

Low simulated radiation limit for runaway greenhouse climates

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The atmospheres of terrestrial planets are expected to be in long-term radiation balance: an increase in the absorption of solar radiation warms the surface and troposphere, which leads to a matching increase in the emission of thermal radiation. Warming a wet planet such as Earth would make the atmosphere moist and optically thick such that only thermal radiation emitted from the upper troposphere can escape to space. Hence, for a hot moist atmosphere, there is an upper limit on the thermal emission that is unrelated to surface temperature. If the solar radiation absorbed exceeds this limit, the planet will heat uncontrollably and the entire ocean will evaporate—the so-called runaway greenhouse. Here we model the solar and thermal radiative transfer in incipient and complete runaway greenhouse atmospheres at line-by-line spectral resolution using a modern spectral database. We find a thermal radiation limit of 282 W m^{-2} (lower than previously reported) and that 294 W m^{-2} of solar radiation is absorbed (higher than previously reported). Therefore, a steam atmosphere induced by such a runaway greenhouse may be a stable state for a planet receiving a similar amount of solar radiation as Earth today. Avoiding a runaway greenhouse on Earth requires that the atmosphere is subsaturated with water, and that the albedo effect of clouds exceeds their greenhouse effect. A runaway greenhouse could in theory be triggered by increased greenhouse forcing, but anthropogenic emissions are probably insufficient.

Earth's surface and troposphere are heated by the Sun. If more sunlight was absorbed, these would warm and, consequently, emit more thermal radiation to space. While thermal radiation from the surface can escape the atmosphere directly, energy balance and temperate climate are maintained. Most of such emission occurs through the water vapour window, the 8–13 μm local minimum in water vapour absorption, coincident with the Wein peak in surface thermal emission at 10 μm .

Warming an Earth-like planet would make the atmosphere moist and hence optically thick across the thermal region, even in the water vapour window. Then, only the upper troposphere (not the surface or lower troposphere) would be able emit radiation directly to space. As sunlight penetrates the atmosphere better than thermal infrared radiation, the upper troposphere is heated from below and its thermal structure is determined by moist convection. The water vapour mixing ratio increases with temperature, causing the moist adiabatic lapse rate to tend towards the saturation vapour pressure curve and the tropopause to acquire a fixed temperature–pressure structure. Thus, the level from which effective thermal emission occurs tends to a fixed temperature and the flux a fixed value. This maximum in outgoing radiation—or radiation limit—means that surface warming no longer leads to more thermal emission. If the net absorption of solar radiation exceeds this limit, then surface temperatures will increase in a runaway greenhouse, evaporating the entire ocean and sterilizing the planet en route.

This radiation limit was first found in a model by Simpson in 1927¹, but seen as a paradox. The physics, as described above, was subsequently elucidated by Nakajima *et al.*², so we refer to the Simpson–Nakajima limit. In the intervening time, the notion of a runaway greenhouse was identified by reference to a separate stratospheric radiation limit, the Komabayashi–Ingersoll

limit^{3,4} (which is unlikely to be reached in practice²). These various radiation limits have recently been reviewed⁵. Numerical calculations performed a few decades ago by several groups^{6–9} converged on a consensus estimate of 310 W m^{-2} for the Simpson–Nakajima limit⁹. Subsequent work addressed some aspects of the problem (for example, the hydrological cycle¹⁰, pedagogic treatment of thermal limits¹¹, and early Mars¹²), but there has been no full revision of the classic calculations.

Here, we present the most complete study of the runaway greenhouse for 25 years, across the full range of temperatures, using modern input spectroscopic data and a line-by-line treatment of the solar and thermal radiation (see the Methods). We limit ourselves to clear-sky (cloud-free) calculations, which embody the first-order physics of the problem. Clouds both reflect solar radiation (making the runaway less likely) and enhance the greenhouse (making it more likely). Omitting them yields hard upper bounds on both solar absorption and thermal emission. This is a robust place to begin a re-evaluation of the problem, with thought experiments on how clouds will modify the results.

The runaway greenhouse has contemporary relevance. There has been high-profile speculation that extreme anthropogenic global change could trigger it¹³, but this is contrary to existing theory^{2,5} and numerical results¹⁴. It also sets the inner boundary of the circumstellar habitable zone^{15,16} in which the Kepler mission is identifying planetary candidates at present¹⁷.

Pure water atmospheres

We begin with the absolute endmember case of a pure water atmosphere: neither any background gas nor any greenhouse gas other than water. With increasing surface temperature, evaporation from the ocean adds mass to the bottom of the atmosphere, so this

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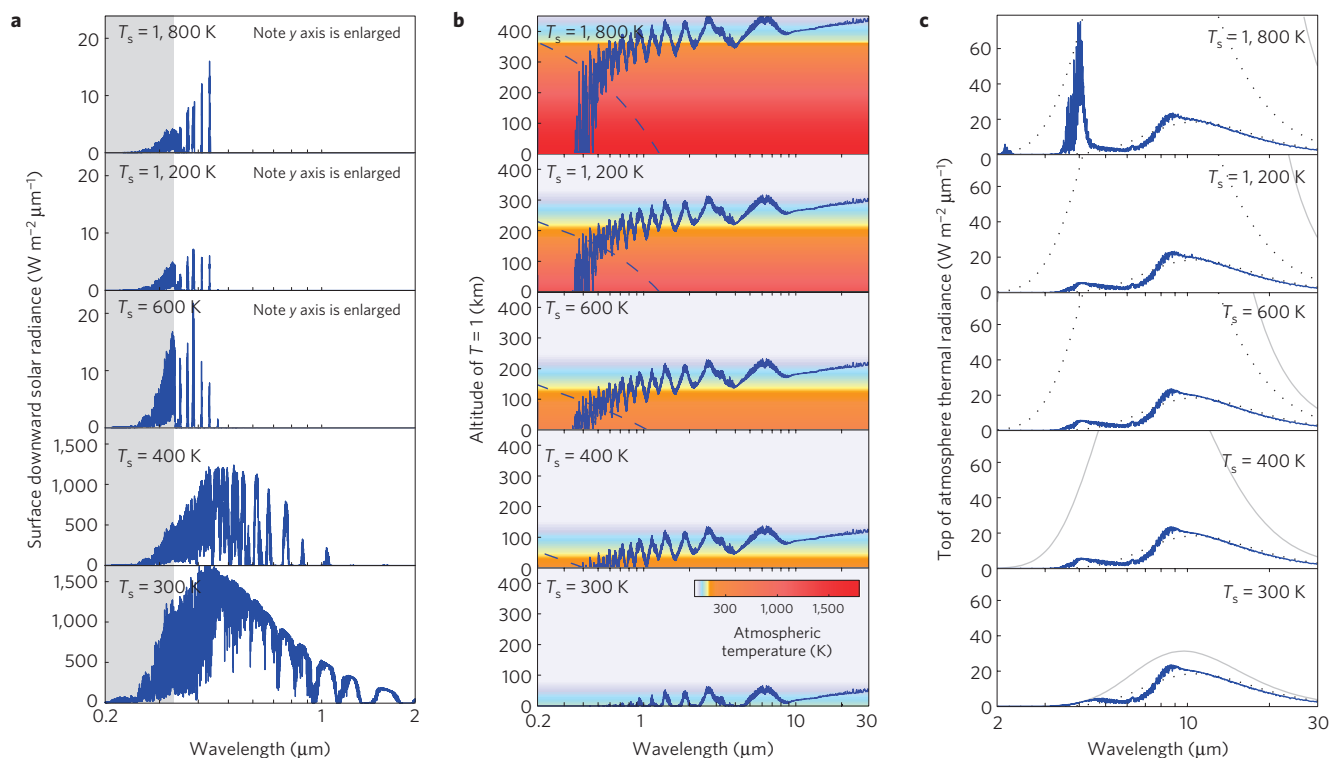


Figure 1 | Spectra and effective absorption and emission levels in a pure water atmosphere. T_s is surface temperature. **a**, Spectrum of downward solar flux at the surface. No line data are available for the grey shaded area. **b**, Altitude at which optical depth is unity. Solid line is absorption optical depth and dashed line is Rayleigh scattering optical depth. Background shading is atmospheric temperature. **c**, Spectrum of outgoing thermal flux at the top of the atmosphere. Solid lines are the black body flux at the surface temperature. Dotted lines are the black-body flux for 280 and 400 K for comparison. See also Supplementary Fig. S2.

is a good approximation for a hot atmosphere in which the water vapour mixing ratio asymptotes to one.

The optical depth of the atmosphere, τ_λ (a function of wavelength), is measured downward from the top. Effective emission to space and attenuation of sunlight occur where $\tau_\lambda \sim 1$ (given Beer's law; $I_1 = I_0 \exp^{-\tau_\lambda}$, where I_0 and I_1 are the incident and transmitted radiance), so plotting the altitude of $\tau_\lambda = 1$ shows where emission to space and absorption of sunlight dominantly occur (Fig. 1).

For thermal emission, $\tau_\lambda = 1$ is either near the surface or not reached for low surface temperatures (T_s), but rises towards high altitudes as the planet warms. While $T_s \lesssim 1,600$ K, the temperature of $\tau_\lambda = 1$ remains between 250 and 300 K, so the top of atmosphere thermal spectrum is bounded by Planck functions for temperatures of 250 and 300 K, and is independent of T_s . Thus, the Simpson–Nakajima radiation limit emerges as 282 W m^{-2} (Fig. 2), lower than previous estimates (for example, 310 W m^{-2} , ref. 9). When $T_s \gtrsim 1,600$ K the upper atmosphere temperature gradient is sufficiently steep that the temperature reaches 400 K at $\tau_\lambda = 1$ in the $4 \mu\text{m}$ water vapour window (Fig. 1c). Hence, a new peak in thermal radiation emerges, sufficient to permit a new stable climate with a steam atmosphere. No surface radiation escapes directly to space. Observation of this emission peak in a (exo)planetary atmosphere would indicate that the planet is in a runaway greenhouse state.

Earth's atmosphere is largely transparent to solar radiation. However, for water-rich atmospheres, increasing temperatures are accompanied by increases in atmosphere pressure and water vapour absorption. The additional pressure increases the Rayleigh scattering optical depths at shorter wavelengths, whereas near-infrared water vapour vibration rotation bands increase the absorption optical depth at longer solar wavelengths. Both processes attenuate sunlight, very little of which reaches the surface, so surface albedo no longer affects the radiation budget. In the limiting case,

for a pure water atmosphere without clouds and the present solar flux, a maximum of 294 W m^{-2} is absorbed, much higher than the previous estimate⁹ of 222 W m^{-2} .

Given a hot, moist and cloud-free atmosphere, the net absorption of sunlight would slightly exceed the thermal radiation limit. This implies that a cloudless runaway greenhouse, steam atmosphere, would be stable under the present insolation. Earth today has a stable temperate climate (the requirements for which are discussed in the next section) implying a climate bistability with respect to the runaway greenhouse (previously seen in a grey atmosphere model¹⁸). Both the solar and thermal calculations represent upper bounds for a pure water atmosphere—clouds could reduce either, moving the bifurcation point.

Previous work suggested that the thermal radiation limit does not depend on the presence of non-condensable greenhouse gases^{2,9}. This is not strictly correct. The radiation limit depends on the minimum absorption cross-section in the $10 \mu\text{m}$ water vapour window; any additional opacity here would raise the $\tau_\lambda = 1$ surface to a higher altitude that radiates at a lower temperature. As purely theoretical tests, we set 1% each of our atmospheres to be carbon dioxide or ammonia, then 1% of both. The radiation limit decreased by 2 W m^{-2} for 1% CO_2 , 6 W m^{-2} for 1% NH_3 and 8 W m^{-2} for both (Supplementary Fig. S7). Ammonia is one of the strongest absorbers around $10 \mu\text{m}$, so deeper reductions to the radiation limit seem unlikely.

These results are sensitive to the absorption cross-sections used. First, using the most detailed spectral line list for water (we use HITEMP2010 (ref. 19)) and correct Rayleigh scattering cross-sections for water are essential. Using a less comprehensive line list (for example, HITRAN 2008), or Rayleigh scattering for air instead of water, gives erroneous results (Supplementary Figs S3–S6). Relative to previous results, our lower thermal emission and

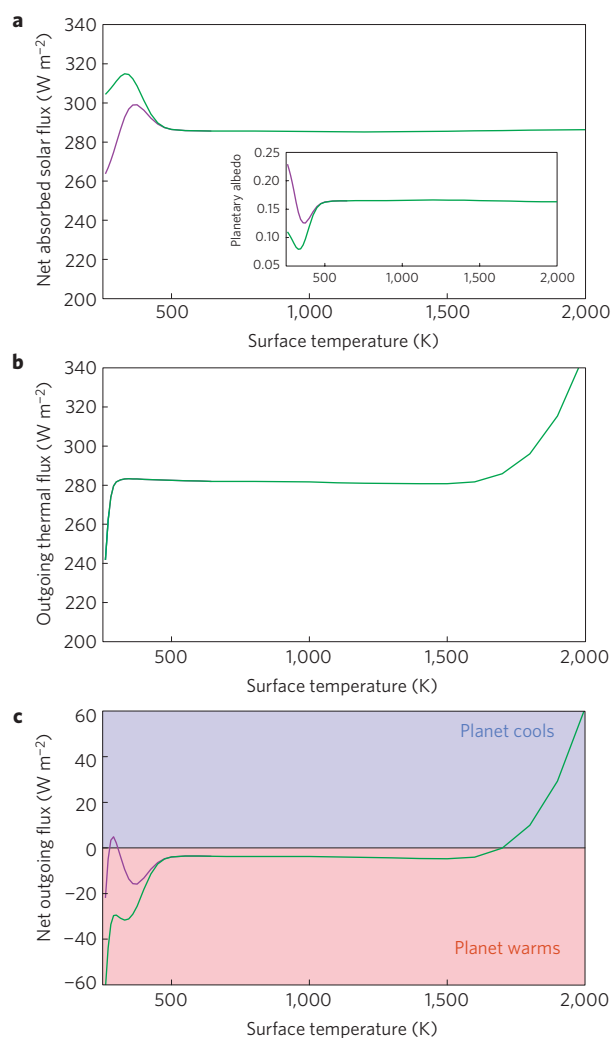


Figure 2 | Top of atmosphere fluxes from a pure water atmosphere. **a**, solar; **b**, thermal; and **c**, net flux. Colours identify different surface albedos: green for 12% and purple for 25%. Steady-state climates are found where the net outgoing flux is zero; stable steady states are where the flux is increasing with increasing temperature as it passes through zero, and unstable steady states are where it decreases with increasing temperature as it passes through zero.

higher solar absorption are due to these absorption coefficient changes and increased spectral resolution. Second, the strength of the water vapour continuum (the smoothly varying absorption in the window regions) is very important. In the infrared, the continuum we use is weaker than indicated by the most recent data²⁰ (Supplementary Fig. S9), so our estimates are conservative and the Simpson–Nakajima limit is probably slightly lower than our estimate. The uncertainty wedge associated with the continuum grows towards shorter wavelengths, and there are no measurements in the visible region. Our (or any other) solar calculations must be regarded as provisional until such measurements are made.

Transition to a runaway greenhouse

Given that a cloud-free steam atmosphere seems to be a stable state at the present solar constant, one should examine both how the stable temperate climate is maintained on Earth and the conditions that would lead to a runaway greenhouse. Hence, we examined transitional atmospheres (up to 400 K) with the same mass of background gas as Earth and various greenhouse gas inventories (Table 1).

Table 1 | Greenhouse gas inventory scenarios used in transitional atmospheres.

Scenario	CO ₂ (ppmv)	CH ₄ (ppmv)	N ₂ O (ppbv)
Baseline	0	0	0
Pre-industrial	287	0.806	275
RCP 8.5, year 2100	936	3.75	435
Extreme anthropogenic	3,000	10	500
Arbitrarily high	30,000	10	500

Pre-industrial³⁹ and RCP 8.5 for year 2100⁴⁰ are standard. Extreme anthropogenic assumes that the large fossil fuel reservoirs (coal and some methane clathrate) are burned rapidly—that is, it is extreme, but certainly possible. Note that concentrations are relative to a standard, 10⁵ Pa atmosphere and the mass of each greenhouse gas is fixed as temperature and surface pressure increase, so actual mixing ratios decrease as temperature increases.

At 280 K, the surface emits directly to space through the water vapour window (Fig. 3). For surface temperatures above 310 K the temperature of the emitting level remains between 250 and 300 K, regardless of the surface temperature. If greenhouse gases other than water are more abundant, $\tau_1 = 1$ is higher in the absorption bands of these gases and less radiation is emitted overall. However, the relative magnitude of this effect decreases in hotter atmospheres with more water. In flux terms (Fig. 4), for the endmember case of a saturated, cloud-free atmosphere with contemporary surface albedo, the net absorbed solar radiation exceeds thermal emission in all scenarios except that with no greenhouse gases other than water, implying that a runaway greenhouse should occur. As this has manifestly not happened to Earth, we are led to the conclusion that a combination of atmospheric subsaturation and an excess of cloud albedo forcing over cloud greenhouse forcing prevents a runaway greenhouse on Earth today.

First, we relax the assumption of saturation. Our nominal relative humidity profile (Methods) yields a stable climate only for the case of no additional greenhouse gases, although a marginally stable result was obtained for pre-industrial greenhouse gas concentrations. The assumed relative humidity profile is a source of uncertainty in one-dimensional (1D) models. With convection parameterization in a 1D model, a nonlinear transition from subsaturation to saturation around 310 K was found¹⁰, introducing an additional bistability in climate that is not found with climatological relative humidity profiles such as ours¹⁰. In Earth's tropics, columns of dry air prevent a local runaway greenhouse²¹. Nonetheless, our results indicate that subsaturation alone is probably not sufficient to prevent a runaway greenhouse today (Fig. 4).

Thus, we turn our attention to clouds. Today, these give an albedo forcing of 50 W m⁻² and a greenhouse forcing of 26 W m⁻² (ref. 22). Climatological mean top of atmosphere fluxes are 239.4 W m⁻² net solar absorbed and 238.5 W m⁻² outgoing thermal²³ (the 0.9 W m⁻² discrepancy is causing global warming). High, cold, clouds have a dominant greenhouse effect (though also reflect). Low cloud has a dominant albedo effect. At first approximation, we simulate this by increasing surface albedo; doubling our 12% albedo approximately represents the low cloud forcing. Our subsaturated pre-industrial cases with present surface temperature and enhanced albedo (Fig. 4) have top of atmosphere solar and thermal fluxes of 263 and 266 W m⁻², reasonably approximating climatology minus high cloud forcing. In all of our scenarios (other than arbitrarily high CO₂), there is a stable climate under this assumption. Thus, we can infer that the excess of cloud albedo over cloud greenhouse forcing, in combination with subsaturation, permits stable temperate climate on Earth.

The clear-sky fluxes are upper bounds: clouds could reduce either flux, making the runaway more or less likely. Previously⁹ it was argued that cloud reflection would dominate over cloud

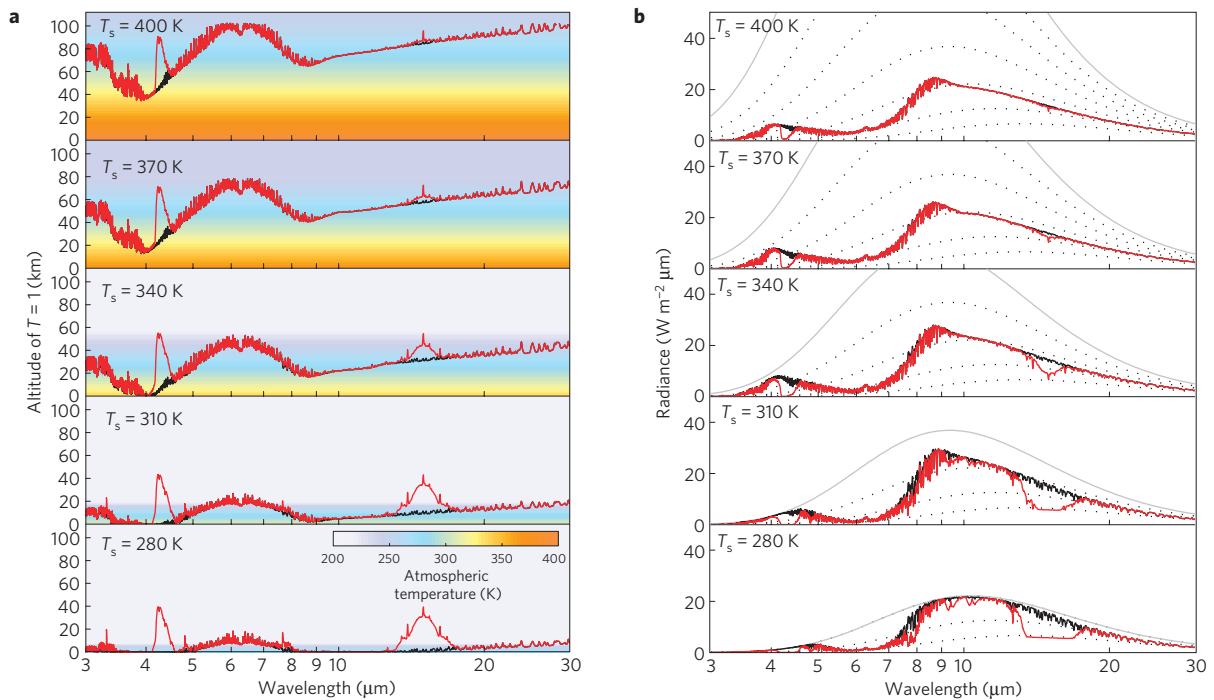


Figure 3 | Spectra of thermal emission level and outgoing thermal radiation for transitional atmospheres. Black is baseline case; red is with 5,000 ppmv CO₂. **a**, Altitude where optical depth is unity. Most outgoing thermal radiation is emitted from this level. Background colour is atmospheric temperature. **b**, Top of atmosphere emission spectra. Solid grey lines are the surface emission for each case. Dotted grey lines are for Planck functions of 220, 250, 280, 310, 340 and 370 K up to the surface temperature. The broad 5–8 μm absorption feature is from water and the smaller absorption features at 4.2 μm and 15 μm are from carbon dioxide.

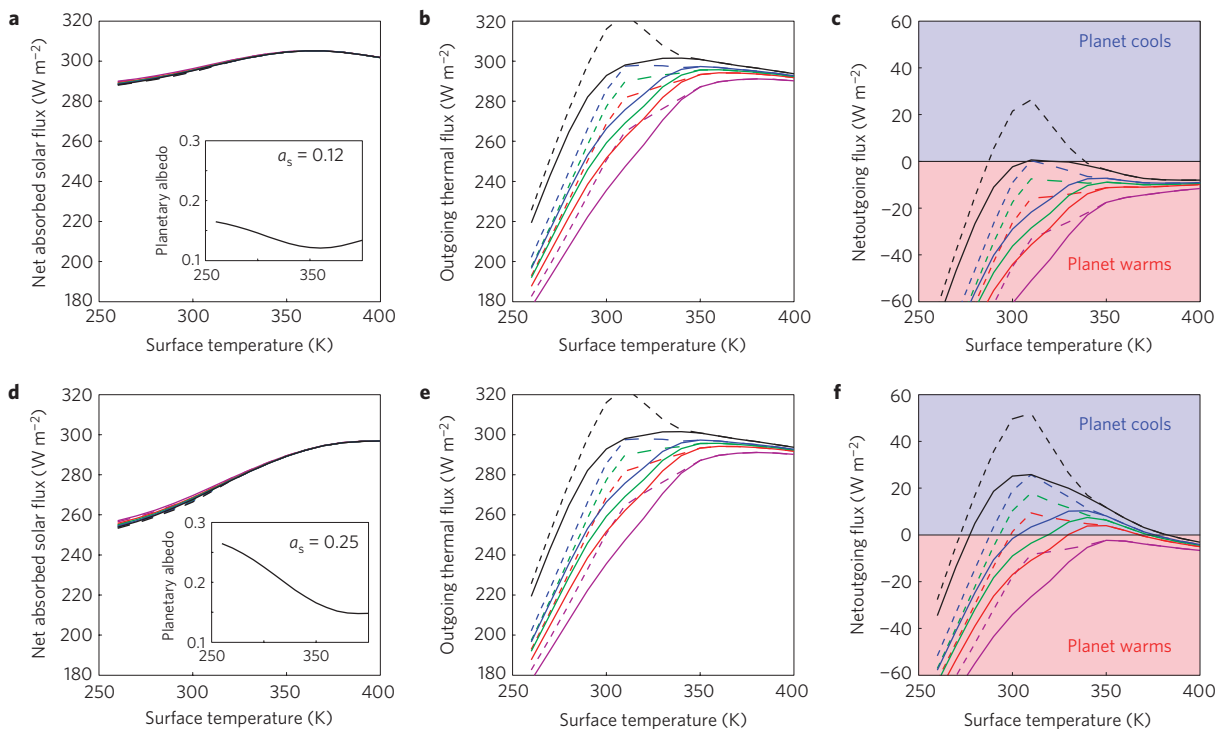


Figure 4 | Top of atmosphere fluxes from transitional atmospheres. **a–c**, With a surface albedo of 12%. **d–f**, With a surface albedo of 25%. Colours identify the greenhouse gas inventories from Table 1 (black is baseline, blue is pre-industrial, green is RCP 8.5 at 2100, red is extreme anthropogenic and purple is arbitrarily high). Solid lines are atmospheres saturated with water vapour throughout, whereas dashed lines are subsaturated where the saturation mixing ratio is less than 5%. Steady-state climates exist where the net outgoing flux is zero, stable where flux increases with temperature and unstable where flux decreases with temperature.

greenhouse in an optically thick atmosphere. However, this misses a critical distinction based on the level of the clouds. The atmosphere is more transparent to sunlight than thermal radiation, so $\tau_{\lambda}(\text{solar}) = 1$ is at lower altitude than $\tau_{\lambda}(\text{thermal}) = 1$, so there are three categories of cloud effect based on these. Below $\tau_{\lambda}(\text{solar}) = 1$, clouds will have negligible effect. Between $\tau_{\lambda}(\text{solar}) = 1$ and $\tau_{\lambda}(\text{thermal}) = 1$, albedo will dominate. Above $\tau_{\lambda}(\text{thermal}) = 1$, cloud greenhouse will probably dominate (although reflection may dominate if the clouds are more than four times thicker than the present global mean²⁴). The largest projected increase in the water vapour mixing ratio is in the upper atmosphere (Supplementary Fig. S1), suggesting that high clouds would increase most (although this is speculative). For near-future global warming, the present best estimates are for clouds to exert a positive forcing²⁵ (that is, enhanced greenhouse dominating).

Steady-state climates exist where the net flux (thermal minus solar) is zero; stable where the net flux increases with temperature and unstable where it decreases with temperature (Fig. 4). For small greenhouse gas inventories, the outgoing thermal flux overshoots the Simpson–Nakajima limit giving a ‘hump’ of stability; excess thermal emission will give a negative feedback that will restore stable, temperate, climate. Using the subsaturated, 25% albedo runs as a reference, the hump of stability is 24 W m^{-2} for pre-industrial, 18 W m^{-2} for representative concentration pathway (RCP) 8.5 at 2100 and 8 W m^{-2} for extreme anthropogenic. With our arbitrarily high greenhouse gas scenario (30,000 ppmv CO_2), this vanishes and there is no stable temperate climate. Greenhouse gases do not simply warm the planet, but also lower or remove the energy barrier between temperate climate and a runaway greenhouse.

Other times and other planets

A runaway greenhouse has manifestly not occurred on post-Hadean Earth—it would have sterilized Earth (there is observer bias). Palaeoclimate gives us a sample of conditions where a runaway greenhouse did not occur, but cannot tell us the size of any safety margin. The so-called ‘hothouse’ climate of the Eocene is the most useful constraint for anthropogenic change. With the solar constant 1% less than today and a few thousand ppmv CO_2 , the mean temperature was $\sim 10 \text{ K}$ warmer than today²⁶. With CO_2 and temperature both higher than we expect in the foreseeable future²⁷, this implies that an anthropogenic runaway greenhouse is unlikely. Deglaciation from Neoproterozoic snowball Earth events probably required that $\sim 10\%$ of the atmosphere was carbon dioxide. The solar constant was 6% less than today, so net solar radiation absorbed would have been 12 W m^{-2} less and climate not yet bistable. By contrast, deglaciation from a snowball Earth event in the future might trigger a runaway greenhouse.

Venus probably experienced a runaway greenhouse in the past, evident now in enrichment of D/H in its atmosphere^{7,28}. Previous work suggested that early Venus was close to the threshold for a runaway greenhouse⁹—our new lower radiation limit and enhanced solar absorption imply that, given the same amount of nitrogen in the atmosphere as Earth, Venus may not have had a habitable period. However, if early Venus had at least as much nitrogen in its atmosphere as it does now this would have had a protective effect (Fig. 5). More nitrogen gives more Rayleigh scattering, decreasing absorbed solar radiation in a transitional atmosphere.

As the solar constant increases with time, Earth’s future is analogous to Venus’s past. We expect a runaway greenhouse on Earth 1.5 billion years hence if water is the only greenhouse gas, or sooner if there are others. Earth’s atmospheric nitrogen inventory has probably changed with time²⁹. Any future decrease would lessen the protective effect of Rayleigh scattering and hasten a runaway greenhouse (in contrast to previous arguments³⁰).

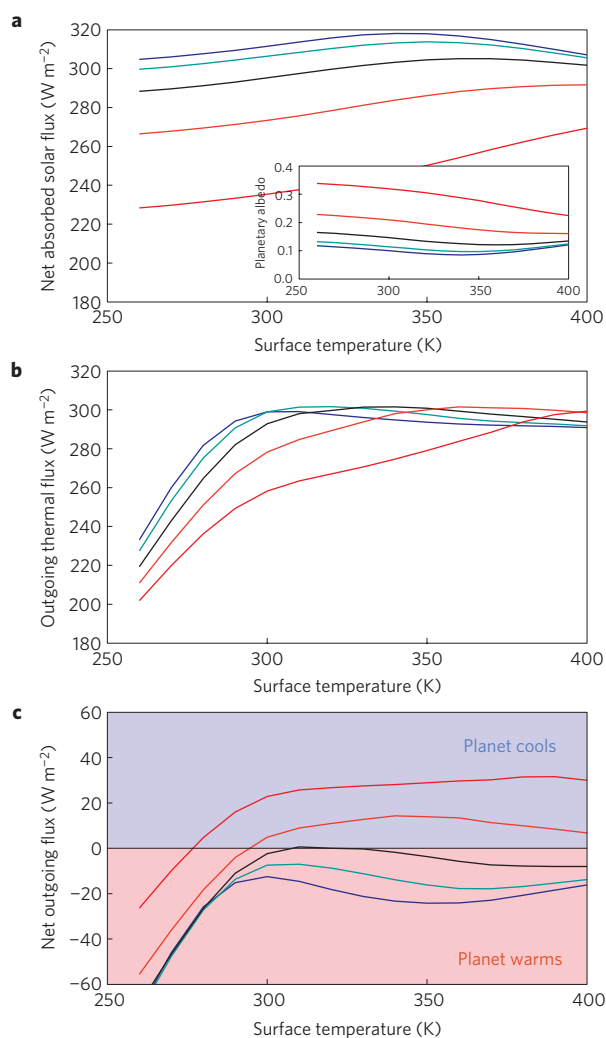


Figure 5 | Top of atmosphere fluxes from an ideal gas atmosphere with a varying amount of background gas (nitrogen). **a**, Solar; **b**, thermal; and **c**, net flux. All runs for baseline (no non-condensable greenhouse gas) with surface albedo of 12%. Colours are: blue for 0.1 bar, turquoise for 0.33 bar, black for 1 bar, orange for 3 bar and red for 10 bar.

Our pure water calculations were aimed at hot atmospheres, but should also apply to water worlds, analogous to a warm version of Jupiter’s moon Europa. In the absence of Rayleigh scattering from background gas, planetary albedo would be lower than Earth. Under Earth’s insolation, without clouds and with 12% surface albedo (the average for Earth, about twice that of sea water), there is no stable temperate climate and a runaway greenhouse would always ensue. Arbitrarily increasing surface albedo to 25% (a proxy for low cloud reflection) gives a marginally stable state at 275 K. With a mean surface temperature this low, ice albedo feedback would probably lead to low-latitude glaciation (snowball Earth-like). A transient warming sufficient to melt the ice would probably cause a transition directly to a runaway greenhouse. Unless mediated by other atmospheric constituents or clouds, there would be no stable temperate climate state.

The runaway greenhouse sets a hard limit for the inner edge of the circumstellar habitable zone^{15,16}. This classic definition neglects multiple climate equilibria so may be misleading. Three major stable climate states exist at the same solar constant: snowball Earth (at least transiently reduced habitability), temperate (habitable) or runaway greenhouse steam atmosphere (sterilizing)¹⁸. Changes to clouds today may be sufficient to transition from temperate to either

other state. Determining surface temperature requires knowledge of the atmospheric state and history: it is not possible to determine a habitability a priori from incident stellar radiation.

The thermal radiation limit depends weakly on the mass of the planet (Supplementary Fig. S8). Everything else being equal, a Mars-size planet would be more susceptible to a runaway greenhouse and a so-called super-Earth less¹¹.

Revisiting the classic planetary sciences problem of the runaway greenhouse with modern modelling tools, we have shown that the thermal radiation limit is lower and that more solar radiation is absorbed. The runaway greenhouse may be much easier to initiate than previously thought. A renewed modelling effort is needed, addressing both Earth and planetary science applications. We have begun this process with a single column, clear-sky model, which has allowed us to advance the core radiative transfer aspect of the problem. Reference calculations are available as Supplementary Information to permit future model testing. Our work should be followed with cloudy column models and then, ultimately, general circulation models (to address the cloud and relative humidity distributions). The latter represents a grand challenge in climate modelling, for which present-generation models may be insufficient: there are difficulties associated with radiative transfer, clouds and dynamics (with a major component being condensable), and no empirical comparison cases.

Methods

Atmospheric structure. We prescribe the surface temperature and atmospheric structure (moist adiabatic, with bottom of atmosphere temperature equal to surface temperature) in a single global mean column⁹. We separately consider pure water atmospheres (which is the limit for extensive evaporation of the ocean and allows us to easily include the non-ideal behaviour of steam) and transitional atmospheres (up to a surface temperature of 400 K where the ideal gas law remains a good assumption). More information is given in the Supplementary Information, along with profiles (Supplementary Fig. S1).

Relative humidity. The standard assumption for 1D models is a linear decrease in relative humidity with pressure, from the surface to the tropopause^{31,32}. Mean subsaturation on Earth today is an area-weighted construction of areas of saturated and unsaturated air. Once water vapour is a major atmospheric constituent (a few per cent or more), regions of subsaturation would induce huge pressure gradients, leading to rapid mixing from saturated regions and permitting further evaporation. Hence, we assume that air below a level with some threshold water vapour mixing ratio (we use 5%) is saturated, with a linear decrease in relative humidity with pressure above.

Spectral data. We use line data for water from the HITEMP2010 spectral database¹⁹, which provides line data, including weak lines, in the range 0–30,000 cm⁻¹ (>0.33 μm; Supplementary Fig. S3) and absorption cross-sections for the far ultraviolet (>50,000 cm⁻¹, <0.2 μm). The line data are converted to cross-sections separately for each atmospheric profile examined using the LBLABC program, written by D.C. There are no spectral data available in the mid ultraviolet (30,000–50,000 cm⁻¹, 0.2–0.33 μm). This corresponds to 6% of incoming solar radiation, so we will underestimate net absorption. It is of utmost importance to use the most detailed line database available for water (Supplementary Figs S3 and S4). For thermal emission, HITRAN2004 and later perform similarly to HITEMP2010 up to 625 K, whereas HITRAN2k allows more outgoing radiation through the 4 and 10 μm windows. For solar radiation, all HITRAN data sets give less absorption than HITEMP2010. At high temperatures (thick atmospheres) the largest differences are seen at shorter wavelengths as net absorption depends on the ratio of absorption to Rayleigh scattering cross-sections (the latter depending on the reciprocal of the fourth power of wavelength). For greenhouse gases other than water, we use line data from HITRAN2008 (ref. 33).

Continuum absorption. The water vapour continuum is smoothly varying absorption that cannot be explained by near-centre contributions of known spectral lines. It is primarily responsible for setting the absorption cross-section in the water vapour window, which in turn largely determines the radiation limit. There has been interesting theoretical work on the source of this recently³⁴, but it will be some time until this provides a mature parameterization at the range of temperature, pressure and mixing ratio conditions that we encounter. Our approach is to use a more standard implementation, which is implemented with χ -factors³⁵ (Clough, personal communication, 2000) and performs similarly in tests to the empirical MT-CKD 2.4 continuum. We then test our computed absorption cross-sections against the best available data in the thermal infrared²⁰

(Supplementary Fig. S9). Our implementation slightly underestimates the strength of the continuum, so the radiation limit may be lower than our estimate and thermal exit from the runaway would require a warmer temperature. Whilst the temperature–pressure range of the data is limited, we take comfort in the fact that the thermal radiation limit is determined in the cool upper atmosphere, which is within observable ranges.

Rayleigh scattering. Previous work⁹ used Rayleigh scattering cross-sections for air throughout, as data for water were not available (J. Kasting, personal communication). Detailed refractive indices for water are now available³⁶, so we derive new constants for Rayleigh scattering (Supplementary Table S1). Water is a weaker Rayleigh scatterer than air, so using Rayleigh scattering cross-sections for air in place of water leads to erroneously low solar absorption, as too much sunlight is scattered (Supplementary Fig S6).

Radiative transfer calculation. We use the SMART code, written by D.C. (ref. 37), for our radiative transfer calculations. This code works at line-by-line resolution, but uses a spectral mapping algorithm to treat different wavenumber regions with similar optical properties together, giving significant savings in computational cost. We evaluate the radiative transfer in the range 50–100,000 cm⁻¹ (0.1–200 μm) as a combined solar and thermal calculation. Our solar source is spectrally resolved and we average the flux from zenith angles of 7.5°, 22.5°, 37.5°, 52.5°, 67.5° and 82.5°. In a few cases, models for one of these zenith angles did not run successfully owing to singularities in the matrix inversion in DISORT (ref. 38), which SMART uses for multiple scattering. In these cases, an appropriate average was made of the remaining five and the effect on the final results is trivial. Spectra shown are reduced using a simulated slit function. Our reference-standard radiative transfer model output is available as Supplementary Information to facilitate future model testing.

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Author contributions

C.G. and K.J.Z. suggested the study. C.G. conducted all of the modelling work, using a model written by D.C., with help from T.D.R. and D.C. All authors contributed to analysing the results.

Additional information

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Competing financial interests

The authors declare no competing financial interests.