastly different from the present day, particularly with regard to the exceedingly cold stratospheric temperatures expected for an anoxic early Earth (von Paris et al., 2008). The longwave (10 – 2200 cm\(^{-1}\)) spectrum is divided into 13 spectral intervals while the shortwave (2200 – 50000 cm\(^{-1}\)) spectrum is divided into 15 spectral intervals (Table 2.1). The division between longwave and shortwave spectral regions is chosen such that solar radiation is largely confined to the shortwave, while terrestrial radiation is largely confined to the longwave. Each spectral interval has 8 gauss points except for the three spectral intervals encompassing 500 – 730 cm\(^{-1}\) which require 16 gauss points to improve accuracy in the stratosphere. In this spectral region CO\(_2\) absorbs very strongly. Low atmospheric pressure in the upper atmosphere amplifies small errors in flux when calculating radiative heating rates, thus extra spectral gauss intervals are required. Continuum absorption for CO\(_2\), N\(_2\), H\(_2\)O self-broadening and H\(_2\)O foreign broadening are fit to the MT_CKD continuum model which is implemented as part of LBLRTM (Clough et al., 2005). For low-CO\(_2\) continuum absorption is negligible, however it
<table>
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<tr>
<th>Interval</th>
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<th># gauss points</th>
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<td>H(_2)O, CO(_2)</td>
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</tr>
<tr>
<td>28</td>
<td>38000–50000</td>
<td>none</td>
<td>8</td>
</tr>
</tbody>
</table>

Table 2.1. Radiative transfer spectral intervals and absorbing species.
becomes increasingly important as \( p\text{CO}_2 \) rises above 0.1 bar (Halevy et al., 2009). While uncertainties remain regarding the proper theoretical treatment of \( \text{CO}_2 \) continuum absorption, the MT_CKD parameterization provides a median solution amongst popular continuum absorption models in use today (Halevy et al., 2009). Rayleigh scattering is included using the parameterization of Vardavas and Carver (1984). \( \text{CO}_2 \) is more than twice as effective at Rayleigh scattering than is \( \text{N}_2 \), therefore Rayleigh scattering becomes increasingly important in \( \text{CO}_2 \)-rich atmospheres. Both ice and liquid cloud particles are treated using Mie scattering. The radiative effect of overlapping cloud layers is treated using the Monte Carlo Independent Column Approximation under the assumption of maximum-random overlap (Pincus et al., 2003; Barker et al., 2008). The solar spectrum is taken to have the identical relative strength with wavenumber as present day and is evenly scaled down to a lower solar constant for Archean simulations.

The new radiative transfer code has been tested against LBLRTM to ensure accuracy for expected Archean greenhouse gas inventories. Figure 2.2 shows the clear-sky downwelling longwave flux at the surface and the clear-sky net longwave flux at the tropopause (defined here as the 200 mb level) calculated for varying \( p\text{CO}_2 \) and \( p\text{CH}_4 \) applied to the US 1976 Standard Atmosphere profile but with ozone and oxygen removed. The net broadband flux at the tropopause is taken to be positive downwards. The net flux at the tropopause gives a good indication of the correlation between surface temperature changes and varying greenhouse gas amounts while the downwelling flux at the surface gives a better indication of the absolute accuracy of the radiative transfer scheme (Goldblatt et al., 2009a). Both
Figure 2.2. Radiative transfer flux validations for increasing CO$_2$ and CH$_4$.
Longwave radiative fluxes from line-by-line calculations (LBLRTM, dashed blue) and from our model (Archean RT, red) assuming the US 1976 Standard Atmosphere but with oxygen and ozone removed. Panels (a) and (b) show the surface downwelling longwave flux and percent error compared with line-by-line calculations for varying CO$_2$ and CH$_4$ respectively. Panels (c) and (d) show the net longwave flux at the tropopause and percent error compared with line-by-line calculations for varying CO$_2$ and CH$_4$. Flux errors in our model remain less than ~1.5% compared with line-by-line calculations.
surface and net flux calculations are accurate to within \(~1.5\%\) of line-by-line results up through end-member cases of \(pCO_2 = 0.3\) bar and \(pCH_4 = 0.01\) bar. Figure 2.3 illustrates various atmospheric profiles of interest. The first panel shows the US 1976 Standard temperature profile (as used for flux calculations in figure 2.2) and an anoxic temperature profile typical of the Archean (\(i.e.\) with oxygen and ozone removed. The second panel shows vertical profiles of water vapor, CO\(_2\), and CH\(_4\) of a typical Archean configuration. In this case CO\(_2\) has been set to 0.06 bar (60,000 ppm), CH\(_4\) has been set to 10 ppm while atmospheric water vapor is determined by the thermal structure. Figure 2.4 shows longwave flux and cooling rate profiles from our correlated \(k\)-distribution radiative transfer scheme and from LBLRTM. The profiles are calculated using the Archean temperature profile shown in figure 2.3a and the greenhouse gas constituent profiles shown in figure 2.3b.

At present we do not treat non-local thermodynamic equilibrium (LTE) processes, thus radiative heating and cooling rates are strictly valid only for atmospheric pressures greater than \(~0.05\) bar (\(~51\) km in anoxic simulations, \(~65\) km in present day simulations) where LTE conditions are met (Fomichev et al., 1998). Above this level, the assumption of LTE begins to break down with decreasing pressure and additional physics is needed to obtain accurate heating/cooling rate profiles. Note that gravity waves, a tunable parameter of the model, control the thermal structure of the upper stratosphere and mesosphere but have no discernable effect on surface climate (Bardeen et al., 2010). For simulations under the present day solar constant, ozone and oxygen absorption coefficients are
added to correlated $k$-distributions, requiring a remapping of spectral intervals presented in Table 2.1.
Figure 2.3. Vertical profiles of temperature and greenhouse gases used for radiative transfer validation. The left panel shows vertical profiles for the anoxic Archean atmosphere compared with the US 1976 standard atmosphere. The right panel shows vertical profiles for water vapor, CO$_2$ and CH$_4$ used for radiative transfer validations in figure 2.4.
Figure 2.4. Radiative transfer flux and heating rate validations against Archean profiles. The top row shows longwave upwelling flux, longwave downwelling flux, longwave net flux, and longwave cooling rates from our radiative transfer compared with line-by-line calculations. The bottom row shows the difference between our radiative transfer code and line-by-line calculations.
Evidence from ancient sediments indicates that liquid water and primitive life were present during the Archean despite the faint young Sun. To date studies of Archean climate typically utilize simplified one-dimensional models that ignore clouds and ice. Here we use an atmospheric general circulation model coupled to a mixed layer ocean model to simulate the climate circa 2.8 billion years ago when the Sun was 20 percent dimmer than today. Surface properties are assumed to be equal to present day while ocean heat transport varies as a function of sea ice extent. Present climate is duplicated with 0.06 bar of CO$_2$ or alternatively with 0.02 bar of CO$_2$ and 0.001 bar of CH$_4$. Hot Archean climates, as implied by some isotopic reconstructions of ancient marine cherts, are unattainable even in our warmest simulation having 0.2 bar of CO$_2$ and 0.001 bar of CH$_4$. However, cooler climates with significant polar ice, but still dominated by open ocean, can be maintained with modest greenhouse gas amounts, posing no contradiction with CO$_2$ constraints deduced from paleosols or with practical limitations on CH$_4$ due to the formation of optically thick organic hazes. Our results indicate that a weak version of the faint young Sun paradox, requiring only that some portion of the planet’s surface maintain liquid water, may be resolved with moderate greenhouse gas inventories. Thus, hospitable late Archean climates are easily obtained in our climate model.
The simplest possible solution to the faint young Sun paradox is to
counteract the weak Sun by adding large amounts of greenhouse gases. For our first
set of simulations we isolate the effects of a reduced solar constant and increased
greenhouse gases. In this chapter we concentrate on the climate of the late Archean
(circa 2.8 Ga) having a solar constant of 1093.6 W m\(^{-2}\), 80\% of the present day value.
This time period and solar constant is chosen as to facilitate comparison with recent
literature that focuses on the same time period using a radiative convective model
(Haqq-Misra et al., 2008). Geochemical evidence indicates that CO\(_2\) and CH\(_4\)
amounts were generally larger in the distant past and thus may feasibly provide a
solution to the faint young Sun paradox.

For all simulations presented in this chapter, the mean sea level pressure is
taken to be 1.013 bar. The composition of the Archean atmosphere is assumed to be
0.00933 bar of argon gas along with variable amounts of CO\(_2\), CH\(_4\) and N\(_2\). Oxygen
and ozone are removed. Partial pressures of CO\(_2\) and CH\(_4\) are varied while N\(_2\) is
calculated as \(pN_2 = 1.013 - pCO_2 - pCH_4 - pAr\) in units of bars. Atmospheric CO\(_2\) is
varied up to a maximum of 0.2 bar while CH\(_4\) is taken to be either 0 or 10\(^{-3}\) bar. Note
that given our selections for mean sea level pressure, \(pCO_2\) and \(pCH_4\), \(pN_2\) is equal to
or slightly greater than its present atmospheric level in all simulations presented
here. Estimates of the late Archean surface pressure from fossilized raindrop
imprints suggest that it was probably not much different than today, though the
authors leave open the possibility that \(pN_2\) could have been as much as twice the
present atmospheric level as an upper limit (Som et al., 2012). Continental
configurations, topography, planetary rotation rate, ocean heat transport, cloud
droplet sizes, land based glacial ice, and surface vegetation are assumed to be those of the present day. Precise changes to these climate system elements remain speculative at best (Rosing et al., 2010), but changes to the solar constant and greenhouse gas inventories are supported strongly by astrophysical theory and geochemical evidence. Thus we begin our examination of Archean climate by looking only at the effects of reduced solar insolation and increased greenhouse forcing. By following this methodology we are making the implicit assumption that many elements of the late Archean were “like” present-day Earth, an assumption also inherent in the surface albedo tuning procedure used in RCM paleoclimate studies (Kasting et al., 1984; Haqq-Misra et al., 2008).

3.2. Standard atmospheres

Our simulations replicate modern climate with 360 ppm of CO₂, 1.7 ppm of CH₄, and a solar constant of 1367.0 W m⁻², yielding a global and annual mean surface temperature of 287.9 K and sea ice margin of 68.5°. Here we calculate annual and global mean clear-sky radiative forcings. Radiative forcing is defined as the change in net downwelling radiative flux (F_{down} − F_{up}) at the tropopause caused by the introduction of a forcing agent (Hansen et al., 2005). The tropopause is defined as the lowest model level where the lapse rate (−dT/dZ) is 2 K/km or less (WMO, 1957). We define radiative forcings relative to our simulation of modern climate. Radiative forcing is a useful concept for estimating first order changes to the planet’s surface temperature caused by perturbations to the radiation field. However, the complex interaction of climate feedbacks with imposed radiative
forcings ultimately determines the exact radiative forcing–surface temperature response.

We first performed a simulation using modern CO₂ and CH₄ concentrations after allowing the stratosphere to adjust to the removal of oxygen and ozone, but with all other aspects of climate held fixed. Molecular nitrogen is increased to maintain the present day mean sea level pressure of 1.013 bar. In this simulation the global and annual mean surface temperature is 287.4 K and the sea ice margin is 67.8° (see Table 3.1). Removing oxygen and ozone allows more solar radiation to reach the surface, yielding a small shortwave radiative forcing of +1.2 W m⁻². However removing O₃ greatly reduces stratospheric temperatures thus reducing downwelling longwave fluxes emanating from the upper atmosphere by ~2 W m⁻². Combining the reduced radiation due to the stratospheric temperature change with the loss of ozone’s greenhouse contribution, the removal of O₃ yields a longwave radiative forcing of −5.2 W m⁻².

Archean simulations assume a solar constant 80% of the present day value. This equates to a clear-sky net downwelling radiative forcing of −56.7 W m⁻². In our model we can achieve a global and annual mean surface temperature of 287.9 K, with 80% of the present day solar constant assuming a CO₂ partial pressure of 0.06 bar (60,000 ppm), hence forth regarded as our “standard” Archean atmosphere (see Table 3.1). Increasing CO₂ from 360 ppm to 0.06 bar yields a longwave radiative forcing of +46.4 W m⁻². However, absorption of solar energy in the near-IR and enhanced rayleigh scattering, both due to increased atmospheric CO₂, results in an additional shortwave radiative forcing of −2.7 W m⁻². The total change in global
Table 3.1. Standard atmospheres.
mean radiative forcing between our present day and standard Archean simulations is calculated by summing the radiative forcings due to the removal of oxygen and ozone (+1.2 W m\(^{-2}\) shortwave, −5.2 W m\(^{-2}\) longwave), the reduction to the solar constant (−56.7 W m\(^{-2}\) shortwave), and the increase in atmospheric CO\(_2\) (−2.7 W m\(^{-2}\) shortwave, +46.4 W m\(^{-2}\) longwave). The remaining global mean energy deficit of −17.0 W m\(^{-2}\) is accounted for through feedbacks involving the Archean hydrological cycle.

The Archean hydrological cycle is altered by fundamental changes to the radiative forcing at the surface caused by weak solar radiation and a strong greenhouse (Figure 3.1a). The positive radiative forcing imposed by increasing the concentration of a well mixed greenhouse species (in this case CO\(_2\)) has a weak latitudinal gradient compared with the negative forcing imposed by decreasing the solar constant, particularly across low latitudes. Weak radiant energy reaching the Archean surface inhibits the hydrological cycle by reducing surface latent heat fluxes (\textit{i.e.} evaporation of water from ocean surfaces) over low latitudes compared with the present day despite mean tropical ocean temperatures that differ by less than 1 K (Figure 3.1b). Zonal mean latent heat fluxes are reduced by on the order of 10 W m\(^{-2}\) across the tropics. Reduced latent heat fluxes naturally result in reduced specific humidities in the lower troposphere of the Archean compared with present day (Figure 3.2b). Similar boundary layer temperatures ensure that the Archean also has reduced relative humidities below ~4 km (Figure 3.2c). The reduction in moisture in lower troposphere in turn leads to reductions in moist convective mass and water fluxes (Figure 3.3) and reductions to low level clouds (below 700 mb)
Figure 3.1. Difference in zonal mean surface forcings and fluxes between Archean and present day atmospheres. Zonal mean broadband surface forcing difference (a) and surface energy flux difference (b) between standard Archean and present day atmospheres. A value of zero (dashed purple) means that the surface forcing for the late Archean and present day atmospheres is identical. Positive values for surface forcing indicate relative warming while negative values indicate relative cooling for the Archean compared to present day. A large negative surface forcing caused by a 20% reduction in the solar constant (red) is offset by increased greenhouse forcing from 0.06 bar of CO$_2$ (dark blue). A slow down of the Archean hydrological cycle reduces water clouds and shortwave cloud forcing, allowing a greater fraction of the incident solar radiation to reach the tropical surface than at present (light blue). Despite identical mean surface temperatures, our standard Archean atmosphere receives more energy at the polar surface and less energy at the tropical surface compared with present day (dash-dot black). Surface latent (blue) and sensible heat fluxes (red) are decreased compared with the present day atmosphere coincident with reductions to the surface radiative forcing. Total (dash-dot black) surface fluxes are reduced by $\sim 17$ W m$^{-2}$ at the equator, however note that reductions to latent heat fluxes dominate.
Figure 3.2. Vertical profiles of temperature, specific humidity, and relative humidity for Archean and modern atmospheres. Global and annual mean vertical profiles of temperature (a, d), specific humidity (b, e), and relative humidity (c, f) for Archean (red), present day (purple), and the present day atmosphere but with oxygen and ozone removed (light blue, dashed). The top panels show profiles for the troposphere and the bottom panels show profiles for the whole atmosphere up to a height of 60 km.
(Figure 3.4c). However above the boundary layer the situation reverses. Our
standard Archean atmosphere has slightly colder temperatures in the upper
troposphere compared with present day. Since the saturation vapor pressure
depends exponentially on temperature, a small decrease in air temperature yields
significant changes in relative humidity for a given amount of water vapor. Lower
temperatures and larger relative humidities aloft imply a greater potential for latent
heat release and thus reduced static stabilities. Thus, while shallow boundary layer
convection remains weak, deep convective mass and water fluxes are enhanced for
the Archean (Figure 3.3). Increased convection aloft helps support the higher
altitude clouds decks in the Archean atmosphere despite an atmosphere with a
reduced total water vapor column. Middle clouds (700 to 400 mb) are
approximately equal to their present day value while high clouds (400 mb and
upwards) occur with an increased frequency (Figure 3.4a, b).

For our standard Archean atmosphere, the climatological mean atmospheric
water vapor column is reduced by ~16% and the cloud water column is reduced by
~26% compared with simulations of present day. Global mean low cloud fractions
and cloud albedos are reduced in nearly exact proportion with the reduction in the
solar constant despite identical global mean surface temperatures, reinforcing the
importance of solar energy for driving the hydrological cycle. While the water vapor
greenhouse is weakened, reductions to low cloud fractions (Figure 3.4c) greatly
reduce the magnitude of the shortwave cloud forcing at the tropopause (Figure
3.4e). The magnitude of the shortwave cloud forcing is reduced by 20.6 W m⁻²
averaged globally and ~30 W m⁻² over equatorial regions, coincident with the
Figure 3.3. Moist convective mass and water fluxes for Archean and modern atmospheres. (a) Tropical moist convective mass flux and (b) moist convective water flux for the Archean (red), present day (purple), and the present day atmosphere but with oxygen and ozone removed (dashed light blue). Profiles are averaged between 22° S and 22° N latitudes. The reduction to surface latent heat flux inhibits shallow convection for our standard Archean atmosphere. However, a generally steeper lapse rate and higher relative humidities aloft allow convection to penetrate to higher altitudes.
Figure 3.4. Zonal mean cloud fractions and forcings for Archean and modern atmospheres. Zonal mean cloud properties for the Archean (red), present day (purple), and the present day atmosphere but with oxygen and ozone removed (dashed light blue). (a) Vertically integrated high cloud fraction describes clouds above 400 mb \((Z > 6 \text{ km})\) (b) Vertically integrated middle cloud fraction describes clouds between 700 and 400 mb \((2 < Z < 6 \text{ km})\), (c) Vertically integrated low cloud fraction describes clouds below 700 mb \((Z < 2 \text{ km})\). (d) Longwave cloud forcing, 10–2200 cm\(^{-1}\). (e) Shortwave cloud forcing, 2200–50000 cm\(^{-1}\). (f) Net cloud forcing, longwave plus shortwave.
largest decreases in the surface latent heat flux. Note that reductions to Archean shortwave cloud forcings are compounded by the fact that there is less incoming solar radiation to reflect. Thus, changes to Archean shortwave cloud forcings appear larger compared to present day values than they would if the solar constant had not been decreased. For comparison, the 19.6% reduction to the cloud albedo found for our standard Archean atmosphere would equate to a 10.8 W m\(^{-2}\) reduction to the shortwave cloud forcing if the solar constant remained equal to the present day value. Fewer water clouds over the Archean tropics partially counteracts the latitudinal differences between Archean solar and greenhouse radiative forcings by allowing a greater fraction of incident solar energy to reach the tropical surface. Nonetheless our standard Archean atmosphere still receives less total radiant energy at the equatorial surface while receiving more radiant energy in polar regions compared with the present day (Figure 3.1a). Note that the reduction in the total (CO\(_2\) + solar + cloud) surface radiative forcing in the tropics is virtually mirrored by reductions to the total (sensible + latent) surface to atmosphere energy flux (Figure 3.1b).

The removal of O\(_3\) and the addition of high CO\(_2\) cause the Archean stratosphere to become very cold and dry with minimum global mean temperatures of \(~150\) K at an altitude of 35 km (Figure 3.2d). This is nearly a 100 K reduction compared with the present day stratospheric temperature maximum. In essence, the Archean lacks a true stratosphere as we think of it today. Although a tropopause as defined by the decrease of the lapse rate still exists, a tropopause defined by a sharp temperature minimum does not exist. Note that the broad temperature
minimum occurs well above the lapse rate tropopause. Below ~25 km, temperature differences are predominantly due to a lack of shortwave heating from the removal of O₃, however note that some additional cooling from high CO₂ is still present. Above ~25 km radiative cooling from high CO₂ becomes increasingly important. At 60 km, cooling of the atmosphere from the removal of O₃ and from increased CO₂ becomes approximately equal. Exceedingly low temperatures above the tropopause mean that the saturation vapor pressure here is quite low. Thus, despite having little water vapor, relative humidities in the anoxic simulations grow large above the tropopause and a tenuous band of high altitude ice clouds is diagnosed located near the temperature minimum. The temperature dependent parameterization of ice cloud particle radii ensures that particles here are quite small (~0.5 μm), limiting their infrared opacity in our model. Both our standard Archean and anoxic present day simulations have increased high cloud fractions (Figure 3.4a) and increased total ice cloud condensate compared with present day. However, for the Archean longwave cloud forcings are slightly reduced compared with the present day (Figure 3.4d). The contribution of ice clouds to the greenhouse effect is mitigated by high concentrations of CO₂ which saturates longwave spectral bands, reducing the greenhouse effect from Archean ice clouds by 4.8 W m⁻² compared with present day despite increased ice cloud fractions and cloud condensate amounts. Conversely, present day simulations but with oxygen and ozone removed enjoy a 3.9 W m⁻² gain in longwave radiative forcing from clouds, mostly compensating for the loss of greenhouse warming from ozone and the stratospheric inversion. Combining effects from liquid and ice clouds, our simulation of present day Earth has a global
mean net cloud radiative forcing of \(-24.9\) W m\(^{-2}\), in general agreement with observations and baseline CAM3 simulations (Collins et al., 2006). However, for our standard Archean atmosphere the global mean net cloud forcing is only \(-9.1\) W m\(^{-2}\). Thus an additional flux at the tropopause of +15.8 W m\(^{-2}\) is gained for the Archean compared with present day due to changes to the radiative interaction of clouds.

### 3.3. Model sensitivity

In order to understand the response of climate to changing greenhouse forcings, simulations are conducted over a wide range of atmospheric CO\(_2\) under both late Archean and present day solar constants. Here we consider climate sensitivities as a function of broadband (shortwave plus longwave) clear-sky radiative forcing implied by varying CO\(_2\) concentrations relative to Archean and present day standard atmospheres respectively (Table 3.1). Climate sensitivity is the change in the global mean surface temperature for a given change in radiative forcing. By plotting climatological data versus radiative forcing instead of versus CO\(_2\) concentration, we have effectively normalized our results allowing for a direct comparison of simulations under present day and late Archean solar constants. While CO\(_2\) strongly affects longwave radiation by the absorption of terrestrial radiation, CO\(_2\) also imparts a small negative forcing on solar radiation by enhancing Rayleigh scattering. Our use of a mixed layer ocean model means that the model captures only the short-term (i.e. decadal to century timescale) response of the climate system to changes in radiative forcing. The response of ocean heat fluxes to changing sea ice coverage is identical for both sets of simulations, under the late
Archean and present day solar constant (Figure 2.1). The response of slow processes such as deep ocean circulations and continental glacier formation are not represented in this study. Mean surface temperatures \( (T_s) \), sea ice extent \( (\phi_{\text{ice}}) \), and climate sensitivity \( (\lambda) \) vary quasi-linearly with radiative forcing for present day solar constant simulations (Figure 3.5a, b, c). The correlation between \( p\text{CO}_2 \) and radiative forcing is indicated in figure 3.5e. Archean simulations exhibit greater climate sensitivity and more complex behavior compared with simulations under the present day solar constant. Comparatively larger climate sensitivities for the Archean suggest a volatile climate where a gentle nudge can yield dramatic climate change relative to present day conditions. While climate sensitivities remain relatively moderate in amplitude and linear with radiative forcing for a warmer late Archean \( (T_s \geq 280 \text{ K}) \), a cooling Archean climate experiences two accelerated climatic transitions indicated by the maxima in figure 3.5c. Sensitivity maxima mark tipping points between climate states of relative stability. Simulations under the present day solar constant exhibit no such abrupt transitions as \( \text{CO}_2 \) is reduced down to our end-member simulation with 1 ppm.

The maxima in climate sensitivity in figure 3.5c centered at \(-12.1 \text{ W m}^{-2}\) marks a transition from a state where sea ice is confined to polar regions to a state where sea ice can extend into mid-latitudes. The sharp increase in Archean climate sensitivity for radiative forcings between \(-5\) and \(-12 \text{ W m}^{-2}\) is caused by fundamental changes to the latitudinal distribution of energy under a faint Sun and strong greenhouse. As discussed in section 3.2, our standard Archean atmosphere receives less radiant energy at the equatorial surface while receiving more radiant
Figure 3.5. Climate sensitivity under changing CO$_2$ for Archean and modern atmospheres. Climate response to changes in clear-sky radiative forcing at the tropopause under present day and late Archean solar constants (1367.0 W m$^{-2}$ and
1093.6 W m$^{-2}$ respectively). Radiative forcings are applied by varying $p$CO$_2$ for present day with oxygen and ozone (purple) and late Archean (red) simulations, referenced from standard atmospheres (see Table 2). A radiative forcing of zero (vertical dotted line) indicates standard atmospheric states for the late Archean and for present day Earth, both having $T_s = 287.9$ K. (a) Mean surface temperature ($T_s$) versus radiative forcing. (b) Annual and zonal mean sea ice margin ($\phi_{\text{ice}}$) versus radiative forcing. The sea ice margin is calculated as the inverse sine of the global and annual mean open ocean fraction. (c) Archean climate sensitivities ($\lambda$) are larger and more complex compared with present day. Maxima indicate tipping points between quasi-stable climate states. (d) Annual, zonal, and hemispheric mean meridional temperature differences ($\Delta T$) between the tropics ($22^\circ$) and the arctic ($74^\circ$) versus radiative forcing. (e) CO$_2$ partial pressure versus radiative forcing.
energy at the poles compared with the present day (Figure 3.1). This fundamental modulation of the latitudinal surface energy distribution causes Archean simulations with surface temperatures close to present day to have reduced meridional temperature differences (Figure 3.5d) and less sea ice (Figure 3.5b) compared with simulations under the modern Sun. The meridional temperature difference shown in figure 3.5d is calculated as the annual, zonal and hemispheric mean surface temperature difference between grid cells centered at 22° and 74° latitude. Reduced latitudinal temperature differences permit ice to advance (or retreat) more easily for a given radiative forcing (Pierrehumbert, 2002). In this study a reduction in Archean $p_{\text{CO}_2}$ from 0.03 to 0.02 bar, a radiative forcing of $-4.3 \text{ W m}^{-2}$, causes an annual mean surface cooling of 8.6 K and an annual and hemispheric mean equatorward expansion of sea ice by 12° latitude. By contrast, for simulations of the present day Earth reducing $\text{CO}_2$ from 360 to 180 ppm yields a nearly identical radiative forcing of $-4.4 \text{ W m}^{-2}$, however mean surface cooling is only 2.3 K and sea ice expands equatorward by only 2°. If Archean ice sheets are allowed to expand into the mid latitudes, mean surface temperatures plummet, meridonal temperature differences increase due to polar amplification, and the ice-albedo feedback weakens. A larger meridonal temperature gradient makes it more difficult to form ice at low latitude for a given mean surface temperature (Pierrehumbert, 2002).

While Archean ice sheets can expand easily from high into mid latitudes, when ice enters into the subtropics (approximately 23° – 40° latitude) the ice-albedo feedback weakens and the climate can stabilize against immediate runaway
glaciation. The strength of the ice-albedo feedback is critically dependent upon the albedo contrast between adjacent ice-free and ice-covered surfaces (Abbot et al., 2011; Lewis et al., 2006; Yang et al., 2012). Here, the broadband cold (warm) snow albedo is 0.79 (0.69), the broadband sea ice albedo is 0.45 (0.37), the open ocean albedo is 0.06, and the land albedo is dependent upon surface type and snow cover. While these values compare well with modern observations, ice surface albedo parameterizations in GCMs remain relatively crude (Warren et al., 2002).

Atmospheric dynamics dictate that the subtropics are dominated by desert climates with high evaporation and low precipitation. Here, encroaching sea ice can remain free from snow cover throughout most of the year since any snowfall will sublimate away quickly (Abbot et al., 2011). Bare sea ice has a much lower albedo than does fresh snowfall. As ice enters the subtropics, a narrow boundary of low-albedo bare sea ice develops at the leading edge of ice sheets. The diminished contrast between ocean and bare sea ice vis-à-vis fresh snowfall acts as a powerful limitation on the ice-albedo feedback mechanism. Note that this mechanism has been proposed to allow exceedingly narrow but stable waterbelt climates (Abott et al., 2011). Figure 3.6 illustrates this mechanism in action. Sea ice reaching into the subtropics is also more susceptible to the formation of melt ponds as opposed to ice present at high latitudes due to more direct solar radiation, thus further lowering the surface albedo at the leading edge of the ice. Additionally, as climate cools latent heat fluxes are greatly diminished with preferential decreases over the tropical oceans. This allows the tropical oceans to maintain more heat and resist ice formation (Pierrehumbert, 2002).
Figure 3.6. Time and temperature evolution of surface albedo, sea ice and snow cover. Annual cycle of latitudinal variations in surface albedo for Archean simulations with mean surface temperatures ranging from 289.1 K to 253.6 K. The solid white line is the sea ice margin. The dashed white line is the edge of snow cover. Snow cover has a substantially higher albedo than does bare sea-ice. As the sea ice margin nears 30°latitude, a thin buffer of relatively low albedo bare sea ice develops, slowing the ice-albedo feedback.
The maxima in climate sensitivity in figure 3.5c at -45.9 W m$^{-2}$ marks the final transition into a completely ice covered world. In our model this corresponds to a glaciation threshold between 300 and 500 ppm of CO$_2$ for the Archean climate. While at face value this is certainly an intriguing result, here the observed climate stability down to low-CO$_2$ is likely attributed to the absence of a dynamical sea ice model. While plausible changes to ocean heat transport may only have a small effect on the tipping point towards runaway glaciation, the equatorward flow of sea ice is a key element in triggering a snowball Earth (Voigt and Abbot, 2012). Even so, GCMs that include dynamic oceans and sea ice still commonly find stable waterbelt climates with sea ice margins located in the tropics (Voigt and Abbot, 2012; Yang et al., 2012; Yang and Peltier, 2012). Furthermore, while we argue here that limitations of the Archean geologic record make it inappropriate to preclude climates with relatively larger polar ice caps compared with the present day, waterbelt climate states where the surface experiences glaciation covering 80% or more of the surface may stretch the believable limits for cold Archean climate solutions. Considering the limitations of our model, here we take a conservative approach and limit the scope of acceptable Archean climate solutions to those with stable sea ice margins poleward of 30° degrees latitude (i.e. with 50% open-ocean or greater). Future studies using coupled dynamic ocean and sea ice models should be used to better estimate tipping points towards complete glaciation.
3.4. Hospitable Archean climates

Hospitable late Archean climates, dominated by open-ocean, are permitted safely within constraints imposed on paleoatmospheric CO$_2$. Here we have adopted the constraint of Driese et al. (2011) which limits paleoatmospheric CO$_2$ at 2.69 Ga to between 10 and 50 PAL (0.0036 – 0.018 bar), with a best guess value of 41 PAL (~0.015 bar). With 0.015 bar of CO$_2$ and no CH$_4$, the global mean surface temperature stays below freezing (Figure 3.7a), however surface temperatures averaged across the tropics reach 284 K (Figure 3.7b) and the sea ice margin stabilizes at 46° latitude (Figure 3.7c). In this case ~72% of the planet surface area remains free from ice. With 0.015 bar of CO$_2$ and 0.001 bar of CH$_4$, the global mean surface temperature is 285 K while mean tropical surface temperatures reach 296 K, only a few degrees colder than present day. The sea ice margin is stable at 68° latitude and thus ~93% of the surface would remain free from ice. In this case photochemical hazes would not be expected to form since the CH4/CO2 < 0.1. Thus a relatively warm, ocean dominated late Archean poses no conflict with imposed constraints on greenhouse gases. Stable cooler climates with more extensive ice coverage are possible at lower CO$_2$ (and CH$_4$). Stable climates with 0.005 bar of CO$_2$ and no CH$_4$ are found having $T_s = 262$ K and a sea ice margin at 38° latitude (~62% of surface free from ice). In this case increasing CO$_2$ from 360 ppm to 0.005 bar yields a radiative forcing of +16.9 W m$^{-2}$ while reductions to low clouds are compounded by cool surface temperatures resulting in a net cloud forcing of −0.9 W m$^{-2}$, a gain of 24.0 W m$^{-2}$ at the tropopause compared with present climate. A range of hospitable climate solutions exist with 0.005 < $p$CO$_2$ < 0.015 bar which have
Figure 3.7. Late Archean global mean surface temperature, mean tropical temperature and sea ice margin versus partial pressure of CO₂. Archean climate response to varying CO₂ partial pressure. (a) Mean surface temperature ($T_s$) versus $p\text{CO}_2$ from this study (red) and from a recent RCM model study (blue, Haqq-Misra et al., 2008) for 0 and 0.001 bar of CH₄ respectively under 80% of the modern solar constant. A recent estimate for paleoatmospheric CO₂ abundance dated to 2.69 Ga is indicated at $p\text{CO}_2 = 0.015$ bar (green, Driese et al., 2011). (b) Mean tropical temperature ($T_{trop}$) between 22° S and 22° N latitudes for GCM simulations. (c) Annual and hemispheric mean sea ice margin ($\phi_{ice}$) for GCM simulations. Purple dashed lines indicate the values from simulations of present day climate.
significant polar ice caps but are still dominated by open ocean. Within this range of
paleoatmospheric CO$_2$, the addition of smaller amounts of methane (10$^{-4}$ bar or less)
can contribute up to $\sim$6 K of further warming without initiating photochemical haze
formation (Haqq-Misra et al., 2008). If we are allowed to consider climates with
large polar ice caps as valid solutions, resolving the faint young Sun paradox within
constraints on greenhouse gases becomes remarkably easy.

Our Archean general circulation model exhibits greater sensitivity to
changing CO$_2$ compared with published results from paleoclimate simulations
utilizing 1D radiative-convective models (Haqq-Misra et al., 2008). This comes as no
surprise since RCMs used for paleoclimate simulations typically set the surface
albedo to a constant, thus ice-albedo and cloud feedbacks to changing surface
temperature are not included (Kasting et al., 1984). Here, as expected we find that
the temperature versus $p$CO$_2$ curve is consistently steeper for GCM simulations than
for RCM simulations (Figure 3.7a). Nonetheless the qualitative similarity between
the model surface temperature responses from GCMs and RCMs (i.e. fairly smooth
and lacking sharp bifurcations) is encouraging but also somewhat surprising. First
order differences between GCM and RCM simulated mean surface temperatures can
be understood in terms of modulations to the albedo and cloud forcings (Figure 3.8).
RCMs parameterize the net radiative effect of clouds and ice within the surface
albedo, typically setting it to a constant value of $\sim$0.22 in all simulations (Haqq-
Misra et al., 2008). In our GCM as CO$_2$ is reduced from 0.2 to 0.005 bar, global mean
surface temperatures fall from 296 to 262 K, the sea ice margin expands from 82$^\circ$ to
38$^\circ$ latitude and the global mean surface albedo increases from 0.11 to 0.32. The
difference in mean surface temperatures between GCM and RCM simulations mirror the variation in surface albedo determined by our GCM relative to the constant value for surface albedo used in RCMs indicating the importance of the ice-albedo feedback (Figure 3.8a). However, cloudiness and Rayleigh scattering moderate the contribution of the surface albedo to the TOA albedo. Both cloud and Rayleigh scattering albedos act to oppose increases to the surface albedo as CO₂ and surface temperature analogously decrease. Rayleigh scattering is directly related to the atmospheric CO₂ content. The global mean cloud albedo varies with surface temperature. On a cold Archean world the hydrological cycle weakens because the saturation vapor pressure varies sharply with temperature. Optically thick water clouds are eliminated in favor of tenuous ice clouds. While the longwave cloud forcing remains relatively constant, the shortwave and net cloud forcings decrease in magnitude with decreasing surface temperature (Figure 3.8b). The persistence of ice clouds (vis-a-vi liquid clouds) as the climate cools serves as a buffer against runaway glaciation for cold early climates in support of earlier theories (Rossow et al., 1982). However, here this effect is weak and does not provide a full solution to the faint young Sun paradox. As the planet cools a larger percentage of the total greenhouse effect comes from clouds rather than from gaseous absorption. For colder climates 20% or more of the total greenhouse effect can come from ice clouds, while for Archean climates with temperatures near present day (boosted by CO₂), ice clouds contribute slightly more than 12% of the total greenhouse (Figure 3.8c). Combining effects of surface, cloud, and atmosphere, the TOA albedo exhibits a much more modest increase (0.31 to 0.42) than does the surface albedo over our
Figure 3.8. Late Archean global mean albedo, cloud forcing, and greenhouse components versus partial pressure of CO$_2$. (a) Global mean top-of-atmosphere (TOA), surface, cloud, and Rayleigh scattering albedos versus atmospheric CO$_2$ for Archean simulations, analogous with figure 8. Radiative-convective models typically use an effective surface albedo of $\sim$0.22 to implicitly incorporates cloud forcings. (b) Cloud longwave, shortwave and net forcing at the tropopause versus atmospheric CO$_2$. (c) Percent contribution of gaseous and cloud absorption to the total greenhouse effect.
range in climate. RCMs change neither surface nor cloud albedo, and therefore accidentally benefit from their partial cancellation, allowing RCM and GCM temperature curves to remain surprisingly similar despite the use of vastly different methods.

An often overlooked shortcoming of RCM paleoclimate studies is that they typically assume that if the mean surface temperature falls below 273 K (Haqq-Misra et al., 2008; Domagal-Goldman et al., 2008) or sometimes 279 K (Goldblatt et al., 2009b) that runaway glaciation to a hard snowball will be the inevitable result. However, in this study a sharp bifurcation to a frozen world is not observed when $T_s$ drops below 273 K. Here we find stable climates with $T_s$ as low as 260 K that maintain open ocean fractions of greater than 50%. Similar climate solutions with $T_s < 273$ K but with open oceans at low and mid latitudes are also commonly found in GCMs that use more complex treatments of oceans and sea ice albeit within the context of different geologic epochs and radiative forcings (Voigt et al., 2011; Yang et al., 2012; Yang and Peltier, 2012; Voigt and Abbot, 2012). Stable climates may even be possible with sea ice margins as low as $10^\circ$ to $20^\circ$ latitude and global mean surface temperatures well below freezing (Abbot et al., 2011; Yang et al., 2012). Radiative convective models remain attractive research tools since they are computationally fast and can incorporate comprehensive self-consistent reducing atmospheric chemistry. The utility of paleoclimate RCMs may be expanded by incorporating simple cloud and ice-albedo feedback parameterizations and through the realization that solutions with $T_s < 273$ K remain viable for a habitable early Earth with substantial liquid water at the surface.
3.5. Discussion

The isotopic composition of marine sediments commonly serves as a proxy for paleo-ocean temperatures. The oxygen and silicon isotopic composition of seawater and water-lain sediments is controlled by temperature dependent exchange of $^{18}$O between water and rock within hydrothermal vent systems and mid-ocean ridges (Kasting et al., 2006). Taken at face value, low-$\delta^{18}$O and $\delta^{30}$Si inclusions are interpreted as indicators of hot seawater temperatures, possibly reaching as high as 350 K during the late Archean (Knauth and Lowe, 2003; Robert and Chaussidon, 2006). However, observed low-$\delta^{18}$O signals found in Archean sediments may be the result of variations in hydrothermal fluid temperature through time rather than variations in the mean seawater temperature (Kasting et al., 2006; van der Boorn et al., 2007; Shields and Kasting, 2007). Observed isotopic data might reflect more widespread hydrothermal activity on the ancient seafloor which slowly tapered off through time (Hofmann, 2005; van der Boorn et al., 2007). Enhanced continental weathering from higher paleoatmospheric CO$_2$ and a greater abundance of brittle volcanic rocks may have acted to suppress $\delta^{18}$O during the Precambrian (Jaffres et al., 2007). Oceanic temperatures are also expected to vary strongly with latitude and depth. The paleolatitudes and depths of formation of the rocks on which the temperature estimates discussed above are based are unknown (Feulner, 2012). Some recent studies have improved upon traditional methods by conducting combined analysis of oxygen and hydrogen isotopes and by analyzing the oxygen isotopic composition of phosphates found in Archean sediments. These studies suggest that seawater temperatures during the mid-Archean were more
moderate, likely between 299 and 308 K and no higher 313 K (Hren et al., 2009; Blake et al., 2010).

In this study our warmest Archean simulation, having 0.2 bar of CO$_2$ and 0.001 bar of CH$_4$ at 80% of the modern solar constant, global and annually averaged seawater temperatures reach 305 K while local seasonal maximum temperatures reach 321 K (Figure 3.9). Coincidentally this is also our only simulation that exhibits year around ice-free conditions. While the seawater temperature predictions of Robert and Chaussidon (2006) are not reached in this study, the predictions of Blake et al. (2010) are achieved with reasonable greenhouse gas amounts. With 0.015 bar of CO$_2$ and 0.001 bar of CH$_4$ seasonal and local maximum seawater temperatures hover just above 305 K. With the inclusion of $10^{-3}$ bar of CH$_4$ our modeled seawater temperatures are in good agreement with recent estimates for paleo-ocean temperatures while remaining safely within constraints imposed on paleoatmospheric CO$_2$.

A combination of high CO$_2$ and/or high CH$_4$ could in theory yield a hot climate for the early Earth. However, large amounts of CO$_2$ (>0.2 bar) would be required to match the hot temperature estimates predicted by Knauth and Lowe (2003) and Robert and Chaussidon (2006) if CO$_2$ and H$_2$O were the only greenhouse gases, hopelessly violating inferred constraints on CO$_2$. Photochemical calculations of Zerkle et al. (2012) predict an upper limit on atmospheric methane obeying the ratio CH$_4$/CO$_2 = 0.2$. Thus if the predicted atmospheric CO$_2$ at 2.69 Ga is 0.015 bar, CH$_4$ is limited to only 0.003 bar, 3 times what is presented in this study. However, such an increase in CH$_4$ would yield only a few additional watts per meter squared
Figure 3.9. Late Archean global mean and seasonal maximum seawater temperatures versus partial pressure of CO₂. Seasonal maximum and global mean mixed layer ocean temperature versus $p\text{CO}_2$ with 0.001 bar of CH₄ included. High seawater temperature estimates of Robert and Chaussidon (2006) are not possible within estimates of paleoatmospheric CO₂. However, our results can be easily reconciled with the more recent seawater temperature estimates of Blake et al. (2010).
of radiative forcing (Figure 2.2d), and that is before the radiative effects of organic hazes are even considered. A hot, dense, carbon-rich atmosphere would have left the scars of intense chemical weathering, however the geologic record suggests that weathering rates were modest during the Archean (Condie et al., 2001; Sleep and Hessler, 2006). Additionally, as one moves backward through geologic time, the Sun becomes fainter and thus the climate would require increasingly large amounts of greenhouse gases to offset. For reference, at the onset of the Archean circa 3.8 Ga the Sun was only 75% as bright as today which translates into an additional radiative forcing of \(-14.3 \text{ W m}^{-2}\) compared to simulations presented in this chapter.

An alternative to the hot ocean viewpoint is that the Archean had a more temperate climate. A temperate Archean is more easily reconcilable with evidence for glaciations observed around 2.3 Ga (Evans et al., 1997) and 2.9 Ga (Young et al., 1998) as opposed to an excessively hot planet. A transition from a hot, ice-free state to a hard snowball state requires large radiative forcings and exhibits a strong hysteresis (Pierrehumbert, 2004). By contrast, a temperate Archean may have easily swayed into and out of more moderate glacial states, not unlike the climate of more recent epochs. Here we find the Archean climate to have a greater sensitivity than present day, thus a relatively small push on the radiative balance can transition a temperate late Archean climate from one where ice is confined to polar regions to one where ice extends into mid-latitudes. The interaction of methane, biology and photochemical hazes may be responsible for Archean climate changes. Thickening photochemical hazes from increased methanogenic respiration may have been responsible for the lesser glaciation believed to have occurred at 2.9 Ga (Domagal-
Goldman et al., 2012). Assuming fractal shaped haze particles, Zerkle et al. (2012) predict haze effective optical depths at 0.550 μm reach 0.3 when methane reaches CH$_4$:CO$_2$ = 0.2. Such a haze would extinguish more than a quarter of the incoming solar energy. The glaciation at the end of the Archean circa 2.4 Ga is widely speculated to have been caused by the rise of oxygen and the subsequent destruction of a methane greenhouse (Kasting, 2005). If the pre-glacial atmosphere contained 0.015 bar of CO$_2$, the loss of 0.001 bar of methane due to rising oxygen levels would yield a ~15 K global mean temperature decline and a ~23° latitudinal expansion of sea ice. Even larger temperature changes would have occurred if higher CH$_4$ concentrations had been present prior to the rise of oxygen.

### 3.6. Summary

In this chapter we have used a general circulation model to study the climate of the late Archean circa 2.8 Ga when the Sun was 20% dimmer than present day. We vary only the solar constant and specified mixing ratios for CO$_2$ and CH$_4$. We find self-consistent climate solutions to a weaker form of the faint young Sun paradox, requiring that only some portion of the planet surface remain free from ice and thus hospitable for early life. While self-consistent hot solutions for the late Archean remain elusive, we find that cooler climates that remain dominated by areas of liquid surface water are easily achieved within published paleoatmospheric constraints on CO$_2$. The combination of 0.015 bar of CO$_2$ and 0.001 bar of CH$_4$ produces surface temperatures very near present day while not violating paleosol constraints on CO$_2$ and avoiding complications from photochemical haze formation.
Climates with weaker greenhouses and mean surface temperatures as low as 260 K can maintain open ocean fractions of greater than 50%, thus ensuring the habitability of the Archean even at relatively low paleoatmospheric greenhouse gas levels. Convincing geological arguments can be made in favor of a more temperate late Archean climate as opposed to an excessively hot planet. One need not extrapolate based on sparse geologic data to there being only climates of extreme hot or cold during the Archean. The change in latitudinal energy distribution compared with present day due to weak solar irradiance and a strong greenhouse effect may have caused the ancient climate to be susceptible to wide swings for relatively small radiative forcings. However, here climates with open-ocean fractions of greater than 50% are maintained despite a 20% reduction in solar constant even with only 0.005 bar of CO₂ and no CH₄. Resolving the faint young Sun paradox may not be as challenging to climate modelers as previously thought.
Chapter 4

Controls on the Archean climate

4.1. Overview

The most obvious means of resolving the faint young Sun paradox is to invoke large quantities of greenhouse gases, namely CO\textsubscript{2} and CH\textsubscript{4}. However, numerous changes to the Archean climate system have been suggested that may yield additional warming, thus easing the required greenhouse gas burden. Here we use a three-dimensional climate model to examine some of the factors that control Archean climate. We examine changes to the Earth’s rotation rate, surface albedo, cloud properties, and total atmospheric pressure following proposals from the recent literature. While the effects of increased planetary rotation rate on surface temperature are insignificant, plausible changes to the surface albedo, cloud droplet number concentrations, and atmospheric nitrogen inventory may each impart global mean warming of 3 to 7 K. While none of these changes presents a singular solution to the faint young Sun paradox, a combination can have a large impact on climate. Global mean surface temperatures at or above 288 K can easily be maintained throughout the entirety of the Archean if plausible changes to clouds, surface albedo and nitrogen content are considered.

As discussed in chapter 3, simulations using general circulation models show that only modest amounts of CO\textsubscript{2} may be needed to marginally rectify the faint
young Sun paradox towards the end of the Archean. Wolf and Toon (2013) found that 0.015 bar of CO₂, the “best guess” estimate for atmospheric CO₂ during the late Archean provided by Driese et al. (2011), can prevent the Earth from freezing over during the late Archean (circa 2.8 Ga with 80% of the present day solar constant). However global mean surface temperatures remain cold ($T_s = 270$ K) and the sea ice margin extends into mid-latitudes ($46^\circ$). This cool climate may be inconsistent with the lack of evidence for glaciation through out most of the Archean (Feulner, 2012). With the addition of $10^{-3}$ bar of methane, surface temperatures and sea ice extent become nearly equal to those of the present day Earth ($T_s = 285$ K, sea ice margin at $68^\circ$ latitude) (Wolf and Toon, 2013). Note also that the CO₂ partial pressure appropriate for the late Archean can be stretched higher than discussed here as Sheldon (2006) suggests $\sim 0.025$ bar as an upper limit. Thus additional warming may be possible via conventional means (i.e. greenhouse gases). CH₄ could feasibly have been higher than $10^{-3}$ bar as well. So long as the ratio of CH₄ to CO₂ remains less than about 0.1, photochemical hazes should not form (Haqq-Misra et al., 2008; DeWitt et al., 2009; Zerkle et al., 2012). While photochemical hazes may provide an effective ultraviolet shield for early Earth (Wolf and Toon, 2010) thick hazes may have a strong cooling effect (Haqq-Misra et al., 2008). Studies using climate models of differing origin find similar greenhouse gas to surface temperature relationships for the Archean (Charnay et al., 2013; Kunze et al., 2013).

However, outstanding questions remain. While solutions to the faint young Sun paradox with present day temperatures or cooler may provide ample planetary surface area with moderate temperatures and open-oceans, they fall short of the hot
seawater temperatures indicated by some geochemical analyses of ancient marine sediments (Knauth and Lowe, 2003; Robert and Chaussidon, 2006). Granted, the hot seawater interpretation for the Archean climate is controversial (Shields and Kasting, 2007; Jaffrés et al., 2007) and more recent estimates indicate seawater temperatures only slightly warmer than those in the present day tropics (Hren et al., 2009; Blake et al., 2010). The faint young Sun paradox also becomes more severe as one probes deeper backwards in time. At the onset of the Archean (circa 3.8 Ga) the Sun was only 75% as bright as today (Gough, 1981), thus more substantial warming is required to solve the paradox compared with the results presented by Haqq-Misra et al. (2008) and Wolf and Toon (2013), both of which consider the Sun at 80% of its present day brightness. Of course, our confidence in geochemical indicators for both surface temperature and CO₂ become correspondingly weaker as one probes deeper back in time. Finally, evidence from banded-iron formations suggest that atmospheric CO₂ during the Archean may be constrained to a mere 3 times the present atmospheric level (PAL) (Rosing et al., 2010). This is considerably lower than CO₂ estimates derived from paleosols (Rye et al., 1995; Hessler et al., 2004; Sheldon, 2006; Driese et al., 2011). However, again this interpretation of the banded iron formations remains controversial (Dauphas and Kasting, 2011; Reinhard and Planavsky, 2011). If we are to consider either the requirement for hot surface temperatures, the climate of earliest Archean, or the strict CO₂ constraint imposed by banded iron formations, other mechanisms for warming the climate may still be needed to resolve the faint young Sun paradox.
The simulations presented in chapter 3 (see also Wolf and Toon, 2013) assume that aside from the strength of the solar constant and atmospheric inventories of CO₂ and CH₄, the Archean Earth remained fundamentally similar to the present day. Model physics parameterizations are left in their default state and the model reacts to changes in the shortwave and longwave radiative forcings only. However other researchers have proposed systematic differences between the Archean and the present day that may have controlled early climate. The Earth had a faster rotation rate in the distant past (Williams, 2000). This would have modulated atmospheric dynamics (Jenkins, 1996). Rosing et al. (2010) suggest that the early Earth may have had fewer cloud condensation nuclei and significantly less emergent land; both would lower the planetary albedo. Goldblatt et al. (2009) suggests that the early Earth may have had an increased inventory of atmospheric nitrogen causing the sea level pressure to be greater than 1 bar. Increased pressures would broaden spectral lines and increase absorption from existing greenhouse species. In this chapter we test these systematic changes to the Archean climate system using a general circulation model. We first conduct a series of sensitivity tests and we then combine warming mechanisms to find optimal warming scenarios for the duration of the Archean.

Again we use a general circulation model based on the Community Atmospheric Model version 3.0 (CAM3) from the National Center of Atmospheric Research (Collins et al., 2004; Collins et al. 2006). The model configuration is discussed in depth in chapter 2. Physics packages remain identical to their default form except where described in the following subsections.
Sensitivity tests are conducted to isolate the warming gained from a variety of atmospheric conditions specific to the Archean and notably different from the present day. For a baseline case we first simulate the late Archean time period with a solar constant of 1093.6 W m\(^{-2}\) (2.8 Ga, 80% of present day) warmed only by CO\(_2\) and H\(_2\)O. Our baseline atmosphere consists of 0.06 bar of CO\(_2\), no methane, 0.00933 bar of argon gas, 0.9438 bar of N\(_2\) and variable amounts of water vapor determined through self-consistent modeling of the hydrological cycle. Note that increasing N\(_2\) above the present atmospheric level (0.79115 bar N\(_2\)) ensures that the mean sea level pressure is 1.013 bar, identical to the present day. The planetary rotation rate, cloud droplet number concentrations, cloud droplet sizes, and land properties are held identical to present day. In this baseline simulation the global mean surface temperature is 287.9 K, matching that of the present day (Table 3.1; see also section 3.2 for an in depth discussion of our baseline case). In the following sections we consider plausible changes to the rotation rate, land surface albedo, cloud droplet properties, and total atmospheric pressure relative to our baseline simulation.

It should be noted that climate sensitivity is state-dependent (Hansen et al., 2005; Caballero and Huber, 2013). Initial conditions affect the climate response to a given forcing. In particular, the surface temperature controls many factors of climate including the distribution of snow cover, sea ice, water vapor, clouds and ultimately the albedo and infrared opacity (via water vapor) of the planet. Our goal in this section is to test the sensitivity of climate to warming mechanisms starting from approximately present day temperatures. Climate sensitivities will likely deviate as one approaches tipping points such as a runaway greenhouse or a
snowball Earth. However, a climate with surface temperatures near that of the present day may be centered between extremes of climate and upturns in climate sensitivity (Hansen et al., 2005). Archean climates, under a weak Sun and warmed by thick greenhouses, tend to have slightly larger climate sensitivities than that of the present day earth over a wide range of conditions (Wolf and Toon, 2013). Climate models of differing origin and construction may yield varying climate responses (Bony et al., 2006).

4.2. Increased rotation rate

The Earth–Moon orbital system can be treated as a closed system subject only to the force of gravity. Differential gravitational forces exerted by the Moon on the Earth causes the oceans to bulge outwards creating oceanic tides. However, the Earth’s rotation drags the tidal bulge, placing it slightly ahead of the Earth–Moon axis. As the Moon pulls back on the slightly off-axis tidal bulge, it exerts a subtle torque on the Earth counter to the direction of rotation. Since angular momentum of the Earth-Moon system is conserved, this process causes the Moon to gain energy at the expense of the Earth. The Earth’s rotation rate is slowed while the Moon moves further from the Earth. This process has been ongoing since the formation of the Earth–Moon system. Extrapolating backwards in time, the Earth must have had a faster rotation rate in the distant past. Analysis of sedimentary deposits within ancient tidal zones and analysis of the fossils of ancient marine invertebrates both indicate that the length of day was likely near 18 hours during the late Archean
(Williams, 2000; Denis et al., 2002). However other studies indicate that the length of day may have been as short as 14 hours circa 4.0 Ga (Zahnle and Walker, 1987).

Jenkins (2000, 2001) found that the obliquity of the early Earth may have played a strong role in controlling climate. High obliquity (70°) could produce relatively warm Archean climates with only a few thousand parts per million of CO₂ by changing the latitudinal distribution of solar insolation. However, it is unclear how the Earth may have migrated to such an extreme obliquity and then back to its present 23.4° obliquity. For all simulations in this work we assume the present day obliquity.

We conduct sensitivity tests of the Archean climate with day lengths of 15, 18, and 24 hours. As expected from dynamical theory, a faster rotation rate causes the Hadley cells to become weaker and smaller in latitudinal extent (Figure 4.1). Zonal, meridional, and vertical motions all decrease in magnitude. It is well established that increasing rotation rates results in a decrease in poleward heat transport, allowing the tropics to stay warmer while cooling the poles (Hunt, 1979; Navarra and Boccaletti, 2002; Jenkins, 1996). As a result the meridonal temperature gradient steepens, but the effect is slight. For an 18 hour day, zonal mean surface temperatures increase by ~1 K in the tropics while decreasing by ~3 K at the poles compared with a 24 hour day. For a 15 hour day, zonal mean surface temperatures increase by ~2 K in the tropics while decreasing by ~5 K at the poles (Figure 4.2). Dynamical effects impart subtle changes to the vertical and latitudinal structure of clouds, however the effect on overall climate appears unimportant. Local differences in net cloud forcings can vary by ±5 W m⁻² from our baseline case,
Figure 4.1. Zonal mean streamfunction for changing rotation rates. Zonal and annual mean streamfunctions for 24 hour (a), 18 hour (b), and 15 hour (c) days respectively. Faster rotation rates reduce wind speeds and the overall strength of circulation as is reflected in the streamfunction. Hadley and Ferrel cells weaken and latitudinally contract.
Figure 4.2. Zonal mean surface temperature for changing rotation rates. Zonal and annual mean surface temperature (a) and the change in surface temperature (b) due to increased rotation rates. Increasing the rotation rate reduces meridional heat transports, thus steepening the meridional temperature gradient. However, global mean temperature differences are small (<1 K).
<table>
<thead>
<tr>
<th>Length of Day (hours)</th>
<th>24</th>
<th>18</th>
<th>15</th>
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</thead>
<tbody>
<tr>
<td>Mean surface temperature (K)</td>
<td>287.9</td>
<td>288.1</td>
<td>288.6</td>
</tr>
<tr>
<td>Sea ice margin (degrees)</td>
<td>74.2</td>
<td>73.5</td>
<td>73.2</td>
</tr>
<tr>
<td>Cloud radiative forcing (W m⁻²)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shortwave</td>
<td>−34.4</td>
<td>−34.6</td>
<td>−34.6</td>
</tr>
<tr>
<td>Longwave</td>
<td>25.3</td>
<td>25.5</td>
<td>25.8</td>
</tr>
<tr>
<td>Net</td>
<td>−9.1</td>
<td>−9.1</td>
<td>−8.8</td>
</tr>
</tbody>
</table>

**Table 4.1. Rotation rate sensitivity tests.**
but global mean net cloud forcings vary by less than 0.3 W m$^{-2}$ over the three rotation rate cases (Table 4.1). In total, with an 18 hour day the global mean surface temperature rises by only a scant 0.2 K from our baseline simulation to reach 288.1 K. With a 15 hour day the global mean surface temperatures rises 0.7 K to 288.6 K. Our findings largely confirm Jenkins (1996) work using an earlier three-dimensional climate model. While identifiable changes to the climate system are found due to increased rotation rates, the overall warming gained is miniscule in the context of that needed to resolve the faint young Sun paradox.

Kienert et al. (2012) using a three-dimensional ocean model coupled to a parameterized statistical atmosphere model found that increasing rotation rates lead to strong cooling of the Archean, requiring 0.4 bar of CO$_2$ bar or more to overcome the faint young Sun. However, recent work with various circulation models agree well with our results presented here (Charnay et al., 2013; Le Hir et al., 2013). Increased rotation rates do not appear to play a significant role in warming the Archean.

4.3. Darker surface albedo

Surface albedo characteristics during the Archean were much different from today. Reductions to the planetary surface albedo have been proposed as at least a partial solution to the faint young Sun paradox (Rosing et al., 2010). There was much less emergent continental crust during the Archean than there is today, instead the Earth would have been dominated by ocean (Dhime et al., 2012). What continents did exist were barren of vegetation. The earliest Archean continents
were likely formed from dark basalts. As the continents aged the surface texture may have changed into lighter colored soils typical of modern day desert regions (Rosing et al., 2010). Presently, permanent land glaciers on Greenland, Antarctica, and in mountainous regions add considerably to the surface albedo. Such semi-permanent land based ice structures may have been absent from the early Earth.

Here we remove all surface vegetation and all continental ice sheets and test “light” and “dark” bare soil surface configurations specified onto the present day continental areas. Note that sea ice and snow cover are still permitted, both of which strongly modulate the surface albedo. For simplicity we do not move or eliminate continental area and instead focus only on changes to land albedo. The albedo of bare soil depends upon moisture content and crudely upon wavelength (visible or near-infrared, demarcated at 0.7 μm). For light colored soils, the water-saturated soil albedo in the visible (near-infrared) is 0.12 (0.24) while the dry albedo is 0.24 (0.48). For dark colored soils, the water-saturated soil albedo in the visible (near-infrared) is 0.05 (0.10) while the dry albedo is 0.10 (0.20). The albedo of ocean is 0.06 across both the visible and near-infrared, thus the albedo of saturated dark soil approaches that of ocean.

Changing the albedo of the land has a limited effect on overall climate as continental area constitutes only ~30% of the total planet surface. For our light (dark) simulation the global mean surface albedo is 0.143 (0.105), while for our baseline simulation it is 0.136 (Table 4.2). Note that the baseline model has a slightly lower albedo than light soil. Modest gains in global mean surface temperature are found for both light and dark soil simulations compared with our
baseline case. The dark soil global mean surface temperature rises 3.7 K to 291.6 K while exhibiting a 23% percent decrease in the global mean surface albedo compared with the baseline simulation. The light soil global mean surface temperature rises 0.9 K to 288.8 K despite having a slightly larger global mean surface albedo than the baseline case. This can be attributed to two factors; reduced low cloud fractions ($p > 700$ mb) over land and significantly decreased surface albedos over the Antarctic continent and Greenland.

Replacing all forested and vegetated land areas with bare soil, results in a lower latent heat flux to the atmosphere because precipitation is more readily lost to runoff. In particular, tropical forested areas tend to hold water in the near surface environment, allowing water to be cycled more easily back into the atmosphere creating clouds in the lowest model layers. Thus by removing all surface vegetation (thus creating a desert land type), we find a reduction in low clouds and a modest reduction to the magnitude of shortwave cloud forcings (Table 4.2). This effect becomes less pronounced for the dark soil simulation because increasing global air temperatures enhance ocean surface evaporation rates, allowing more moisture into the atmosphere.

Reduced surface albedos in the polar regions exist due to the removal of semi-permanent ice-sheets. While large surface albedos are still found over the poles due to the accumulation of snow and the growth of sea-ice, they remain slightly less than those found in our baseline simulation (Figure 4.3b). Periodic melts of snowpack over Antarctica and Greenland reveal a low albedo soil surface instead of glacial ice (which has been removed from the model in these simulations).
Figure 4.3. Zonal mean surface temperature for changing land albedos. Zonal and annual mean surface temperature (a) and surface albedo (b) for default, light, and dark land surface configurations. The mean surface temperature is affected both by changing the specified surface albedo and through a modest positive cloud forcing that occurs when removing forested land types in favor of bare soil (i.e. desert).
<table>
<thead>
<tr>
<th>Soil Surface Configuration</th>
<th>Present Day</th>
<th>Light</th>
<th>Dark</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean surface temperature (K)</td>
<td>287.9</td>
<td>288.8</td>
<td>291.6</td>
</tr>
<tr>
<td>Sea ice margin (degrees)</td>
<td>74.2</td>
<td>75.6</td>
<td>77.9</td>
</tr>
<tr>
<td>Surface albedo</td>
<td>0.136</td>
<td>0.143</td>
<td>0.105</td>
</tr>
<tr>
<td>Cloud fraction</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Low</td>
<td>0.309</td>
<td>0.293</td>
<td>0.296</td>
</tr>
<tr>
<td>Middle</td>
<td>0.233</td>
<td>0.214</td>
<td>0.232</td>
</tr>
<tr>
<td>High</td>
<td>0.540</td>
<td>0.548</td>
<td>0.541</td>
</tr>
<tr>
<td>Cloud radiative forcing (W m⁻²)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shortwave</td>
<td>-34.4</td>
<td>-31.9</td>
<td>-33.9</td>
</tr>
<tr>
<td>Longwave</td>
<td>25.4</td>
<td>25.6</td>
<td>25.9</td>
</tr>
<tr>
<td>Net</td>
<td>-9.0</td>
<td>-6.3</td>
<td>-8.0</td>
</tr>
</tbody>
</table>

**Table 4.2. Surface albedo sensitivity tests.**
Overall warmer climates found in both light and dark soil simulations ensure that sea ice distributions are reduced, further lowering polar surface albedos. Changes to the surface albedo and surface temperature are more pronounced in the northern hemisphere since this is where the majority of the continental area resides in our model (Figure 4.3a).

Our results are in general agreement with recent analysis of the effect of land albedo on Archean climate (Goldblatt and Zahnle, 2011; Charnay et al. 2013; Kunze et al., 2013). Reducing the surface albedo results in a non-negligible gain in global mean temperature and may constitute a minor contributor towards warming the early Earth. However a solution to the faint young Sun paradox cannot rely on warming from a reduced surface albedo alone.

### 4.4. Changes to clouds

Alternative solutions to the faint young Sun paradox have been proposed based on fundamental changes to cloud properties on the early Earth. Notably the early Earth likely had fewer cloud condensation nuclei (CCN) (Andreae, 2007; Rosing et al., 2010). CCN are important because at typical tropospheric temperatures water vapor requires the presence of aerosols to seed condensation and form cloud droplets. Where CCN are plentiful, cloud droplets tend to be more numerous but smaller in size, creating clouds that are more reflective and have longer lifetimes against removal (via precipitation). Where CCN are few, cloud droplets are sparse but grow larger in size, creating clouds that are less reflective
and rain out more quickly (Kump and Pollard, 2008). Thus, a reduction to cloud albedos driven by reductions to CCN concentrations may provide a substantial warming mechanism for the early Earth (Rosing et al., 2010). At present CCN are dominantly sourced by sea salt, dust, industrial pollution, biomass burning, and the oxidation of biologically produced compounds. The present day CCN burden may be \(~100 \text{ cm}^{-3}\) over biologically productive regions of the ocean, while possibly reaching \(1000 \text{ cm}^{-3}\) or more over active continental areas (Andreae, 2007). While the Archean obviously lacked anthropogenic pollutants, it was also anoxic and had much sparser biological activity than today. Archean life was primarily microbial and had not yet colonized the land (Schopf, 2006). Thus, one might expect that sea salt and dust were the only available CCN for the early Earth.

Sea salt aerosols are produced as winds loft particles off whitecaps in turbulent oceans. Detailed three-dimensional modeling of sea salt CCN concentrations, supported by observations, predict values between 10 and \(50 \text{ cm}^{-3}\) over most of the world’s oceans. In isolated regions of persistent high wind speeds (such as the “roaring 40s” on the present Earth) sea salt CCN concentrations may approach \(100 \text{ cm}^{-3}\) (Fan and Toon, 2011). The early Earth would have had less continental area, so there may have been more surface area uninterrupted by land, promoting stronger zonal mean winds. However this effect would have been offset by weakened overall circulation due to increased rotation rates. Presently, CCN found over biologically and photochemically inactive ocean regions are primarily sea salts, numbering only \(~10 \text{ cm}^{-3}\) (Andreae, 2007). Similar conditions would likely have existed during the Archean.
Dust serves as CCN predominantly over desert regions, however it is likely that all emergent continental crust was desertified and thus dusty. At present, over inland deserts dust may constitute up to 40% of the total CCN concentration and create 23.8% of the cloud droplet number concentration (CDNC) (Karydis et al., 2011). This equates to local maximum values of CDNC sourced by dust to between 40 and 100 cm$^{-3}$, however regional and global average values are much lower (Karydis et al., 2011). Pre-industrial CCN concentrations over the continents are believed to have been between 50 and 200 cm$^{-3}$ (Andreae, 2007), however this estimate includes the contribution of biologically sourced CCN which would have been absent over Archean continents. Archean CCN concentrations over land were probably much lower than pre-industrial estimates.

CAM3 utilizes bulk parameterizations that determine the exchange between water vapor, cloud condensate and precipitate (Rasch and Kristjánsson, 1998; Zhang et al., 2003). Cloud condensate amounts are controlled by dynamical and thermodynamical processes, however liquid CDNC and liquid cloud droplet effective radii are specified parameters rather then following from a self-consistent calculation based on the activation of CCN. CCN and CDNC are both closely related to the atmospheric aerosol burden with CDNC by definition numbering less than the CCN burden (Karydis et al., 2011; Hegg et al., 2012). Note that the model treats ice and liquid cloud types with fundamentally different parameterizations. Ice cloud properties are determined by temperature and condensate mixing ratio alone and do not rely on fixed parameters for CDNC and effective radius (Kristjánsson and Kristiansen, 2000).
In default setting, appropriate for the present day Earth, liquid CDNC are taken to be 400 cm$^{-3}$ over land, 150 cm$^{-3}$ over ocean and 75 cm$^{-3}$ over ice. We preform simulations with 50%, 20%, and 10% of default present day CDNC, scaled evenly across land, ocean, and sea ice values (Table 3). For a given amount of cloud condensate, the cloud droplet effective radius varies as CDNC$^{-1/3}$. In the default setting, liquid cloud droplets achieve their maximum size of 14 µm over ocean and sea ice, while for our test simulations with reduced CDNC cloud droplet maximum sizes found over ocean can be 17.6, 23.9, and 30.2 µm respectively. Liquid cloud droplet sizes are slightly smaller over land owing to a larger aerosol burden (and thus more numerous CCN and CDNC). Rosing et al. (2010) suggest cloud droplet sizes of 20 and 30 µm may be appropriate for the Archean. For reference, presently liquid cloud droplet sizes are observed to reach 17 µm over unproductive oceans far from human activities (Bréon et al., 2002). Both liquid and ice cloud optical properties are determined by mie scattering coefficients calculated at the cloud droplet effective radii.

Increasing liquid cloud droplet sizes has a two-fold effect on climate. Clouds that consist of larger but less numerous droplets are inherently less reflective. Clouds droplet extinction efficiencies are ~2 across visible and infrared wavelengths for all effective radii of interest here. Thus for a given cloud water path, the cloud optical depth is inversely proportional to effective radius. Secondly, larger cloud droplets will more easily precipitate out of the atmosphere, thus total cloud water paths will be reduced. CAM3 parameterizes the conversion of liquid cloud condensate to rain following the method suggested by Chen and Cotton (1987).
The rate of conversion of liquid cloud condensate to precipitate is proportional to the cloud droplet effective radius. However, other parameterizations use precipitate conversion rates as large as \( r^{5.37} \) (as opposed to \( R^1 \) in this study), resulting in much more efficient rainout, fewer clouds, and more surface warming than is found here (Kump and Pollard, 2008; Charnary et al., 2013).

Figure 4.4 shows the zonal mean cloud condensate amount (g/kg air) including both ice and liquid for all four cloud simulations (default, 50%, 20%, 10%). Note that liquid cloud condensate dominates over ice condensate by a ratio of 5:1. For each successive reduction in CDNC, cloud condensate amounts fall. As cloud decks become thinner the climate warms as expected. As CDNC are reduced, low (>700 mb) and mid (700 – 400 mb) level cloud fractions are reduced at middle and high latitudes but remain roughly equal over the tropics (Figure 4.4, Figure 4.5b,c). Note that the Archean has inherently fewer convective clouds due to weak solar insolation (Wolf and Toon, 2013). The magnitude of shortwave cloud forcings is strongly reduced at mid-latitudes, by up to \( \sim 33 \) W m\(^{-2} \) in the southern hemisphere and \( \sim 20 \) W m\(^{-2} \) in the northern hemisphere for the 10% CDNC simulation compared with the default (Figure 4.5e). Changes to cloud forcings are less over the tropics (\( \sim 7 \) W m\(^{-2} \) reduction in magnitude). On global mean, shortwave cloud forcings are reduced in magnitude by 12.9 W m\(^{-2} \) for the 10% CDNC simulation compared with the default (Table 4.3). High altitude (<400 mb) cloud fractions are reduced over the tropics, however they are increased over high latitudes (Figure 4.5a). This behavior is a feature of warming climates and is associated with polar amplification. It is not caused by our specified changes to
Figure 4.4. Zonal mean cloud condensate for changing cloud droplet number concentrations. Zonal and annual mean cloud condensate amounts from simulations with the default cloud number droplet concentration (CDNC) (a), 50% (b), 20% (c), and 10% (d) of the default values (see table 3). Cloud condensate drops significantly as CDNC is reduced because cloud droplets are larger and thus rainout more efficiently.
Cloud fraction

Figure 4.5. Zonal mean cloud fractions and forcings for changing cloud droplet number concentrations. Zonal and annual mean cloud fractions and cloud forcings for simulations with varying cloud droplet number concentration (CDNC). Low cloud fractions ($p > 700$ mb) (c), and middle cloud fractions ($700 > p > 400$ mb) (b) both see significant reductions over mid-latitudes as CDNC is decreased. High cloud fractions ($p < 400$ mb) (a) decrease over the tropics and increase over the poles. However this feature is caused by overall warming to the planet along with polar amplification and is not affected by our choice of CDNC. Longwave cloud forcings (d) do not change significantly, however the magnitude of shortwave cloud forcings (e) are greatly reduced, particularly over the mid-latitudes. As CDNC is decreased, the net cloud forcing (f) grow less negative, thus the cooling effect of clouds is greatly weakened.
<table>
<thead>
<tr>
<th>CDNC Configuration</th>
<th>100% (Default)</th>
<th>50%</th>
<th>20%</th>
<th>10%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean surface temperature (K)</td>
<td>287.9</td>
<td>291.2</td>
<td>294.6</td>
<td>296.6</td>
</tr>
<tr>
<td>Sea ice margin (degrees)</td>
<td>74.2</td>
<td>78.2</td>
<td>80.6</td>
<td>82.0</td>
</tr>
<tr>
<td>CDNC (# cm⁻³)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Land</td>
<td>400</td>
<td>200</td>
<td>80</td>
<td>40</td>
</tr>
<tr>
<td>Ocean</td>
<td>150</td>
<td>75</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td>Sea ice</td>
<td>75</td>
<td>37.5</td>
<td>15</td>
<td>7.5</td>
</tr>
<tr>
<td>Cloud droplet radii (µm)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Land</td>
<td>8</td>
<td>10.1</td>
<td>13.7</td>
<td>17.2</td>
</tr>
<tr>
<td>Ocean</td>
<td>14</td>
<td>17.6</td>
<td>23.9</td>
<td>30.2</td>
</tr>
<tr>
<td>Sea ice</td>
<td>14</td>
<td>17.6</td>
<td>23.9</td>
<td>30.2</td>
</tr>
<tr>
<td>Cloud albedo</td>
<td>0.134</td>
<td>0.117</td>
<td>0.095</td>
<td>0.082</td>
</tr>
<tr>
<td>Water column (Kg m⁻²)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water vapor</td>
<td>20.2</td>
<td>24.4</td>
<td>30.5</td>
<td>34.6</td>
</tr>
<tr>
<td>Cloud liquid water</td>
<td>0.086</td>
<td>0.077</td>
<td>0.065</td>
<td>0.059</td>
</tr>
<tr>
<td>Cloud ice water</td>
<td>0.017</td>
<td>0.015</td>
<td>0.013</td>
<td>0.012</td>
</tr>
<tr>
<td>Cloud radiative forcing (W m⁻²)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shortwave</td>
<td>−34.4</td>
<td>−30.2</td>
<td>−24.8</td>
<td>−21.5</td>
</tr>
<tr>
<td>Longwave</td>
<td>25.4</td>
<td>25.8</td>
<td>25.4</td>
<td>25.4</td>
</tr>
<tr>
<td>Net</td>
<td>−9.0</td>
<td>−4.4</td>
<td>0.6</td>
<td>3.9</td>
</tr>
</tbody>
</table>

Table 4.3. Cloud droplet number concentration sensitivity tests.
liquid CDNC and liquid cloud droplet sizes. Similar behavior is observed in simulations that rely only on greenhouse gas warming with no alterations made to the default cloud parameterizations (not shown). There is little change to the longwave cloud forcings (Figure 4.5d). Longwave cloud forcings are imparted most strongly by high-altitude ice clouds. In this set of simulations ice cloud particles have not been altered, thus we would not expect to find significant changes here.

Simulations of the present day Earth using CAM3 indicate that clouds have a net radiative forcing of $-24.9 \text{ W m}^{-2}$ and thus impart a strong cooling influence on climate (see also Collins et al., 2006). For our baseline simulation of the late Archean using default CDNC and cloud droplet sizes, clouds have a net radiative forcing of only $-9.0 \text{ W m}^{-2}$ (see discussion in section 3.2). For simulations with 20% and 10% CDNC, global mean net cloud forcings become positive with values of +0.6 and +3.9 W m$^{-2}$ respectively (Table 4.3). With 10% CDNC, cloud net radiative forcings are positive virtually everywhere on the planet (Figure 4.5f). Reducing CDNC to 10% has the largest effect on climate, increasing global mean surface temperatures by 8.7 K to 296.6 K. Thus while perhaps not a full solution to the faint young Sun paradox in its own right, reduced cloud condensation nuclei (and thus reduced CDNC) during the Archean may provide a fairly sizeable boost to surface temperatures. Even given our maximum change in net cloud forcing found to be +12.9 W m$^{-2}$ for our 10% CDNC case, our results fall within the ‘plausible’ scenarios outlined by Goldblatt and Zahnle (2011).

It has also been proposed that feedbacks involving increased high altitude (ice) clouds may help warm the early Earth (Rossow et al., 1982; Rondanelli and
Lindzen, 2010). Due to their low radiating temperatures, high altitude clouds can contribute strongly to greenhouse effect. Very cold temperatures are found aloft in our Archean simulations, with minimum temperatures near 150 K, due to the absence of an ozone layer and increased radiative cooling from high-CO₂ concentrations (Wolf and Toon, 2013). The model diagnoses large cloud fractions located near the deep atmospheric temperature minimum found at 1 mb (~30 km) (Figure 4.6). Exceedingly low ambient temperatures cause very low saturation vapor pressures and thus high relative humidities are easily achieved and large cloud fractions are found. While cloud ice water mixing ratios found here are much larger for Archean simulations compared with the present day (~10⁻⁸ versus ~10⁻¹⁸, really there are no clouds in most of the present day stratosphere, only over Antarctica in the polar night), they remain two orders of magnitude lower than typical cloud ice water mixing ratios found in the upper troposphere (~10⁻⁶).

It is unclear how ice particles might be systematically different during the Archean compared with the present day. Ice particle growth is primarily controlled by temperature because the amount of available water vapor is limited by temperature. Ice particle sizes increasing monotonically with temperature (Kristjánsson and Kristiansen, 2000). In our baseline Archean simulation, ice particles in the upper ice cloud deck have sizes of ~0.5 μm near 1 mb. Here we have explored tuning the upper level ice cloud deck based on the work of Urata and Toon (2013) in studying warm early Martian climates. We conducted simulations where ice clouds above the tropopause are artificially forced to have larger particle sizes (10 μm, 50 μm, 100 μm) and 100% cloud fractions across the whole sky. However,
Figure 4.6. Zonal mean cloud fraction contour for Archean and present day atmospheres. Zonal and annual mean cloud fractions for present day simulations (a) and for Archean simulations (b). The removal of ozone allows the Archean stratosphere to become very cold. Large cloud fractions are diagnosed in the Archean stratosphere because the saturation vapor pressure is quite low. Only a small amount of water vapor is needed to saturate the Archean stratosphere and cause ice particles to condense.
these simulations fail to yield discernable warming. The upper cloud deck found in Archean simulations contains too little cloud water to have a significant impact on climate even when tuned to optimize the cloud greenhouse effect.

4.5. Increased atmospheric nitrogen

An accounting of the Earth’s nitrogen budget suggests that the total inventory of nitrogen existent on the Earth, including both in the atmosphere and in the solid Earth, may total $14.8\pm5.3 \times 10^{18}$ kg. This is equal to 3 PAL N$_2$ as a best guess and 5 PAL as an upper limit (Goldblatt et al., 2009). Low biological productivity during the Archean would have reduced the rate of nitrogen removal from the atmosphere via fixation and burial within sediments. Coupled with increased outgassing of nitrogen trapped in the mantle, the Archean bio-geological system could have favored higher atmospheric N$_2$ inventories (Goldblatt et al., 2009). Increasing the nitrogen content (and thus the total pressure) of the young atmosphere results in increased molecular absorption from existing greenhouse house species through the pressure broadening of spectral lines. Som et al. (2012) attempt to constrain the surface air pressure of the late Archean by analyzing the imprints left from ancient raindrops in the 2.7 Ga Ventersdorp Supergroup, South Africa. Their most probable estimate places surface air pressure between 0.527 and 1.115 bar, thus the atmospheric nitrogen content was not likely much different than today (in fact their estimate implies possibly reduced surface air pressures compared with the present day). However, their absolute upper limit on surface pressure is 2.128 bar, leaving open the possibility that the atmospheric nitrogen
content was indeed much larger than the present day. Marty et al. (2013) constrain $N_2$ in the early atmosphere through an analysis of the $N_2/^{36}Ar$ ratios in fluid inclusions trapped in Archean quartz. Marty et al. (2013) conclude that the total surface pressure at 3.5 Ga was no more than 1.1 bar and possibly as low as 0.5 bar. Nonetheless, given the overall low-confidence in ancient geochemical indicators, here we will explore the idea of increased atmospheric $N_2$ as originally proposed by Goldblatt et al. (2009).

Here we simulate atmospheres having 1, 1.5, 2, and 3 PAL $N_2$ in addition to our baseline simulation (Table 4.4). Note that 1 PAL of $N_2$ is equal to 0.79115 bar. Our baseline simulation achieves a mean sea level pressure of 1.013 bar (identical to the present day) by invoking a slightly larger atmospheric $N_2$ inventory (~1.19 PAL $N_2$) to make up for the removal of oxygen. While increasing $N_2$ in the atmosphere enhances the greenhouse forcing imparted by a static amount of the greenhouse gases, it also increases Rayleigh scattering (Figure 4.7). However, the positive longwave forcing from the pressure broadening effect outpaces the negative shortwave forcing from enhanced scattering to space. Increasing surface temperatures and increasing surface pressures have competing effects on the hydrological cycle. Warming surface temperatures increase convection and thus increase clouds, both tending to cool to the surface. However, increasing surface pressure increases the absolute value of the moist adiabatic lapse rate, thus decreasing the frequency of convective instabilities and thinning cloud cover, both tending to warm the surface (Goldblatt et al. 2009). These feedbacks partially offset. Here we find modest reductions to the magnitude of shortwave cloud forcings as the
Figure 4.7. Global mean radiative forcing versus partial pressure of N₂. Global mean instantaneous clear sky radiative forcing from increasing the nitrogen content of the atmosphere. Here the atmosphere is assumed to have a mean surface temperature of 287.9 K, 0.06 bar of CO₂, with neither CH₄, nor O₂, nor O₃. Atmospheric water vapor is determined by self-consistent hydrological cycle modeling. Increasing N₂ increases the longwave forcing by enhancing pressure broadening of existing spectral lines. However, increasing N₂ also leads to a negative shortwave forcing by enhanced Rayleigh scattering. In total, the positive longwave forcing wins out and climate warms.
<table>
<thead>
<tr>
<th>$p_{\text{N}_2}$ (PAL)</th>
<th>1</th>
<th>1.19</th>
<th>1.5</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sea level pressure (bar)</td>
<td>0.861</td>
<td>1.013</td>
<td>1.256</td>
<td>1.651</td>
<td>2.443</td>
</tr>
<tr>
<td>Mean surface temperature (K)</td>
<td>286.1</td>
<td>287.9</td>
<td>289.9</td>
<td>292.9</td>
<td>297.6</td>
</tr>
<tr>
<td>Sea ice margin (degrees)</td>
<td>71.3</td>
<td>74.2</td>
<td>77.3</td>
<td>80.6</td>
<td>86.8</td>
</tr>
<tr>
<td>Water column (Kg m$^{-2}$)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water vapor</td>
<td>18.8</td>
<td>20.2</td>
<td>22.0</td>
<td>24.6</td>
<td>30.8</td>
</tr>
<tr>
<td>Cloud liquid water</td>
<td>0.081</td>
<td>0.086</td>
<td>0.091</td>
<td>0.094</td>
<td>0.091</td>
</tr>
<tr>
<td>Cloud ice water</td>
<td>0.016</td>
<td>0.017</td>
<td>0.018</td>
<td>0.019</td>
<td>0.020</td>
</tr>
<tr>
<td>Cloud radiative forcing (W m$^{-2}$)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shortwave</td>
<td>-34.3</td>
<td>-34.4</td>
<td>-34.4</td>
<td>-33.8</td>
<td>-31.1</td>
</tr>
<tr>
<td>Longwave</td>
<td>25.3</td>
<td>25.3</td>
<td>25.6</td>
<td>26.1</td>
<td>26.2</td>
</tr>
<tr>
<td>Net</td>
<td>-9.0</td>
<td>-9.1</td>
<td>-8.8</td>
<td>-7.7</td>
<td>-4.9</td>
</tr>
</tbody>
</table>

Table 4.4. Nitrogen sensitivity tests.
surface pressure is increased. The strongest effect is found in the 3 PAL N₂ simulation where the magnitude of the global mean shortwave cloud forcing is reduced by 3.3 W m⁻². For comparison, simulations where the total surface pressure is kept uniform, increasing the surface temperature tends to add slightly to the strength of shortwave cloud forcings (Figure 3.8). Simulations with increased surface pressure also show a small increase in the cloud greenhouse effect. Steeper lapse rates result in cold temperatures and larger relative humidities in the upper troposphere, causing a greater incidence of high ice clouds. However, this effect on climate remains small, contributing <1 W m⁻² of additional forcing to the cloud greenhouse effect.

As a primary result, our simulations generally agree with the predictions made by Goldblatt et al. (2009). With 2 PAL N₂ global mean surface temperatures increase by 5.0 K to 292.9 K. With 3 PAL N₂ global mean surface temperatures increase by 9.7 K to 296.6 K. Our 3 PAL N₂ simulation exhibits slightly more warming from increasing N₂ than is shown by Goldblatt et al. (2009). Differences are likely accounted for by the subtle positive cloud feedbacks found here and from magnified pressure broadening effects as here we use 0.06 bar of CO₂ as a baseline case as opposed to 0.02 bar CO₂ and 10⁻⁴ bar CH₄ as a maximum in Goldblatt et al. (2009).

4.6. Optimal warming solutions

For a second set of simulations we combine the above discussed warming mechanisms to construct optimal climate solutions for the duration of the Archean
(3.8–2.5 Ga, corresponding to solar constants from 75% to 82% of the present day). We attempt to make realistic assumptions for the Archean climate system and then determine the atmospheric \( \text{CO}_2 \) needed to maintain modern day global mean surface temperatures (~288 K).

We chose a rotation rate of 18 hours per day for all simulations in this section. While the Earth’s rotation rate has varied continuously over time, the rate of despining was slow during the Archean (Denis, 2002). Regardless, the chosen rotation rate has only a minor effect on global climate compared with other influences discussed here (see section 4.2). We assume that cloud droplet number concentrations were 20 percent of their present day value (relative to the default settings in CAM3, see Table 4.3). This choice is consistent with estimates of CDNC if sea salt and dust are assumed to be the only available CCN for the early Earth (Fan & Toon, 2011; Karydis et al., 2011). This assumption yields liquid cloud droplet effective radii of 13.7 \( \mu \text{m} \) over land and 23.9 \( \mu \text{m} \) over oceans and sea ice. Averaged globally, liquid cloud droplet effective radii are 19.1 \( \mu \text{m} \) in the lower troposphere. Note that this choice for the cloud droplet effective radii is less than that postulated by Rosing et al. (2010), but is ~2 \( \mu \text{m} \) greater than is observed in unproductive regions of the oceans today (Breon et al., 2002). We assume land surface characteristics corresponding to dark bare soil on all land surfaces. Thus we implicitly assume that all emergent continents are dominated by basaltic rock. The albedo of dark bare soil is nearly indistinguishable from open ocean. Furthermore, replacing forested grid cells with bare soil \textit{(i.e.} desert) imparts a modest reduction to low level clouds over land (see section 4.3). We test two separate atmospheric
nitrogen inventories, corresponding to 1 PAL N₂ and then 2 PAL N₂. The 1 PAL N₂ case is safely within bounds set by geochemical indicators (Som et al., 2012; Marty et al., 2013). While 2 PAL N₂ is larger than their most probable values, it remains within the absolute upper limit postulated by Som et al. (2012) for 2.7 Ga and well within the theoretical limit given by Goldblatt et al. (2009). The timeframe for the draw down of atmospheric N₂ remains uncertain. However, logic would dictate that atmospheric N₂ inventories were likely to have been larger during the early Archean and smaller towards the end of the Archean. However, for simplicity here we keep N₂ constant at all times. We also include 10⁻⁴ bar of methane in our simulations. Photochemical ecosystem models predict that up to 0.035 bar of CH₄ could have been present in the anoxic early atmosphere (Kharecha et al., 2005), thus 10⁻⁴ bar is a rather modest number. Atmospheric CH₄ would have been strongly modulated by the presence of early life and atmospheric chemistry, however here it is a fixed parameter. The inclusion of 10⁻⁴ bar of methane can yield a ~6 K increase in global mean surface temperature (Haqq-Misra et al., 2008).

Optimal simulations benefit strongly from the combined effects of systematic warming mechanisms along with modest atmospheric CH₄. For the later Archean, optimal simulations have no trouble warming the early Earth to global mean surface temperatures of 288 K well within constraints on paleo-atmospheric CO₂ (Figure 4.8). With 2 PAL N₂, at the end of the Archean (2.5 Ga, 82% solar constant) only 0.0024 bar of CO₂, about 6 PAL, is needed to maintain present day surface temperatures. If we are limited to only 1 PAL N₂ still only 0.0048 bar of CO₂, about 12 PAL, is required at 2.5 Ga. Both solutions require CO₂ concentrations well below
Figure 4.8. CO₂ needed to maintain global mean surface temperatures of 288 K. Amount of carbon dioxide needed to maintain current global mean surface temperatures (288 K) over the course of the Archean. The dark line shows the amount of CO₂ needed, if CO₂ alone is used to warm the Archean to present day surface temperatures. Warmed by CO₂ only, current temperatures cannot be maintained within constraints given on CO₂ (marked on figure, Hessler et al., 2004; Sheldon 2006; Rosing et al. 2010; Driese et al. 2011). The orange and red lines shows optimal warming solutions with 1 and 2 PAL N₂ respectively. Optimal warming solutions also include 10⁻⁴ bar of CH₄, an 18 hour rotation rate, dark soil surface, and 20% CDNC. For 2 PAL of N₂, warm Archean solutions can be maintained throughout the Archean at CO₂ amounts below paleosol constraints on CO₂. Temperatures significantly warmer than 288 K could easily be achieved if CO₂ were raised up to the paleosol constraints (~0.01 to 0.02 bar CO₂) and if CH₄ were raised such that CH₄:CO₂ ≤ 0.1. The CO₂ constraint given by Rosing et al. (2010) remains difficult to satisfy.
the upper most limits proposed by Driese et al. (2011) and Sheldon (2006) of 0.018 bar and 0.025 bar respectively for the late Archean. At the beginning of the Archean (3.8 Ga, 75% solar constant) with 2 PAL N₂ and the warming mechanisms discussed above, present day global mean surface temperatures can be maintained with only 0.0147 bar of CO₂, about 37 PAL. This again remains within the bounds on paleoatmospheric CO₂ defined by Driese et al. (2011) and Sheldon (2006). Thus with 2 PAL N₂ we can maintain a warm Earth within paleosol constraints at all times of the Archean. However, with only 1 PAL N₂, 0.032 bar of CO₂, about 80 PAL, is needed to reach mean surface temperatures of 288 K at 3.8 Ga. This CO₂ concentration marginally exceeds the upper most limits derived from paleosols. However, CO₂ constraints are only defined for the later Archean and it is generally believed that there may have been more atmospheric CO₂ during the early Archean as opposed to the late Archean. Conversely, solutions that rely only on a CO₂ greenhouse, with no CH₄ or additional warming mechanisms, cannot keep the early Earth warm within proposed constraints on paleo-atmospheric CO₂ at any time during the Archean.

The ratio of CH₄ to CO₂ remains less than 0.1 in all of our optimal warming simulations. Thus in all cases Titan-like photochemical hazes would not be expected to form (Domagal-Goldman et al., 2008; Haqq-Misra et al., 2008; Zerkle et al., 2012) and thus there is no need to incorporate their effects in this study. Hazes may have cooled the Earth via the anti-greenhouse effect, but may also have provided some benefits. A haze layer may have shielded early life forms from harsh UV and may also have protected NH₃, a potent greenhouse gas, from photolysis (Sagan and
Chyba, 1997; Wolf and Toon, 2010). With either 1 PAL N₂ or 2 PAL N₂, the CO₂ required to reach present day surface temperatures for the late Archean remains below paleosol derived constraints. Thus we could allowably increase both CO₂ and CH₄, warming climate significantly above 288 K while still avoiding the issue of photochemical hazes.

CO₂ constraints for the early Archean remain unknown, as geochemical indicators primarily exist from the late Archean. It is conventionally believed that more CO₂ was in the atmosphere during the early Archean and then was slowly drawn down over time by the carbonate-silicate cycle. Given more lax constraints, one may suppose a case for the earliest Archean with 0.04 bar of CO₂ and 0.004 bar of CH₄ without significant haze formation. In such a case, for the earliest Archean (3.8 Ga, 75% solar constant) along with optimal warming assumptions given above, the global mean surface temperature reaches 307.5 K in our model while the tropical sea surface temperatures reach 315 K. While these temperatures still fall short of those suggested by Kauth and Lowe (2003) and Robert and Chaussidon (2006), with the aid of non-greenhouse warming mechanisms “hot” Archean temperatures may be approached. Our study of hot Archean climates is limited as CAM3 encounters numerical instabilities when global mean surface temperatures push past ~313 K (Wolf and Toon, 2014).

Even under our optimal warming assumptions and 2 PAL N₂, present day surface temperatures cannot be achieved near the CO₂ constraint of Rosing et al. (2010). While increased CH₄ could theoretically resolve this issue, large CH₄ concentrations would likely lead to the formation of thick photochemical hazes.
Possibly other greenhouse gases such as NH₃, C₂H₆ or OCS could help rectify this discrepancy however they remain unproven. However, the plurality of CO₂ constraints place the upper limit well above that of Rosing et al. (2010). Hessler et al. (2004) place a lower limit on CO₂ at 3.2 Ga at 0.0025 bar in direct contradiction to Rosing et al. (2010). The Rosing et al. (2010) constraint remains an outlier among geologically derived constraints on atmospheric CO₂. Some have argued that the methodology used by Rosing et al. (2010) is not robust (Dauphas & Kasting, 2011; Reinhard & Planavsky, 2011). Certainly the debate on paleoatmospheric CO₂ will continue as new measurements come to light. At present, we find the Rosing et al. (2010) constraint on CO₂ to be too low to allow us to resolve the faint young Sun paradox.

4.7. Summary

In this chapter we have examined systematic changes to the Earth’s rotation rate, surface albedo, cloud properties, and atmospheric nitrogen content in hopes of incorporating non-greenhouse means of warming the early Earth. While the overall effect of an increased rotation rate is slight, assumptions of darker land surface albedos, reduced cloud condensation nuclei, and increased atmospheric N₂ all yield modest global mean surface temperature increases; from 3 to 7 K each given plausible assumptions. One can argue convincingly for the validity of the above discussed warming mechanisms, however they remain difficult to explicitly prove through the geological record. While no one mechanism provides a singular solution to the faint young Sun paradox, an aggregate solution which incorporates
modest CH$_4$ concentrations along with the discussed warming mechanisms can resolve the faint young Sun paradox for all times of the Archean without violating constraints on CO$_2$ derived from paleosols. Hot Archean temperatures may even be possible. Combining lowered expectations for Archean seawater temperatures (Jaffrés et al. 2007; Hren et al. 2009; Blake et al. 2010), relatively lenient atmospheric CO$_2$ constraints (Sheldon, 2006; Dreise et al. 2011), wide uncertainties on atmospheric CH$_4$ concentrations (Kharecha et al. 2005), and numerous additional mechanisms for providing surface warming (Goldblatt et al. 2009; Rosing et al. 2010), there may be numerous permutations for resolving the faint young Sun paradox. Recent results from numerous different general circulation models indicate that sufficiently warming the early Earth does not now appear to be a problem (Wolf and Toon 2013; Charnay et al. 2013; Kunze et al. 2013). Inter-comparison of models remains an important tool for gaining confidence in our theoretical results. However, the precise combination of solar constant, surface temperature, CO$_2$, CH$_4$, N$_2$ and other effects may remain difficult to pin down precisely without better geological constraints.
Chapter 5

A fractal aggregate model of early Earth photochemical hazes

5.1. Overview

The atmosphere of the Archean Earth (3.8 – 2.5 Ga) was much different from the present one. The prevailing view of the Archean atmosphere is that it consisted primarily of N₂ with lesser amounts of CO₂, CH₄, H₂, and H₂O. Solar evolutionary models predict that the young Sun was up to ~25% less luminous than now (Sagan and Mullen, 1972; Gough, 1981), but the lack of evidence for glaciation combined with positive evidence for primitive life indicates that Archean surface temperatures were generally as warm or warmer than today (Knuath and Lowe, 2003; Sleep and Hessler, 2006; Robert and Chaussidon, 2006). Resolving this paradox is important for understanding the environment of Earth at the time of the origin of life. While a dense CO₂ atmosphere can theoretically warm the early Earth, the absence of siderite in fossil weathering profiles constrains the amount of CO₂ present in the young atmosphere (Rye et al. 1995; Sheldon, 2006; Driese et al., 2011). Combined greenhouse warming from CO₂, CH₄, NH₃, and other less abundant gases is typically invoked to resolve this apparent paradox. Past studies indicate that N₂ – CH₄ photochemistry produces an organic haze at high altitudes that could cool the planet, offsetting any greenhouse warming (McKay et al. 1999; Pavlov et al., 2001; Haqq-Misra et al., 2008). We show here that the haze may remain optically thinner
at visible wavelengths then previously asserted and therefore have less cooling effect on climate, but could simultaneously be optically thick at ultraviolet (UV) wavelengths and thus could shield reduced gases from photolysis. The key is to consider the fractal nature of the haze particles.

Atmospheric conditions similar to those that prevailed on the young Earth exist currently on Titan, a moon of Saturn. Titan’s atmosphere is strongly reducing, consisting of 98.4% $N_2$ along with 1.6% $CH_4$ and other hydrocarbons. Photochemical production of organic aerosols on Titan produces an optically thick haze that has been much studied (Tomasko et al., 2008). Similar organic aerosols are observed to form readily even in weakly reducing early Earth-like laboratory gas mixtures having the ratio of $CH_4$ to $CO_2$ as low as 0.1 (DeWitt et al., 2009). A Titan-like photochemical haze should have formed high in the Archean atmosphere as well. Sagan and Chyba (1997) argued that an organic haze may have provided a strong UV shield, protecting underlying constituents and primitive organisms from photochemical destruction. Of particular interest, if $NH_3$ is protected by a strongly UV absorbing haze and allowed to accumulate, a surface mixing ratio of $10^{-5}$ could alone provide sufficient greenhouse warming to keep Archean surface temperatures above freezing (Sagan and Mullen, 1972). However, recent modeling studies have suggested that the haze would be of a similar optical thickness in both the UV and the visible (Pavlov et al., 2001; Haqq-Misra et al., 2008). Thus any increase in UV shielding is accompanied by a thickening at visible wavelengths, which would cause intense antigreenhouse cooling and freeze the planet.

Previous models of the Archean haze have inaccurately made the simplifying
assumption of particle sphericity. Hydrocarbon aerosols have long been known to form fluffy aggregates that exhibit fractal structure (Bar-Nun et al., 1988). The fractal structure affects both the microphysical and radiative properties of the haze. Given the expected chemical and microphysical similarities between Titan and early Earth, we simulate haze particles as fractal aggregates using a scheme introduced by Cabane et al. (1993) and later used by Rannou et al. (1995; 1997; 2003) to model fractal aggregate hazes in Titan’s atmosphere. The fractal model is successful in reproducing the scattering properties of Titan’s haze (Tomasko et al., 2008).

We first discuss the modeling procedure with detailed discussions of fractal microphysics and fractal optical properties. Then we conduct a series of microphysical simulations of the haze. This first set of simulations is not coupled to radiative transfer code and benefits from much shorter equilibration times. Finally, we couple the haze to the radiative transfer code and assess its effect on early climate.

5.2. Model

Modeling experiments of the Archean haze layer were conducted using the Community Atmosphere Model version 3 (CAM3) coupled to the Community Aerosol and Radiation Model for Atmospheres version 2.3 (CARMA) microphysical model. The model utilizes the sectional microphysical modeling capabilities of CARMA within the CAM3 framework. CARMA has been modified to simulate fractal aggregate particles. CAM3 controls advection and wet deposition while CARMA controls production, vertical diffusion, coagulation, and dry deposition (Bardeen et
al., 2008). The model is configured with $4^\circ \times 5^\circ$ horizontal resolution and 66 vertical levels extending to a minimum pressure of $\sim 5 \times 10^{-6}$ mb. Note that we do not have a reducing photochemistry model to determine haze production. Thus, haze production rates are specified.

CARMA has been configured with 40 mass dependent size bins, spanning aggregate radii from 1 nm to $\sim 1$ mm. Haze aerosols are produced in a temporally and horizontally uniform manner and are initially sourced into the smallest mass bin (corresponding to $r = 1$ nm). The vertical distribution of haze production is specified by a user defined source function. Here, the haze source function is adapted from the photochemical modeling results of Pavlov et al. (2001). Using a one-dimensional chemical model, Pavlov et al. (2001) found that early Earth haze production likely occurred between altitudes of 40 and 80 km, peaking near 65 km. Pavlov's calculation agrees with the earlier photochemical modeling work of Zahnle (1986). Here the haze source function is chosen to be lognormally distributed about a peak production altitude of 65 km (figure 5.1). Haze particles are assumed to have a material density of $\rho_m = 0.64 \text{ g cm}^{-3}$ (Trainer et al., 2006).

5.3. Fractal Microphysics

Our model simulates haze particles as fractal aggregates rather than as spherical “liquid drop” particles. The method used improves upon a scheme first introduced by Cabane et al. (1993) and later used by Rannou et al. (1995, 1997, 2003) to model fractal aggregate hazes in Titan’s atmosphere. Fractal aggregates are composed of many primary spherical particles (called monomers) that adhere
Figure 5.1. Early Earth haze production rate global mean vertical profile. The source function for haze production rates between $10^{12}$ and $10^{15}$ g yr$^{-1}$. The vertical profile of the haze source function has been adapted from the 1-D photochemical modeling work of Pavlov et al. (2001). In this study the haze is assumed be produced in a temporally and horizontally uniform manner.
together forming irregularly shaped, fluffy aggregates. Though the shape of each individual fractal aggregate can appear quite complex, fractal aggregates have the unifying property of self-similarity. The structural properties of aggregate particles can be quantified to a first approximation by a single parameter, the fractal dimension, denoted $D_f$. The fractal dimension describes the morphological dimension of the aggregate particles; $D_f = 3$ describes a compact spherical particle while $D_f = 1$ describes a linear chain of monomers. In the case of fluffy aggregates ($D_f < 3$), the particle radius is not so easily defined. Fractal geometry provides the needed mathematical framework to proceed. The geometric radius of a fractal aggregate is defined by the equation,

$$n_{mon} = \alpha \left( \frac{R_f}{r_{mon}} \right)^{D_f}$$  \hspace{1cm} (eq. 5.1)

where $n_{mon}$ is the number of monomers contained in the aggregate, $\alpha$ is the dimensionless packing coefficient (taken here to be unity), $R_f$ is the fractal (or geometric) radius, and $r_{mon}$ is the average monomer radius (Mandlebrot, 1977).

In this work the fractal dimension, $D_f$, and the average monomer radius, $r_{mon}$, are free parameters. The number of monomers per aggregate, $n_{mon}$, is proportional to the aggregate mass and thus after appropriate selection of $D_f$ and $r_{mon}$ each aerosol mass bin can be assigned a corresponding fractal radius, $R_f$. The fractal radius can be thought of as the geometric collision radius of an aggregate particle. As such, $R_f$ is used in expressions for the Knudsen number and for coagulation kernels in the microphysical code. Note that the fractal radius of an aggregate with $D_f < 3$ is always greater than the radius of an equal mass sphere, thus fractal
aggregates experience enhanced areas for coagulation and diminished non-continuum effects when compared with spherical particles.

Use of the above defined fractal radius in Stokes law and in the calculation of diffusion coefficients is not fully accurate. Fractal aggregates are permeable, thus fluid can flow through their structure (Li and Logan, 2001; James et al., 2002). This effect causes a decrease in the Stokes drag force and an increase in the molecular diffusivity compared with predictions based on $R_f$. Here, the aggregate permeability scheme suggested by Vainhstein et al. (2004) is used to calculate the fractal mobility radius, $R_m$. The fractal mobility radius is the radius of an equal mass sphere that experiences the same Stokes drag force as the aggregate in question. The effect of permeability is most noticeable for smaller aggregates ($n_{mon} < 1000$) with low fractal dimensions ($D_f < 2$) where $R_m$ can be up to 25% less than $R_f$. The fractal mobility radius of an aggregate with $D_f < 3$ is always greater than the radius of an equal mass sphere, thus fractal aggregates experience reduced fall velocities and reduced diffusion coefficients when compared with spherical particles.

Studies of Titan hydrocarbon aerosols have assumed that all aggregates, regardless of size, have a fractal dimension of 2 (Cabane et al., 1993; Rannou et al., 1995, 1997, 2003; Tomasko et al., 2008). This value is in general agreement with theoretical calculations of diffusion limited cluster-cluster aggregation in both the ballistic and continuum regimes. (Meakin, 1983; Julien, 1984). Though valid as a first approximation, the assumption of a constant fractal dimension across all size bins misses an important nuance of fractal aggregate microphysics. Terrestrial carbonaceous fractal aggregate aerosols are observed to restructure, becoming
more compact (higher $D_f$) as the number of monomers contained in the aggregate increases (Xiong & Friedlander, 2001; di Stasio et al., 2002; Onischuk et al., 2003; Kostoglou et al., 2006). The fractal dimension of early Earth (and Titan) haze aggregates therefore should not be considered constant, but rather will vary across our modeled aggregate size distribution. Hydrocarbon aerosols initially condense into nanometer sized spherical particles. The initial growth phase is characterized by a high fractal dimension ($D_f \sim 3$) as monomers are built molecule by molecule forming spherical particles with radii ranging from 10 to 100 nm. In the secondary growth phase, spherical monomers coagulate into short linear chains of low fractal dimension ($n_{mon} < 100$, $D_f \sim 1.5$). As aggregates grow large restructuring becomes important. Chain-like aggregates tend to be electrically charged. During the restructuring process oppositely charged aggregate limbs attach resulting in more compact arrangements. Laboratory experiments have shown that electrical restructuring increases the fractal dimension of carbonaceous aggregates to $D_f \sim 2.4$ (Onischuk et al., 2003). Aggregate restructuring is also triggered by Brownian motion of monomers within an aggregate and by surface energy minimization of condensed water trapped within an aggregate (Xiong & Friedlander, 2001; Kostoglou et al., 2006). Restructuring is parameterized in the microphysical code by incorporating a size bin dependent fractal dimension (figure 5.2). Incorporating some means of aggregate restructuring is necessary to ensure realistic particle size distributions. The end result of the complex restructuring process is that the fractal dimension tends to increase with aggregate mass.
Figure 5.2. Fractal aggregate restructuring parameterization. The fractal dimension of aggregate aerosols varies with the number of monomers per aggregate (or analogously with the aggregate mass). Initial growth is characterized by a high fractal dimension as nanometer sized particles coalesce to form spherical monomers. The second growth stage is characterized by a low fractal dimension as spherical monomers coagulate to form linear chain-like structures. As aggregates grow larger, they tend to collapse into more compact arrangements resulting in a higher fractal dimension. Illustrated is the parameterization used in this study.
5.4. Fractal Optics

As mentioned above, previous models of the early Earth haze layer have assumed that all particles are spheres and have therefore used Mie theory to quantify the haze optical properties (Pavlov et al., 2001; Haqq-Misra et al., 2008). Fractal aggregates, however, interact quite differently with radiation than do equal mass spheres. Of particular note, the optical size parameter (the ratio of particle circumference to wavelength of light) for fractal particles is based on the monomer radius, while for Mie theory it is based on the total particle radius. Conceptually, one may think of the scattered electric field from an aggregate as the sum of the scattered fields from each monomer contained within.

For spheres, once the size parameter exceeds about 2, the optical properties tend to have reached values that no longer change significantly for larger sized particles or for shorter wavelengths. Hence if we consider a wavelength of 0.5 μm, spherical particles larger than about 160 nm will have similar visible and ultraviolet optical properties. Particles larger than 160 nm were found in previous simulations of the Archean Earth assuming monodisperse spherical particles (Pavlov et al., 2001; Haqq-Misra et al., 2008). Consequently, they found that no ultraviolet shielding could occur without also causing strong antigreenhouse cooling.

Considerable work has been conducted on the theory of scattering and absorption by fractal aggregate particles (Draine & Flatau, 1994; Xu, 1995; Botet et al., 1995, 1997). Here, we adopt the mean-field approximation of multiple scattering by fractal aggregates proposed by Botet et al. (1997). This is a mean-field approximation of T-matrix theory for calculating scattering by non-uniform
particles. The benefit of the mean-field method is that it requires substantially less computational time as compared to full T-matrix theory. Rather than explicitly calculating the scattering intensities by each individual monomer within an aggregate, instead the mean-field approximation calculates an ensemble averaged scattering intensity that depends on the mean aggregate structure. The mean aggregate structure is determined by a monomer-monomer positional correlation function, instead of the exact positions of all monomers as is required by full T-matrix theory. Monomer-monomer positional correlation functions are determined from the fractal dimension assuming spherically symmetric aggregates.

The Botet mean-field code has been successfully validated against the full T-matrix theory calculations of fractal aggregate particles and provides superior results compared with Rayleigh-Debye-Gans theory (Xu, 1995; Botet et al., 1997; Lattuada and Ehrl, 2009; Soos et al, 2009). Validations against the full T-matrix theory have spanned monomer size parameters of ~0.5 to 5 with monomer numbers up 1000 and fractal dimensions varying from 1.8 to 3.0 (Botet et al., 1997; Lattuada and Ehrl, 2009). However, only refractive indices with negligible imaginary parts have been considered. In our study, monomer optical size parameters are ~1 and monomers numbers range can reach ~10^4. Laboratory experiments support the validity of T-matrix codes for calculating optical properties of fractal aggregates however data remains sparse (Xu and Gustafson, 1997; Chakrabarty et al., 2007).

The mean-field code requires inputs of n_{mon}, D_f, and r_{mon} along with the incident wavelength and complex index of refraction. Here, the indices of refraction
determined for Titan analog hydrocarbon hazes are used (Khare et al., 1984). The incorporation of oxygenated species into the haze has been shown to more than double the imaginary part of the refractive index (S26), however data is only available at a single wavelength, $\lambda = 0.532$ $\mu$m (Hasenkopf et al. 2010). It unclear how the wavelength dependence of optical constants for oxygenated haze particles would differ, if at all, from those derived for Titan analogs. The code outputs the extinction ($Q_e$), absorption ($Q_a$), and scattering ($Q_s$) coefficients as well the asymmetry parameter ($g$).

As illustrated in figure 5.3, fractal aggregates are considerably more absorbing in the UV while being slightly more transparent in the visible and near-IR than equal mass Mie particles. This trend becomes more pronounced as $n_{mon}$ increases. An aggregated composed of $10^3$ monomers is $\sim 15$ times more absorbing in the UV than an equal mass Mie particle. Figure 5.4 shows the asymmetry parameter for early Earth fractal haze particles plotted against wavelength. Fractal particles are slightly less forward scattering at short wavelengths ($\lambda < 0.5$ $\mu$m) while being more forward scattering at longer wavelengths when compared to equal mass Mie particles. Figure 5.5 shows the single scattering albedo as a function of wavelength for a 1000 monomer aggregate and an equal mass Mie particle. The aggregate particle is more absorbing than a Mie particle at wavelengths below 200 nm, while being more scattering in the mid-visible.

We have also conducted a series of sensitivity studies that indicate the UV shielding characteristics of fractal aggregates are largely independent of the chosen refractive indices. Cases using imaginary refractive indices 5 times larger and 5
Figure 5.3. The ratio of fractal aggregate optical efficiencies to Mie optical efficiencies versus wavelength for equal mass particles using Khare’s optical constants. The ratio of absorption (solid line) and extinction (dashed line) efficiencies calculated for fractal aggregate particles to those calculated for equal mass Mie particles. A value of unity indicates that the aggregate particle is radiatively indistinguishable from the Mie particle. The particles illustrated have $D_f = 2$, $r_{mon} = 50$ nm, and $n_{mon} = 10, 100,$ and 1000 corresponding to fractal radii of 0.16 µm, 0.5 µm and 1.6 µm respectively (see equation 1). The particles are assumed to have the refractive indices of Titan analogs (Khare et al., 1984). Fractal aggregates particles are an order of magnitude more absorbing in the UV while being slightly less absorbing in the visible than equal mass Mie particles. The gradient increases for larger aggregates containing more monomers.
Figure 5.4. Fractal aggregate asymmetry parameters versus wavelength. The asymmetry parameter for fractal aggregates with $r_{mon} = 50$ nm, $D_f = 2$ comprised of 10 and 1000 total monomers (dashed lines). The asymmetry parameter for equal mass spherical particles (solid lines).
Figure 5.5. Fractal aggregate single scattering albedos versus wavelength. The single scattering albedo for fractal aggregates with $n_{\text{mon}} = 1000$, $r_{\text{mon}} = 50$, and $D_f = 2$ (dashed line). The single scattering albedo for equal mass spherical particles (solid line).
times smaller than those found by Khare et al. (1984) both indicate shielding by fractal aggregates preferentially at short wavelengths (Figure 5.6). UV shielding is also found in a test case using a constant refractive index across all wavelengths (Figure 5.7). The fractal optical model preserves UV shielding regardless of the wavelength dependence of the chosen optical constants. This is because the optical size parameter (the ratio of particle circumference to wavelength of light) for fractal aggregates is based on the monomer radius, not the total aggregate radius.

In scattering atmospheres the extinction optical depth does not provide an accurate estimate of the attenuation of downwelling radiative flux. The extinction optical depth overestimates attenuation by neglecting the fact that forward scattered photons remain in the downwelling beam. A more appropriate measure is the effective optical depth given by

$$\tau_{\text{eff}} = \left[ 1 - \frac{\omega}{2} - \frac{g \omega}{2} \right] \tau_{\text{ext}}$$  \hspace{1cm} \text{(eq. 5.2)}

where $g$ is the asymmetry parameter, $\omega$ is the single scattering albedo, and $\tau_{\text{ext}}$ is the extinction optical depth (Chylek and Wong, 1995). Early Earth hazes are strongly forward scattering in UV so that $g > 0.8$, and thus $\tau_{\text{eff}}$ is only a few percent greater than the absorption optical depth, $\tau_{\text{abs}}$, in this regime. At infrared wavelengths, forward scattering becomes less pronounced and for Titan analog tholins $\omega$ trends toward 1. Then $\tau_{\text{eff}}$ can be an order of magnitude greater than $\tau_{\text{abs}}$. Visible wavelengths represent a transition region between the two regimes.
Figure 5.6. Fractal aggregate absorption enhancement sensitivity for scaled optical constants. The ratio of absorption efficiencies calculated for fractal aggregate particles to those calculated for equal mass Mie particles. A value of unity indicates that the aggregate particle is radiatively indistinguishable from the Mie particle. The particles illustrated have $D_f = 2$, $r_{mon} = 50$ nm, and $n_{mon} = 1000$. Cases are shown corresponding to refractive indices for Titan analogue hazes determined by Khare et al. (1984), five times greater Khare’s values, and five times smaller. In each instance aggregates absorb strongly in the UV while remaining relatively transparent in the mid-visible.
Figure 5.7 Fractal aggregate absorption enhancement sensitivity under wavelength independent optical constants. The ratio of absorption efficiencies calculated for fractal aggregate particles to those calculated for equal mass Mie particles. A value of unity indicates that the aggregate particle is radiatively indistinguishable from the Mie particle. The particles illustrated have $D_f = 2$, $r_{mon} = 50$ nm, and $n_{mon} = 10, 100, 1000$ corresponding to fractal radii of $0.16 \, \mu m$, 0.5 $\mu m$ and 1.6 $\mu m$ respectively. A constant refractive index of $n = 1.70 + i0.023$ is assumed at all wavelengths. Even after removing the wavelength dependence of refractive indices, fractal aggregate particles are more absorbing in the UV while being slightly less absorbing in the visible than equal mass Mie particles. This is due to the strong interaction of monomers contained in the aggregate with shortwave radiation.
5.5. Results from uncoupled simulations

Here, results from 10 uncoupled haze simulations are presented. This is done as to explore a wider range in parameter space in a computational efficient manner. Simulations take only 4 to 6 years to reach equilibrium as opposed to 60 or more for radiatively coupled haze simulations. A summary of the basic parameters and pertinent results for each run is shown in Table 5.1. Four runs were conducted using spherical (liquid drop) microphysics. Six runs were conducted using fractal microphysics. Haze production rates (Φ) were chosen to vary from $10^{12}$ to $10^{15}$ g yr$^{-1}$ in accordance with laboratory based estimates for both the pre-biotic and post-biotic Earth (DeWitt et al., 2009). In this section particular attention is paid to the simulations with a haze production rate of $10^{14}$ g yr$^{-1}$. For comparison, the current rate of mass production in the global sulfur cycle is about $1\cdot2 \times 10^{14}$ g yr$^{-1}$ (Pham et al., 1995), while the current rates of organic C burial and of methane production are of the same order of magnitude (Pavlov et al., 2001).

In this work, $r_{mon}$ is a free parameter that affects both the fractal microphysical and fractal optical models. Recent observations of Titan fractal aggregate hazes have constrained monomer radii to be 50 nm or smaller based on measurements of the phase function and linear polarization (Tomasko et al., 2008). Here we choose $r_{mon} = 50$ nm as a standard value, however values of 20 nm and 100 nm are also explored in limited cases to ensure the results presented here are not artifacts of convenient parameter selection. The haze effective optical depth as a function of wavelength is calculated using the model outputted particle size distributions in conjunction with the relevant optical parameters derived from the
<table>
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<th>$\tau_{vis}$ (564 nm)</th>
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Table 5.1. Uncoupled haze simulations.
mean-field approximation for multiple scattering aggregates for the fractal model and Mie theory for the liquid drop model.

The highest particle number densities occur in the production region (figure 5.8). The haze particles grow rapidly due to coagulation in the production region where number densities are large and collisions frequent. Nanometer sized haze particles can diffuse upwards, while larger particles fall into the lower atmosphere where they continue to grow in size before eventually being removed by wet deposition in the troposphere or dry deposition at the ground.

The annual mean zonally averaged haze mass concentration for the fractal model is plotted in figure 5.9. The haze is distributed in a zonally uniform manner with higher mass concentrations over the poles than over equatorial regions. The steep gradient is caused by meridional mesospheric circulations that transport haze from the summer pole to the winter pole where the haze particles then descend into the lower atmosphere. There is a slight bias towards mass loading over the southern hemisphere pole due to its stronger polar vortex that enhances transport (Bardeen et al., 2008). The bulk of the haze mass is concentrated between 10 and 30 km altitude.

Fractal aggregate hazes have longer atmospheric lifetimes than do liquid drop particles. Aggregate sedimentation velocities are reduced as a result of their larger mobility radii. Atmospheric lifetimes calculated for each simulation are listed in Table 5.1. Haze particles have atmospheric lifetimes of up to ~3 years depending on the production rate. The atmospheric lifetime of haze particles decreases with increasing production rate in both fractal and liquid drop cases. Higher production
**Figure 5.8. Zonal mean haze number density.** Zonal mean annual average number density for a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{mon} = 50$ nm. Particles are initially produced in the model with a radius of 1 nm. Haze number densities peak in the photochemical production zone where sub-monomer sized particles dominate the population. Outside the production zone number densities decrease rapidly due to coagulation of monomers to form larger particles.
**Figure 5.9. Zonal mean haze mass concentration.** Zonal mean annual average haze mass concentration for haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{mon} = 50$ nm. The haze mass distribution features steep latitudinal gradients with the bulk of the mass located over the polar regions. In our model the haze layer remains thin over the equatorial regions, however we expect that radiative feedbacks would smear out the distribution leading to a more latitudinally uniform coverage of haze.
rates lead to the formation of increasingly heavier haze particles that fall out of the atmosphere more rapidly regardless of the microphysical model used.

Particle effective radii reach micron sizes in the lower atmosphere. The effective radius is the area averaged particle size, which is the size most relevant for optical properties. Figure 5.10 shows the effective radii for the liquid drop model (solid line), the effective radii of equal mass spheres for the fractal model (dashed line), and the effective fractal radii (dash-dot line) for a haze production rate of $10^{14}$ g yr$^{-1}$. Note that the effective radii of equal mass spheres is an indicator of the mass per aggregate rather than its true geometric size. In the troposphere the effective radii of equal mass spheres is larger than the effective radii of liquid drops by a factor of $\sim 1.5$, thus showing that fractal aggregate particles grow to be more massive than do liquid drop particles. This trend is due to the increased geometric cross sections of aggregates, which simultaneously reduces sedimentation velocities and enhances coagulation allowing for the growth of more massive particles.

Geometrical information regarding fractal aggregates is described by the effective fractal radius (see equation 5.1). In the troposphere, effective fractal radii are larger than the liquid drop effective radii by a factor of $\sim 4$ and are more than double the effective radii of equal mass spheres. These ratios are even larger in the production region. Fractal aggregate particles grow to significantly greater geometric sizes while being only slightly more massive than liquid drop particles thus reflecting the fluffy and porous nature of their structures.

At each model grid point the average number of monomers per aggregate is calculated using the equation,
\[ n_{\text{mon}} = \frac{MD}{ND} \left( \frac{3}{4\pi\rho_m r_{\text{mon}}^3} \right) \]  

where \( MD \) is the mass concentration, \( ND \) is the particle number density, \( \rho_m \) is the haze material density, and \( r_{\text{mon}} \) is the average monomer radii. \( MD \) and \( ND \) are outputted by the microphysical model. Fluffy micron sized aggregates containing large numbers of monomers dominate the lower atmosphere (figure 5.11). Above \( \sim 50 \) km monomer and sub-monomer sized particles dominate the haze population. The number of monomers per aggregate increases with increasing haze production rate. For a haze production rate of \( 10^{14} \) g yr\(^{-1} \) we find early Earth haze particles typically contain \( \sim 10^4 \) monomers. For comparison, Titan aerosols are estimated to contain \( \sim 10^3 \) monomers per aggregate in its stratosphere, however estimates are not well constrained due to uncertainties in the fractal dimension, average monomer size and haze optical constants, all of which are required to determine \( n_{\text{mon}} \) from observations of Titan (Tomasko et al., 2008).

Varying the choice of \( r_{\text{mon}} \) has minimal effect on the total atmospheric mass, atmospheric lifetime, and the average mass per particle, however \( r_{\text{mon}} \) has a significant effect on the geometric size of aggregates. The similarity between the effective radii of equal mass spheres (figure 5.12a) for all choices of \( r_{\text{mon}} \) demonstrates that the average mass per particle is nearly identical in all cases. However, fractal effective radii vary significantly with \( r_{\text{mon}} \) (figure 5.12b). Since the average mass per particle is approximately constant for all choices of \( r_{\text{mon}} \), one can show that \( R_f \propto r_{\text{mon}}^{(D_f-3)/D_f} \). For \( D_f = 2 \) this equation reduces to \( R_f \propto r_{\text{mon}}^{1/2} \). In the lower atmosphere, aggregates formed by 20 nm monomers have effective radii \( \sim 50\% \)
Figure 5.10. Global mean vertical profile of spherical and fractal aggregate effective radii. The liquid drop effective radii (solid line), equal mass sphere effective radii (dashed line) calculated for aggregate particles and the effective fractal radii (dash-dot line) are shown for a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{mon} = 50$ nm. Fractal aggregates grow both more massive and geometrically larger than liquid drop particles.
5.11. Global mean vertical profile of the mean aggregate monomer number.
The average number of monomers per aggregate plotted versus height for haze production rates ranging from $10^{12}$ to $10^{15}$ g yr$^{-1}$ and $r_{\text{mon}} = 50$ nm. Larger production rates lead to higher monomer numbers.
greater than aggregates formed by 50 nm monomers and double that of aggregates formed by 100 nm monomers. Figure 5.13 shows the average number of monomers per aggregate plotted as function of altitude for a haze production rate of 10\(^{14}\) g yr\(^{-1}\) with average monomer radii of 20, 50 and 100 nm respectively. Since the average mass per aggregate is equal for all choices of \(r_{\text{mon}}\), it follows that \(n_{\text{mon}} \propto r_{\text{mon}}^{-3}\).

Table 5.1 lists the globally averaged effective optical depth in the mid-visible (564 nm, denoted \(\tau_{\text{vis}}\)) and in the UV (197 nm, denoted \(\tau_{\text{uv}}\)) for all simulations. For all haze production rates a fractal aggregate haze layer is an order of magnitude or more optically thick in UV while remaining equally thin in the visible when compared with a liquid drop haze layer. Results from the liquid drop simulation predict that the haze would still too optically thin in the UV (global mean, \(\tau_{\text{uv}} = 0.73\)) to provide effective shielding, but would be thick enough in the visible (\(\tau_{\text{vis}} = 0.55\)) to initiate antigreenhouse cooling. For the liquid drop model the ratio of effective optical depths is \(\tau_{\text{uv}} / \tau_{\text{vis}} = 1.3\). In this case the young Earth would be cooled by the haze while not benefiting from a significant accumulation of photolytically shielded greenhouse gases. However the liquid drop model is inappropriate since haze particles form fractal aggregates (Bar-Nun et al., 1988). The fractal simulation reveals that the haze layer was a highly effective UV shield. A fractal aggregate haze layer produced at a rate of 10\(^{14}\) g yr\(^{-1}\) would have sustained a steady state haze layer that was optically thick in the UV while remaining reasonable in the mid-visible. For an average monomer size of 50 nm, the global mean optical depths are \(\tau_{\text{uv}} = 11.23\), \(\tau_{\text{vis}} = 0.50\), and thus the ratio of effective optical depths is \(\tau_{\text{uv}} / \tau_{\text{vis}} = 22.4\). While some
Figure 5.12. Sensitivity of fractal effective radii to changing assumed monomer radii. (a) The equal mass sphere effective radii calculated for aggregate particles for a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{mon} = 20, 50,$ and $100$ nm respectively. The average mass per particle remains relatively unchanged for all choices of $r_{mon}$. (b) The fractal effective radii is sensitive to the choice of $r_{mon}$. A smaller choice for the average monomer size yields geometrically larger aggregates for a given haze production rate.
Figure 5.13. Sensitivity of aggregate monomer number to changing assumed monomer radii. The number of monomers per aggregate for a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{\text{mon}} = 20, 50, \text{and } 100$ nm. A smaller choice for $r_{\text{mon}}$ yields a larger number of monomers per aggregate.
Figure 5.14. Effective optical depth for spherical and fractal haze layers versus wavelength. The global mean effective optical ($\tau_{\text{eff}}$) depth plotted for both the liquid drop and fractal models for a haze production rate of $10^{14}$ g yr$^{-1}$ and $r_{\text{mon}} = 50$. Fractal aggregate particles (red line) provide strong UV shielding while remaining thin at visible wavelengths. A steep gradient is exhibited at wavelengths below 0.8 $\mu$m. On the contrary a spherical haze layer (blue line) exhibits no such gradient, shielding all wavelengths more or less equally.
Figure 5.15. Sensitivity of fractal haze effective optical depth to changing assumed monomer radii. The effective optical depth ($\tau_{\text{eff}}$) plotted for the fractal models for $\Phi = 10^{14}$ g yr$^{-1}$ and $r_{\text{mon}} = 20, 50, \text{and } 100 \text{ nm}$ respectively. Though subtle variations are present, in all cases the haze layer is thick in the UV while remaining relatively thin in the visible. The choice of $r_{\text{mon}}$ is not observed to affect the primary results of this work.
antigreenhouse cooling would still occur, the atmosphere would be virtually opaque to incoming ultraviolet radiation.

Figure 5.14 shows the globally averaged haze effective optical depths for both the liquid drop and fractal models as a function of wavelength. Notice that a fractal aggregate haze layer is optically thinner in the mid-visible and IR while being more than an order of magnitude thicker in UV when compared to the liquid drop model. The radiative properties of the haze layer show some sensitivity to the free parameter \( r_{\text{mon}} \), however the basic result holds true. Figure 5.15 shows the global mean effective optical depth versus wavelength for average monomer radii of 20, 50, and 100 nm. Both the 20 and 50 nm cases have similar wavelength dependence with \( \tau_{\text{av}} / \tau_{\text{vis}} \sim 22 \). For the case of \( r_{\text{mon}} = 100 \) nm, the wavelength dependence of the optical depth is less drastic. However, \( r_{\text{mon}} = 100 \) falls above the upper limit for average radii proposed by Tomasko et al. (2008) for Titan monomers and is not likely to represent reality. Though the magnitude of the optical depth is sensitive to the monomer radius, \( \tau_{\text{av}} / \tau_{\text{vis}} \) remains high in all cases and thus the choice of \( r_{\text{mon}} \) does not effect the primary results of this work.

5.6. Results from radiatively coupled simulations

Here we conduct a limited number of simulations with radiatively coupled hazes. A control simulation is conducted with 0.015 bar CO\(_2\) and 0.003 bar CH\(_4\). We also assume an 18 hour rotation rate, dark surface albedo and 50\% of the present day cloud droplet number concentration (see chapter 4) to emulate possible Archean conditions. The resultant global and annual mean surface temperature of
our control simulation is 296.2 K. We then conduct radiatively coupled haze simulations with both spherical and fractal physics with haze production rates ranging from $10^{12}$ to $5\times10^{13}$ g yr$^{-1}$. For each simulation the monomer sizes are assumed to be 50 nm and the aggregate restructuring parameterization follows that shown in Figure 5.2. While the haze layer itself reaches a steady state in 4 to 6 years, the response of climate is much slower, taking 60 years or more for sea ice and surface temperature to equilibrate.

Hazes simulated with spherical and fractal physics can both yield significant antigreenhouse cooling (figure 5.16a). However, global mean cooling from a haze layer described by spheres remains greater than that from a fractal haze layer. In both cases, global mean cooling of near 10 K occurs with annual haze production rates of only $5\times10^{12}$ g yr$^{-1}$. At this production rate, global mean haze effective optical depths at 220 nm (550 nm) are 0.98 (0.094) for a fractal haze and 0.15 (0.10) for a spherical haze (figure 5.16b). With a haze production rate of $5\times10^{13}$ g yr$^{-1}$, global mean surface temperatures plummet 24 and 30 K for fractal and spherical haze cases respectively. In this case haze effective optical depths at 220 nm (550 nm) are 6.39 (0.53) for a fractal haze and 0.62 (0.50) for a spherical haze.

However, due to the variation in the latitudinal distribution of the haze layer (figure 5.9), haze optical depths have a significant latitudinal gradient. Figure 5.17 shows the zonal mean effective optical depths for both fractal and spherical hazes at a production rate of $10^{13}$ g yr$^{-1}$. Optical depths over the arctic may be 3 to 4 times greater than that over the tropics. Thus both antigreenhouse cooling and ultraviolet shielding have a strong zonal dependence.
Figure 5.16. Global mean surface temperature and haze effective optical depth as a function of haze production rate. Panel (a) shows the global mean surface temperature for our control simulation with no haze (black) and then for fractal (red) and spherical (blue) haze models with varying haze production rates. Hazes can cause significant cooling. Global mean haze effective optical depths are shown in panel (b). A fractal aggregate haze has significantly greater optical depths in the UV compared with the visible.
Figure 5.17. **Zonal mean haze effective optical depth.** Stratospheric dynamics dictate that the majority of the haze mass is transported to the polar regions. Thus hazes are 3 to 4 times optically thicker over the poles compared with the tropics.
5.7. Discussion

Fractal aggregate hazes are observed on Titan and are readily produced in laboratory simulations using gas mixtures expected for the Archean Earth. The work presented here shows that a fractal aggregate haze layer could provide a strong UV shield for the early Earth since $\tau_{uv}/\tau_{vis} \geq 10$. Earlier work that assumed spherical particles argued that a haze layer could not provide significant UV shielding without causing dramatic antigreenhouse cooling since $\tau_{uv}/\tau_{vis} \sim 3$ (Pavlov et al., 2001). For comparison, here a spherical haze layer yields $\tau_{uv}/\tau_{vis} \sim 1$.

This work reopens the idea first proposed by Sagan and Mullen (1972) that the early Earth may have been home to a reducing atmosphere. A fractal aggregate haze layer as described in this work could have shielded reduced gases CH$_4$ and NH$_3$ from photolysis and allowed them to accumulate to high concentrations in the young atmosphere while still allowing visible light to penetrate down to the surface.

The presence of NH$_3$ in the atmosphere is of dual importance. NH$_3$ concentrations of $10^{-5}$ or more added to the Archean atmosphere could alone resolve the faint young Sun paradox (Sagan & Mullen, 1972). However, without UV shielding an ammonia concentration of $10^{-5}$ would be irreversibly converted into N$_2$ in less than 10 years (Kuhn and Atreya, 1979). Ammonia resupply rates during the Archean could not have supported an ammonia concentration above $10^{-8}$ without a strong UV shield provided by a Titan-like haze layer (Kasting, 1982). Sagan and Chyba (1997) determined that the atmospheric lifetime of NH$_3$ can be approximated by,
\[ t_{NH_3} = \frac{t'_{NH_3}}{e^{-\tau_{uv} \sec \Theta}} \]

where \( t_{NH_3} \) is the atmospheric lifetime of ammonia in the presence of an overlying haze layer, \( t'_{NH_3} \) is the atmospheric lifetime of ammonia in the absence of haze, \( \tau_{uv} \) is the haze optical depth at 200 nm, and \( \Theta \) is the zenith angle taken to be 45°. With an optical depth of 1, the atmospheric lifetime of ammonia is extended to \( \sim 41 \) years. With an optical depth of 5, the atmospheric lifetime of ammonia is extended to \( \sim 12,000 \) years. With an optical depth of 10, the atmospheric lifetime of ammonia is extended to \( \sim 10,000,000 \) years. The precise amount of NH_3 in the atmosphere will depend on HCN chemistry in the early atmosphere and oceans (Zahnle, 1986; Feng et al. 2011). However, the UV shielding nature of a fractal haze layer removes one major NH_3 sink. An ammonia rich atmosphere could have built up to high concentrations and persisted throughout much of the Archean period even if NH_3 sources were small.

The presence of high concentrations of NH_3 in the young atmosphere is also important because NH_3 plays an important role in pre-biotic synthesis. Highly reducing atmospheres produce amino acids and other organic molecules in spark discharge experiments (Miller, 1953). More recently it was confirmed in the laboratory that complex organics form in reducing atmospheres under UV irradiation (Trainer et al, 2006). More oxidizing atmospheres are far less conducive to pre-biotic synthesis of complex organics. The strong UV shielding characteristics of a fractal aggregate haze layer would allow the young atmosphere to remain reducing, thus making the young Earth a favorable environment for pre-biotic
synthesis. The haze layer itself would be composed of complex organic molecules, which would precipitate down into the primordial oceans providing a source of organics to the surface comparable to current carbon burial rates and methane production rates. A strongly UV shielding haze layer is also desirable because it would protect young organisms from destruction by the intense high frequency radiation of the young Sun (Ribas et al., 2005). A fractal aggregate haze layer quite elegantly provides a solution to faint young Sun paradox through the UV shielding of reduced gases while itself creating a large source of biological precursors.

5.8. Summary

The work presented in this chapter shows the ability of a Titan-like haze layer to act as an efficient UV shield on the early Earth. While antigreenhouse cooling can remain significant for fractal hazes, the ratio of the optical depth in the UV to the visible can be 10 or greater. By comparison, spherical models of hazes predict that optical depths would be about the same across the visible and UV spectrum. A UV shielding haze layer provides numerous interesting consequences for the early Earth. A UV shield may have allowed reduced gases like NH₃ to build up in the atmosphere and provide additional greenhouse warming. However the extent to which the constituents and chemistry of atmosphere would change is not precisely known. Furthermore a UV shield could have protected early life from harsh radiation sans an ozone layer. Future work will couple self-consistent haze chemistry with our general circulation model.
Chapter VI

Earth's future under the ever brightening Sun

6.1. Overview

Much of this work has been dedicated to discussing issues relevant to the early Earth; a paradigm dominated by the faint young Sun. Here we briefly discuss the inverse problem. While the standard solar model dictates that Sun was much dimmer in Earth's early history, it also predicts that the Sun will become much brighter as time marches forward. Mankind will have little choice but to adapt.

As the Sun slowly grows brighter over its main sequence lifetime, habitability on Earth's surface will eventually become threatened probably leading to moist and then runaway greenhouse climates. One-dimensional climate models predict that a catastrophic thermal runaway will be triggered by a 6 percent increase in the solar constant above its present level. However, here simulations using a three-dimensional climate model with fixed carbon dioxide and methane indicate that surface habitability may be maintained at significantly larger solar constants. A 15.5 percent increase in the solar constant yields global mean surface temperatures of 312.9 K, well short of moist and runaway greenhouse states. Numerical limitations prevent simulation of climates much warmer than this. Nonetheless, our results imply that Earth's climate may remain safe against both water-loss and thermal runaway limits for at least another 1.5 billion years and probably for much longer.
Life has proven to be remarkably resilient, surviving and recolonizing the Earth after snowball glaciations (Pierrehumbert et al., 2011), abrupt warming episodes (McInerney & Wing, 2011), and catastrophic meteor impacts (Toon et al., 1997). However, in the distant future a Venusian climate, which life may not survive, probably awaits our home planet. At present the Sun is increasing in brightness by 1% every ~110 million years (Gough, 1981). Increased solar radiation should at some point trigger moist and then runaway greenhouse climates, ending any hope for continued surface habitability. Surface habitability of a planet is traditionally defined as the ability to support liquid water.

The limits of Earth (and Earth-like exoplanet) habitability have been estimated using one-dimensional (1D) radiative convective climate models (Kasting, 1988; Kasting et al., 1993; Selsis et al., 2007; Kopparapu et al., 2013). However 1D models remain fundamentally deficient when it comes to probing the hot limit of planetary habitability. Three-dimensional dynamical calculations are critical for determining the distribution of water vapor and clouds in the atmosphere. Both clouds and the relative saturation of the atmosphere play important roles in determining the strength of the water vapor greenhouse feedback and thus the onset of a thermal runaway (Pierrehumbert, 1995; Rennó, 1997; Selsis et al., 2007; Goldblatt et al., 2013).

A classical runaway greenhouse is characterized by a positive feedback loop where rising surface temperatures accelerate evaporation, causing water vapor to become the dominant constituent in the atmosphere. Strong absorption by water vapor causes the atmosphere to become opaque to outgoing thermal radiation and
thus surface temperatures rocket upwards as the planet is unable to sufficiently shed energy to space. A stable climate may not be reached until surface temperatures reach 1600 K with the entirety of Earth’s oceans turning to vapor in the process (Goldblatt et al., 2013). Recent calculations using cloud-free 1D models with fully saturated atmospheres (100% relative humidity) predict that Earth will enter a catastrophic thermal runaway when the solar constant becomes 6% brighter than the present day (Kopparapu et al., 2013). This limit will occur in ~650 million years (Gough, 1981).

While a catastrophic runaway greenhouse would unquestionably sterilize the planet, habitability may become threatened before this ultimate tipping point is reached. A more stringent estimate for the hot limit to planetary habitability is based on the so-called moist greenhouse climate. A moist greenhouse is a stable climate state where hot temperatures throughout the atmosphere reduce the effectiveness of the tropical cold trap, allowing the stratosphere to become moist. If the stratosphere becomes sufficiently wet, the rate of hydrogen lost to space becomes large owing to water vapor photolysis. Kasting et al. (1993) estimate from 1D models that if the Earth’s mean surface temperature were to reach 340 K, the water vapor mixing ratio in the stratosphere would grow to ~3×10⁻³, increasing the photolysis rate sufficiently as to allow Earth’s oceans to effectively evaporate away to space in less than the age of the Earth. The moist-greenhouse limit is typically taken as the inner edge of planetary habitability in the context of exoplanet studies (Kasting et al., 1993; Selsis et al., 2007; Kopparapu et al., 2013).

Cloud-free 1D models with saturated atmospheres predict that Earth will
reach moist greenhouse conditions \((T_s = 340 \text{ K})\) when the solar constant increases by only 1.5% above its present level (Kopparapu et al., 2013). Thus our home planet may be subject to a moist greenhouse climate in a mere \(\sim 170\) million years (Gough, 1981). When considering Earth’s future, the moist greenhouse limit becomes of primary importance only if the water-loss timescale becomes shorter than the timescale for the Sun’s luminosity to trigger a runaway greenhouse. With \(T_s = 340\) K, water-loss rates are likely too slow to be of primary concern (Kopparapu et al., 2013). However, water-loss rates are proportional to the water vapor concentration in the stratosphere. Since the saturation vapor pressure increases exponentially with temperature, water-loss rates would increase rapidly as surface temperatures push past 340 K, hastening the loss of Earth’s oceans as the solar constant inches higher.

Here we re-examine Earth’s fate under the brightening Sun using a 3D climate model. We use the Community Atmosphere Model version 3 provided by the National Center for Atmospheric Research (Collins et al., 2004; Collins et al., 2006). Simulations are configured with thermodynamic ocean and sea ice models. We assume the modern land configuration, however we remove semi-permanent glacial features from Antarctica, Greenland and in high mountain regions. We have updated the native radiative transfer scheme to a correlated-\(k\) model that treats \(\text{CO}_2\), \(\text{H}_2\text{O}\) and \(\text{CH}_4\) (Wolf and Toon, 2013). Correlated-\(k\) distributions for line absorption are derived from the HITRAN 2008 database (Rothman et al., 2009) accessed via LBLRTM (Mlawer et al., 1997). The HITEMP database is not used here since the added spectral line density for water vapor is needed only if temperatures rise
above 350 K (Kopparapu et al., 2013). Continuum absorption for H₂O, CO₂, and N₂ are derived from the MT_CKD continuum model (Clough et al. 2005). Clouds are treated using bulk microphysical parameterizations for condensation, precipitation, and evaporation that control atmospheric water vapor, ice cloud condensate, and liquid cloud condensate fields (Rasch & Kristjánsson, 1998). For simplicity we have omitted O₂ and O₃ from the model. While varying ultraviolet fluxes have a significant effect on ozone heating and stratospheric chemistry, ozone feedbacks on surface temperatures are minimal (Segura et al., 2003). However, the amount of stratospheric water vapor could be sensitive to the presence of O₃. For example the current Earth has much higher stratospheric temperatures and water vapor compared with simulations shown here under the present day solar constant. All simulations assume 500 ppm of CO₂ and 10 ppm of CH₄ with a mean sea level pressure of 1.013 bar. N₂ is the broadening gas.

6.2. Implications for Earth

Our baseline simulation has a solar constant of 1367.0 W m⁻², yielding a global mean surface temperature of 289.5 K. We then incrementally increase the solar constant. The global mean solar forcing can be approximated as \( \Delta S(1-A)/4 \), where \( \Delta S \) is the change in the solar constant and \( A \) is the top-of-atmosphere albedo from our baseline simulation (0.354). A one percent increase in the solar constant then equals a radiative forcing of +2.22 W m⁻². Thus a 2% increase in the solar constant is approximately equivalent to doubling CO₂ (Hansen et al., 2005).

With a 15.5% increase in the solar constant (+34.2 W m⁻² solar forcing) we find
that the global mean surface temperature stabilizes at 312.9 K. Annual mean
tropical surface temperatures (23° S < φ < 23 °N) reach 318.5 K while annual mean
polar surface temperatures (φ ≥ 66°) reach 296.5 K (Figure 6.1a). Our results
predict that stratospheric water vapor mixing ratios will remain near 10⁻⁶, three
orders of magnitude below that needed to initiate significant water loss to space
(Figure 6.2b). While such a hot climate would undoubtedly provide great challenges
for humanity, Earth will remain safe from both water-loss and thermal runaway
limits to habitability even for a 15.5% increase in solar constant.

Numerical instabilities emerge in the convection and vertical diffusion
schemes when the solar constant is increased by 16%, limiting the range of our
study, however this is not indicative of a tipping point to a thermal runaway. While
the climate sensitivity (the change in mean surface temperature per unit forcing)
 begins to steepen for increases to the solar constant beyond 10% (Figure 6.1b),
further increases to the solar constant appear possible without endangering the
habitability of Earth. The relatively steep upturn in climate sensitivity found beyond
a 14% increase in solar constant is concurrent both with rapid increases to the
water vapor column and also with an inflection in the cloud water column (Figure
6.3a,c). At the inflection, the water vapor greenhouse continues to increase in
strength while cloud forcings weaken (less negative) allowing even more sunlight to
reach the surface (Figure 6.3b,d). There is a competition between water in the
vapor and condensate phases. As climate warms, the saturation vapor pressure
increases exponentially. If temperatures get too warm, water vapor may not readily
condense to form clouds, thus favoring a steam atmosphere over a cloudy
Figure 6.1. **Surface temperatures and climate sensitivity versus increasing solar constant.** (a) Annual mean surface temperatures versus increase in solar constant above the present value. The tropical mean surface temperatures are averaged between 23° north and south latitude. The polar mean surface temperatures are averaged for latitudes greater than 66°. (b) Climate sensitivity versus increase in solar constant. Diamonds indicate results from simulations.
Figure 6.2. Global mean vertical profiles of temperature, water vapor mixing ratio and relative humidity for increasingly hot climates. In all panels the dark blue line is from our baseline simulation, the coolest simulation ($T_s = 289.5$ K) with the solar constant equal to present day. The dark orange line is from our hottest simulation ($T_s = 312.9$ K) found with a 15.5% increase in the solar constant. (a) Global and annual mean temperature profiles for all simulations. (b) Global and annual mean water vapor mixing ratio profiles. (c) Global and annual mean relative humidity profiles.
atmosphere. Results here imply that for hot climates, there may be a state transition where upon clouds begin to dissipate. A similar feature has recently been proposed to explain enhanced climate sensitivities for hot paleoclimates (Caballero & Huber, 2013). Clouds remain a considerable uncertainty in modern climate models and more work is needed to determine how clouds may behave in hot climates.

Our results contrast with estimates from recent 1D modeling studies (Kopparapu et al., 2013). It will take ~1.5 billion years before the solar constant increases by 15.5%. Thus at an absolute minimum, our results indicate an increase in the lifetime of Earth against a thermal runaway by a factor of 3 and an increase in the lifetime against moist greenhouse conditions by a factor of 10 compared with recently published 1D results.

The 3D representation of atmospheric dynamics and their effect on the hydrologic cycle is chiefly responsible for extending the habitability of Earth compared with 1D models. Large-scale dynamics yield subsiding air over the subtropics, creating dry (subsaturated) columns that efficiently radiate surface energy to space compared with moist equatorial regions (Pierrehumbert, 1995). This excess energy radiation to space is evident in the zonal mean outgoing longwave fluxes which peak near 30° latitude while being lower over the equator (Figure 6.4d). Heat is ultimately transported from the equator to cold polar regions, homogenizing zonal mean temperatures and corresponding outgoing longwave fluxes (Figure 6.4a,d). While the tropics may remain in a state of local runaway, a planet-wide runaway cannot be reached until global mean values reach the radiation limit (Pierrehumbert, 1995; Ishiwatari et al., 2002; Goldblatt et al.,
Figure 6.3. Water vapor column, cloud water column, clear-sky greenhouse forcing and cloud forcing versus increasing solar constant. (a) Global and annual mean water vapor column, (b) clearsky greenhouse forcing, (c) cloud water column, and (d) cloud radiative forcing versus increase in solar constant.
Figure 6.4. Zonal mean surface temperature, cloud forcing, absorbed solar energy, and outgoing longwave energy for increasingly hot climates. Figure 4: Annual and zonal mean (a) surface temperature, (b) cloud net radiative forcing, (c) net absorbed solar flux and (d) outgoing longwave flux. 282 W m\(^{-2}\) is the theoretical emitted-radiation limit deduced by Goldblatt et al. (2013) for pure water vapor atmospheres. In all panels the dark blue line is from our baseline simulation ($T_s = 289.5$ K) with the solar constant equal to present day while the dark orange line is from our hottest simulation ($T_s = 312.9$ K) with a 15.5% increase in the solar constant.
There is an inherent limit to how much radiation can be emitted from a hot, moist (saturated) atmosphere (282 W m\(^{-2}\) for a pure water vapor atmosphere, Goldblatt et al., 2013). If the absorbed solar energy is above this limit a thermal runaway should occur (Figure 6.4c). However, in a subsaturated atmosphere absorbed solar and outgoing longwave radiation can exceed this limit locally, without triggering a runaway. Here the atmosphere remains subsaturated in all simulations (Figure 6.2c). Global mean relative humidities are near 75% at the surface and 50% above 700 mb. One-dimensional studies typically assume fully saturated atmospheres and thus they artificially amplify the strength of the water vapor greenhouse (Kasting, 1988; Kasting et al., 1993; Goldblatt et al., 2013; Kopparapu et al., 2013).

Clouds also provide a cooling influence on climate, a feature lacking from many 1D studies. For our baseline simulation, the contribution of clouds to the planetary albedo is 0.179. Up to the inflection near a 14% increase in solar constant, global mean cloud forcings become stronger (more negative) (Figure 6.3d). Cloud forcings preferentially increase in magnitude (more negative) over the equator and thus clouds may provide an effective mechanism for regulating tropical temperatures as climate warms (Ramanathan & Collins, 1991; Figure 6.4b). However, if clouds were artificially removed from our baseline simulation, the planet would gain a net global mean radiative forcing of +23.4 W m\(^{-2}\). If the planet were forced to artificially maintain a saturated atmosphere everywhere (without changing the cloud structure), our baseline simulation would increase its global mean greenhouse effect by +17.6 W m\(^{-2}\), owing to a ~33% increase in the global
mean water column. While such test cases are physically unrealistic, they
demonstrate that both the presence of clouds and subsaturated columns impart a
sizable cooling influence on the Earth. These effects are driven by dynamics, thus
3D simulations are needed to capture these effects. One-dimensional studies of hot
climes err in that they are approximating the planet as a single column, thus
artificially eliminating natural dynamical processes that slow global mean warming.

6.3. Implications for other planets

Our results pose no contradiction for Venus having achieved a runaway state
long ago. Venus orbits at a distance of 0.723 AU and thus today it receives 2615.1 W
m⁻², a ∼91% increase compared to Earth’s present insolation. Neglecting any orbital
migrations, even in its earliest history (circa 3.8 Ga) when the Sun was only 75%
percent as bright as today, Venus would have received 1961.3 W m⁻² of solar
radiation. This is equivalent to a ∼43% percent increase in the present day solar
energy received by Earth. While stable climates are found here with a 15.5%
increase in the solar constant, the steepening of the climate sensitivity curve (Figure
6.1b) leaves little hope that habitability could be maintained with a 43% increase in
the solar constant. Any habitable period on Venus was probably tantalizingly brief.

Our results also have implications for the study of exoplanet habitability. For
an Earth-like exoplanet, having an atmosphere in constant contact with oceans, our
results indicate that the inner edge of the habitable zone can be moved closer in
towards the parent star compared with results from 1D cloud-free models. A
similar result is also found from 3D climate models of exoplanets in spin-orbital
resonance with M dwarf stars (Yang et al., 2013). The results of Kopparapu et al. (2013) place the present day Earth precariously close to the inner edge of habitability. They suggest with the present solar luminosity, if Earth were placed at an orbit of 0.97 AU a complete thermal runaway would occur while with an orbit at 0.99 AU a moist greenhouse climate would occur. In this study, if the Earth-Sun distance decreased to 0.93 AU, the mean surface temperature would still be 27 K below that of a moist greenhouse climate and 60 K below the boiling point of water.

6.4. Summary

Ideally one would like to simulate a true thermal runaway with a 3D climate model. However at present off-the-shelf general circulation models are not up to the task. Cloud physics, convection and vertical diffusion (and likely other parameterizations) will need careful examination and modification to increase their robustness and appropriateness for hot climates. However, even in a limited study such as this, some important concepts come to light. Clouds, subsaturation, and equator to pole heat transport all help delay the onset of moist and runaway greenhouse climates. Three-dimensional dynamical modeling is critical for determining realistic cloud, water vapor, and energy distributions, all of which may strongly modulate hot climates. Our preliminary 3D results imply that Earth may remain habitable for far longer than is indicated from 1D simulations.
Chapter VII

Concluding Remarks

The faint young Sun paradox has lingered, stubbornly resisting resolution for over 4 decades since it was first recognized by Sagan and Mullen (1972). Geological evidence clearly indicates that the early Earth was as at least as warm as today (Knauth and Lowe, 2003; Robert and Chaussidon, 2006; Hren et al. 2009; Blake et al. 2010). However, the well-established standard solar model predicts that our Sun was up to 25% dimmer in the ancient past (Gough, 1981). This implies an apparent contradiction. How did the early Earth manage to stay warm despite such weak solar irradiance? In this work we have presented a comprehensive study of the faint young Sun paradox and the climate of Earth’s Archean period (3.8 to 2.5 Ga) using a sophisticated general circulation model. We used a modified version of the Community Atmosphere Model version 3 from the National Center for Atmospheric Research to study early climate (Collins et al., 2004). Refer to Chapter 2 for a detailed description of the model.

A primary result of our work is that the faint young Sun paradox no longer appears quite so problematic. For the late Archean (2.8 Ga, 80% solar constant), modest amounts of carbon dioxide and methane may provide sufficient warming (see Chapter 3, also Wolf and Toon, 2013). A combination of 0.015 bar of CO₂ and 0.001 bar of CH₄ in a 1 bar atmosphere can offset a 20% reduction in the solar
constant yielding global mean surface temperature of 285 K and mean tropical surface temperatures of 296 K, only a few degrees cooler than the present day. The global and annual mean sea ice margin stabilizes at 68° latitude and thus ~93% of the surface would remain free from ice. In this case photochemical hazes would not form since the ratio of CH$_4$ to CO$_2$ is less than 0.1. A CO$_2$ partial pressure of 0.015 bar agrees with the “best guess” estimate of Driese et al. (2011) for the late Archean. However, note that the upper limit on atmospheric CO$_2$ for the late Archean may be as high as 0.025 bar as indicated by Sheldon (2006). Thus warmer climates still are possible via conventional means of warming (i.e. greenhouse gases).

Other fundamental changes to the Archean climate system may result in even further warming. Recent literature has suggested that reductions to the Archean surface albedo and cloud condensation nuclei (Rosing et al., 2010), along with increases to the total surface pressure via increased N$_2$ (Goldblatt et al. 2009), may help warm the early Earth. Darker land surface area; cloud condensation nuclei sourced from only sea salts and dust; and increased atmospheric N$_2$ all may yield modest global mean warming. For reasonable assumptions, the global mean surface temperature may increase by 3 to 7 K for each mechanism respectively (see Chapter 4, also Wolf and Toon, 2014b). However, while one can argue convincingly for the validity of the discussed warming mechanisms, they remain difficult to explicitly prove without better geologic data from the time period. Note that the effect an increased rotation rate on climate is slight, and does not appear to play a significant role in overcoming the faint young Sun.
While changes to the surface albedo, clouds, and atmospheric nitrogen cannot provide a singular means of rectifying the faint young Sun paradox, optimal warming solutions that incorporate plausible changes to each along with modest CO\textsubscript{2} and CH\textsubscript{4} can resolve the faint young Sun paradox for all times of the Archean without violating constraints on greenhouse gases (see Chapter 5). Conversely, solutions that rely only on a CO\textsubscript{2} greenhouse, with no CH\textsubscript{4} or additional warming mechanisms, cannot keep the early Earth warm within proposed constraints at any time during the Archean. With double the present day level of atmospheric N\textsubscript{2}, a dark land surface albedo, 20% of the present day cloud condensation nuclei, and 10^{-4} bar of CH\textsubscript{4}, only 0.0024 bar of CO\textsubscript{2}, about 6 times the present level, is needed to maintain present day surface temperatures (~288 K on global mean) at the end of the Archean (2.5 Ga, 82% solar constant). Note this is a factor of 7 less CO\textsubscript{2} than is predicted by Dreise \textit{et al.} (2011) and a factor of 10 below the upper limit predicted by Sheldon (2006) for the late Archean. Even if we are limited to only the modern atmospheric N\textsubscript{2} inventory (as suggested by Marty \textit{et al.}, 2013), still only 0.0048 bar of CO\textsubscript{2}, about 12 PAL, is required at 2.5 Ga to maintain present day surface temperatures.

At the beginning of the Archean (3.8 Ga, 75% solar constant) with 2 PAL N\textsubscript{2} and the above discussed plausible changes to land, clouds, and nitrogen, present day global mean surface temperatures can be maintained with only 0.0147 bar of CO\textsubscript{2}, about 37 PAL. This again remains within the bounds on paleoatmospheric CO\textsubscript{2} defined by Dreise \textit{et al.} (2011) and Sheldon (2006). However, with only 1 PAL N\textsubscript{2} in the early Archean, 0.032 bar of CO\textsubscript{2}, about 80 PAL, is needed to reach mean surface
temperatures of 288 K at 3.8 Ga, marginally exceeding the upper most limits on CO₂ derived from paleosols. However, CO₂ constraints are only defined for the later part of the Archean and are absent from earlier times. It is generally believed that atmospheric CO₂ has been drawn down slowly over time, thus the early Archean very well could have had larger concentrations of CO₂ than are expected for the late Archean. Climates hotter than 288 K could feasibly be maintained by maximizing the allowable CO₂ and CH₄ along with plausible warming mechanisms mentioned above.

We also find a range of solutions to a weaker form of the faint young Sun paradox. While it is generally believed that the entirety of the Archean was hot and ice free, this may not have been the case. The high seawater temperature interpretation of the isotopic composition of Archean cherts is not universally accepted (Kasting et al., 2006; Jaffres et al., 2007; Shields and Kasting, 2007). More recent studies place Archean seawater temperatures inline with present day tropical ocean temperatures (Hren et al., 2009; Blake et al., 2010). Furthermore, the Archean geologic record is spatially and temporally sparse and the paleolatitudes of geological samples are unknown (Feulner, 2012). High crustal reworking rates imply that very little unaltered material from this time period remains left today (Dhuime et al., 2012). Typical meridional gradients of temperature may imply a warm tropical zone while the poles remained cool. A solution to a weak form of the faint young Sun paradox may only require that liquid water and thus habitable conditions were present on some part of the planet surface.
Here, climates with relatively weak greenhouses can still maintain large swaths of open-ocean during the late Archean. Even with only 0.005 bar of CO$_2$ and no CH$_4$, open-ocean fractions of greater than 50% can be maintained circa 2.8 Ga despite global mean surface temperatures of only \( \sim 260 \) K, thus ensuring that a large part of the young Earth remained habitable (see Chapter 3). A runaway glaciation may be avoided even down to much lower atmospheric CO$_2$. Stable waterbelt climates are possible (Abbot et al. 2011). However while we argue that the limitations of the Archean geologic record make it inappropriate to preclude climates with relatively larger polar ice caps compared with the present day, waterbelt climate states where the surface experiences glaciation covering 80% or more of the surface may stretch the believable limits for a cold Archean climate solution.

We have also examined the properties of photochemical hazes that may have enshrouded the early Earth. After the rise of methanogens, CH$_4$ likely became a significant part of the early atmosphere. Modeling and laboratory studies infer that if the ratio of atmospheric CH$_4$ to CO$_2$ rises above about 0.1, Titan-like hydrocarbon hazes would have formed high in the atmosphere of the early Earth (Trainer et al., 2006; DeWitt et al., 2009; Zerkle et al., 2012). In chapter 5 we demonstrated that it is critically important to accurately account for the fractal aggregate morphology of such hazes. A haze layer composed of fractal aggregate hydrocarbon particles can make a highly effective ultraviolet shield, with an optical depth in the UV an order of magnitude or more greater than that in the visible. Such a shield could have protected NH$_3$ from photolysis and protected early surface dwelling life from harsh
radiation. Life, methane, and climate may have had a complex connection during the Archean. Future studies will link hazes with a reducing photochemical model to our general circulation model.

The evolution of the solar constant over time has dominated our view of early climate and served as the primary motivator of this work. While a weak Sun poses challenges to habitability on the early Earth, the future evolution of the Sun may pose equally great challenges. The brightening of our Sun will not stop. The Sun will continue to increase in brightness by about 1% every ~110 million years (Gough, 1981) and will eventually threaten the habitability of the Earth. However, in chapter 6 we show that Earth will maintain habitable conditions for at least another 1.5 billion years and probably for much longer. When the Sun increases in brightness by 15.5%, global mean surface temperatures will increase to 312.9 K, a gain of +23.4 K from our control simulation. While such a climate would certainly pose grave challenges for humanity, the climate would remain safe from moist and runaway greenhouse states. Water in its liquid form would still dominate the planet’s surface. Work will continue in this area of study.

Model inter-comparison remains an important tool for gaining confidence in our theoretical results. Recently results from general circulation models of differing origin indicate that sufficiently warming the early Earth does not now appear to be a significant problem (Wolf and Toon 2013; Charnay et al. 2013; Kunze et al. 2013). Plausible greenhouse gas concentrations along with realistic changes to land albedos, clouds, and nitrogen can yield warm climates for the duration of the Archean (Charnay et al., 2013; Wolf and Toon, 2014b). Numerous permutations of
factors may suffice. However, the precise combination of solar constant, surface temperature, CO₂, CH₄, N₂ and other effects may remain difficult to pin down precisely without better geological constraints.
References


Briegleb, B.P., Bitz, C.M., Hunke, E.C., Lipscomb, W.H., Holland, M.N., Schramm, J.L.,


Hofmann, A. “The geochemistry of sedimentary rocks from the Fig Tree Group, Baerberton greenstone belt: Implications for tectonic, hydrothermal and surface processes during mid-Archaean times.” *Precambr. Res.* 143, 23–49 (2005)


Mandlebrot, B.B. “Fractals: Form, Chance and Dimension.” Freeman, San Francisco, CA (1977)


Rannou, P., McKay, C.P., Botet, R., and Cabane, M. “Semi-empirical model of


