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RESEARCH ARTICLE

Habitability constraints by nutrient availability in atmospheres of rocky exoplanets

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Abstract

Life as we know it requires the presence of liquid water and the availability of nutrients, which are mainly based on the elements C, H, N, O, P and S (CHNOPS) and trace metal micronutrients. We aim to understand the presence of these nutrients within atmospheres that show the presence of water cloud condensates, potentially allowing the existence of aerial biospheres. In this paper, we introduce a framework of nutrient availability levels based on the presence of water condensates and the chemical state of the CHNOPS elements. These nutrient availability levels are applied to a set of atmospheric models based on different planetary surface compositions resulting in a range of atmospheric compositions. The atmospheric model is a bottom-to-top equilibrium chemistry atmospheric model which includes the atmosphere–crust interaction and the element depletion due to the formation of clouds. While the reduced forms of CNS are present at the water cloud base for most atmospheric compositions, P and metals are lacking. This indicates the potential bio-availability of CNS, while P and metals are limiting factors for aerial biospheres.

Contents

Introduction 1
Defining nutrient availability levels 3
Atmospheric model 4
Results 6
  Applying nutrient availability levels 7
  Hydrogen, oxygen and water 8
  Carbon 8
  Nitrogen 9
  Sulphur 10
  Phosphorus 10
  Chlorine and fluorine 11
Summary and discussion 11
  Implications for surface biospheres 11
  Implications for aerial biospheres 12
  Implications for atmospheric biosignatures 12
Conclusions 14

Introduction

Some of the key questions for humankind are what the conditions favourable for the creation, evolution and persistence of life on Earth and other planets are. In recent times, many exoplanets have been detected and the question of the potential for biological and chemical processes to exist on these planets arose. These show a vast diversity, some without analogue in our own solar system. Especially for the rocky planets, a large diversity of different atmospheric compositions is expected (e.g. Leconte *et al.*, 2015; Grenfell *et al.*, 2020). While the extent of these atmospheres can range from a large envelope of primordial gas to almost no atmosphere at all, the composition can be very diverse, covering reducing and oxidizing atmospheres. For highly irradiated planets, the atmosphere can be composed of evaporated minerals due to the high temperature, which can melt the planetary surface. Recent observations with JWST for example constrain the presence of atmospheres on Trappist-1b (Ih *et al.*, 2023) and hint to a water-rich atmosphere of GJ 486b (Moran *et al.*, 2023). The upcoming next generation ground and space based telescopes (e.g. ELT Gilmozzi and Spyromilio, 2007, ARIEL Tinetti *et al.*, 2018) will allow to further investigate the composition of atmospheres of rocky exoplanets (see e.g. Lopez-Morales *et al.*, 2019; Wunderlich *et al.*, 2021; Ito *et al.*, 2021). The detailed understanding of the planets itself becomes important for interpreting observations, especially for the detection of biosignatures (Catling *et al.*, 2018; Meadows *et al.*, 2018; Lisse *et al.*, 2020; Lyons *et al.*, 2021). These signatures of life are mostly confined to water-based life, as it is the only known form of life. However, other solvents like NH_3 or CH_4 could potentially replace the liquid water for the formation of life (e.g. Ballesteros *et al.*, 2019).

In order to understand and constrain the existence of liquid water on the surface of a planet, the habitable zone has been defined (e.g. Huang, 1959; Kasting *et al.*, 1993). This concept has been further adjusted by including energy budgets and constraints on other molecules such as CO_2 for further needs for the evolution of life (e.g. Hoehler, 2007; Ramirez, 2020) and can explain the water on Earth and on the young Mars (Lammer *et al.*, 2018). However, water can also be found outside of this habitable zone, for example in sub-glacial oceans on Europa and Enceladus (Tjoa *et al.*, 2020) as well as in clouds of rocky planets, sub-Neptunes and gas giants (Benneke *et al.*, 2019; Charnay *et al.*, 2021; Madhusudhan *et al.*, 2021; Mang *et al.*, 2022). This shows that a large diversity of water environments is to be expected for exoplanets. Whereas the liquid water in oceans and below frozen surfaces provide the potential for subsurface biospheres on rocky planets and moons, the presence of water clouds on any planet could allow for some aerial biospheres.

The potential of an aerial biospheres for Venus, or as a general scenario, has been discussed previously (e.g. Morowitz and Sagan, 1967; Woese, 1979; Dartnell *et al.*, 2015), and recently gained further interest (Greaves *et al.*, 2021; Kotsyurbenko *et al.*, 2021; Rimmer *et al.*, 2021; Patel *et al.*, 2022) following from the proposed detection of phosphine (PH_3) in Venus' atmosphere (Greaves *et al.*, 2021).

While Bains *et al.* (2021, 2022) discussed that there is no known process available to abiotically produce a PH_3 feature in the Venus atmosphere, Omran *et al.* (2021) investigate the general possibility of phosphine production on rocky exoplanets. Assuming the production by some kind of living organism, this would introduce the necessity of a biosphere which is only present in a cloud layer. Individual organisms would need to remain afloat for long enough to metabolize and reproduce, before eventually falling towards the surface of Venus (Seager *et al.*, 2021a, 2021b; Patel *et al.*, 2022). This concept of aerial biospheres enlarges the possibilities of potential habitability from the presence of liquid water on the surface to all planets with liquid water clouds. See Seager *et al.* (2021b) for a discussion on the potential of aerial biospheres on mini-Neptunes. The water clouds of such planets could provide the chemical potential for the formation of life (Hallsworth *et al.*, 2021). This motivates a further investigation of the chemical environment of such aerial biospheres.

For the formation of life, the presence of liquid water is not the only hurdle to overcome, but also further elements and conditions need to be biologically available (see e.g. Deamer *et al.*, 2022). All life as we know it is made up of carbon, hydrogen, nitrogen, oxygen, phosphorus and sulphur, in the following referred to as CHNOPS elements (Horneck *et al.*, 2016). If these elements are available, chemical reactions triggered by stellar radiation, lightning, high energetic particles and cosmic rays can lead to the production of molecules, which allow the formation of amino acids and other pre-biotic molecules from which life can eventually form (see e.g. Bailey *et al.*, 2014; Ferus *et al.*, 2017; Ranjan *et al.*, 2021; Barth *et al.*, 2021). For these reactions, the red-ox state in which the elements are present is of particular importance, because elemental speciation dictates bioavailability.

The importance of the CHNOPS elements for life can be illustrated by the Redfield ratio. Redfield (1934) described, that organic matter in all of Earth's oceans comprises of a fairly constant ratio of CNP. This has been further expended to further elements (e.g. Anderson and Sarmiento, 1994; Ho *et al.*, 2003), revealing a stoichiometric ratio of C_{124}, N_{16}, P_1 and $S_{1.3}$, with further traces of metals. This universal incorporation of these nutrient elements underlines their importance to life as we know it and shows that the P and S are limiting factors for life.

In order to investigate the pre-biotic atmospheres from which life might evolve, we investigate and characterize the chemical state of the elements present in atmospheres of rocky exoplanets, with liquid water thermally stable at some point in the atmosphere. We use a chemical equilibrium model for the lower parts of atmospheres of rocky exoplanets with various surface compositions, which allow the study of all elements present in the gas phase. We especially focus on the different CHNOPS elements which are necessary for the formation of pre-biotic molecules in exoplanetary atmospheres.

Defining nutrient availability levels

The characterization of the habitability potential of different atmospheric compositions should take not only the presence of liquid water, but also the availability of essential elements into account. Therefore, we introduce a concept of nutrient availability levels.

The fundamental substance for any discussion of biology as we know it is liquid water. Therefore, the presence of liquid water is used as a prerequisite for every nutrient availability level. We considered any atmosphere without water condensates as uninhabitable. We defined atmospheres with only liquid water, but no nutrient elements, as having level 0 nutrient availability.

We define further nutrient availability levels as having the stability of water as a precondition and are based on the presence of the elements C, N and S, as H and O are by definition already present in the form of H_2O. We define an element i as present, if the concentration of an i bearing molecule is greater than one part per billion (10^{-9}, ppb). As the current understanding of the formation of life does not allow to preciously define a lower threshold for the needed concentrations of any molecule, we set this threshold to 1 ppb in the gas phase as this allows for the inclusion of atmospheric trace species. In principle, also atmospheric species with lower concentrations could accumulate locally by for example dissolution in cloud droplets. However, such processes are not included in this work and therefore the trace species concentration of 1 ppb is applied to define the presence of a molecule. If one or two of C, N and S are present, the atmosphere is classified as level 1 and level 2, respectively. In order to distinguish the different kinds of level 1 and 2, the present elements are added to the notation. For example, if C is present, but not N and S, the atmospheric composition classifies as level 1C. Following on this, the nutrient availability level 3 requires the presence of all three elements.

For the nutrient availability level 3, we further distinguish between the chemical redox state of the CNS elements. If all three elements are present in a reduced form (CH_4 for C, NH_3 for N and H_2S for S), the nutrient availability level is classified as level 3red. The need for this additional distinction is motivated as under such reducing conditions processes such as lightning can result in the formation of pre-biotic molecules, such as HCN, amino acids and nucleobasis (e.g. Miller, 1953; Miller and Urey, 1959; Toupance *et al.*, 1975; Ferris *et al.*, 1978; Parker *et al.*, 2011; Ferus *et al.*, 2017; Pearce *et al.*, 2022). Similarly to the reduced case, we define the oxidized regime as level 3ox with the presence of CO_2, NO_2 and SO_2. It is also possible, that different redox states of an element coexist in an atmosphere. If for carbon, nitrogen and sulphur the oxidized and reduced from coexist, with all species present at concentrations more than 10^{-9}, the atmosphere is categorized as nutrient availability level 3redox. In Woitke *et al.* (2021), we have shown that the coexistence of CO_2 and CH_4 is a fundamental result of chemical equilibrium for specific element abundances.

Table 1. *Definition of the nutrient availability levels*

Nutrient availability	$H_2O[l]$	C	N	S	P
Not habitable	✗	–	–	–	–
Level 0	✓	✗	✗	✗	✗
Level 1C	✓	✓	✗	✗	–
Level 1N	✓	✗	✓	✗	–
Level 1S	✓	✗	✗	✓	–
Level 2CN	✓	✓	✓	✗	–
Level 2CS	✓	✓	✗	✓	–
Level 2SN	✓	✗	✓	✓	–
Level 3	✓	✓	✓	✓	–
Level 3red	✓	red	red	red	–
Level 3ox	✓	ox	ox	ox	–
P	✓	–	–	–	✓

Notes: The tick marks and crosses indicate the presence and absence of the element in the header of the table, respectively. An element is considered present, if at least one molecule incorporating the element in question is present in the gas phase at concentrations above 10^{-9}. The state of the elements indicated with '–' do not matter for the definition of the respective nutrient availability level.
'red': indicates that the reduced form of the respective element (CH_4, NH_3, H_2S) is present at concentrations higher than 10^{-9}.
'ox': indicates the presence of the oxidized state (CO_2, NO_x, SO_2) at concentrations higher than 10^{-9}.
While these six molecules are the most prominent CNS bearing molecules, further molecules contributing to the presence of these elements are CO, COS, HNO_3, S_2O, H_2SO_4 and S_x.

Molecular nitrogen shows a very strong triple bond, which makes it much less accessible for life than other nitrogen sources such as and NH_3 or NO_2. Therefore, we distinguish between the presence of nitrogen in a more easily accessible form and N_2. We note that multiple processes exist, which transform N_2 to the more easily accessible forms of nitrogen (see e.g. Burris and Wilson, 1945; Stüeken, 2016). Examples for these can be of natural origin (e.g. lightning or UV and cosmic ray radiation) as well as biotic by microbes.

In addition to the nutrient availability levels based on CNS, the presence of phosphorus is a further constraint for the formation of life. As it is a limiting factor for the biosphere on Earth (Syverson *et al.*, 2021), we use it as a category on its own. All nutrient availability levels are summarized in Table 1.

These nutrient availability levels are defined to investigate whether or not atmospheres with liquid water clouds can provide basic necessities for possible aerial biosphere. In order to principally allow the presence of an aerial biosphere, all nutrients need to be available. However, even in the case of their presence, further questions of whether such an aerial biosphere can exist need to be investigated. This includes but is not limited to processes that form more complex chemicals from the nutrients, but also whether life can remain afloat for long enough periods of time.

Atmospheric model

In order to investigate the implications of the concept of the different nutrient availability levels defined in the previous section on atmospheres of rocky planets, we create atmospheric models with different element abundances to which we apply the concept of the nutrient availability levels. The atmospheric model is presented in Herbort *et al.* (2022) and uses the chemical equilibrium solver GGchem (Woitke *et al.*, 2018). The bottom-to-top atmosphere model describes the lower atmosphere with a polytropic hydrostatic equilibrium atmosphere which takes element depletion by cloud formation into account. The base of the atmosphere is in chemical phase equilibrium with the crust, as described in Herbort *et al.* (2020). In every atmospheric layer, the chemical equilibrium is solved and all thermally stable condensates are removed while the gas phase is the total element abundance of the atmospheric layer above. This depletes the element abundances of all elements affected by cloud formation.

Each bottom-to-top atmosphere is fully defined by the surface pressure (p_{surf}), surface temperature (T_{surf}), set of total element abundances (ϵ_{tot}) and the polytropic index (γ).

As in Herbort *et al.* (2022), we investigate (p, T) profiles with a constant polytropic index $\gamma = 1.25$. The surface conditions are defined such that the atmosphere reaches the reference temperature of $T_{\mathrm{ref}} =$ 300 K in the atmosphere at $p_{\mathrm{ref}} = 1$ bar. This is guaranteed for a (p_{gas}, T_{gas}) profile defined by

$$T_{\mathrm{gas}} = \frac{T_{\mathrm{ref}}}{p_{\mathrm{ref}}^{1-1/\gamma}} \cdot p_{\mathrm{gas}}^{1-1/\gamma}. \tag{1}$$

We investigate a surface temperature range from 300 K to 1000 K. The surface pressures for the different surface temperatures are given by equation (1) to ensure that all atmospheric profiles reach the reference point.

In our calculations, the included 18 elements (H, C, N, O, F, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Cr, Mn and Fe) can form 471 gas species and 208 condensates. The total element abundances ϵ_{tot} for the different compositions used in this paper are listed in Table A1 and briefly introduced in the following. The compositions of Bulk Silicate Earth (BSE, Schaefer *et al.*, 2012), Continental Crust (CC, Schaefer *et al.*, 2012), Mid Oceanic Ridge Basalt (MORB, Arevalo and McDonough, 2010) and CI chondrite (CI, Lodders *et al.*, 2009) are used to represent a variety of different rock compositions. In order to investigate different levels of hydration, we use sets of element abundances based on BSE with increased abundances of H and O (BSE12, BSE15). The enrichment of respectively 12 and 15 weight percent H and O results in a crust composition rich in hydrated minerals (for further details see Herbort *et al.*, 2020). The Earth set of total element abundances is the result of a fit of our atmosphere-crust chemical and phase equilibrium model. At $p = 1.013$ bar and $T = 288.15$ K, this set of element abundances results in a modern Earth-like gas phase, except for the CH_4, which is in disequilibrium (for further details see Appendix A Herbort *et al.*, 2022). The Archean model represents a strongly reducing atmosphere (presence of CH_4 and NH_3) and stable H_2O[l]. In order to also include non-solar system-based elemental ratios, three models based on element abundances from polluted white dwarfs (PWD Melis and Dufour 2016) are included. As not all elements are included, the missing abundances (especially H) are completed from BSE, MORB and CI. In order to investigate hydrogen-rich atmospheres, a model with solar abundances (Asplund *et al.*, 2009) is taken into account. For further discussions and effects of these elemental compositions for different temperature regimes, see Herbort *et al.* (2020) and Herbort *et al.* (2022).

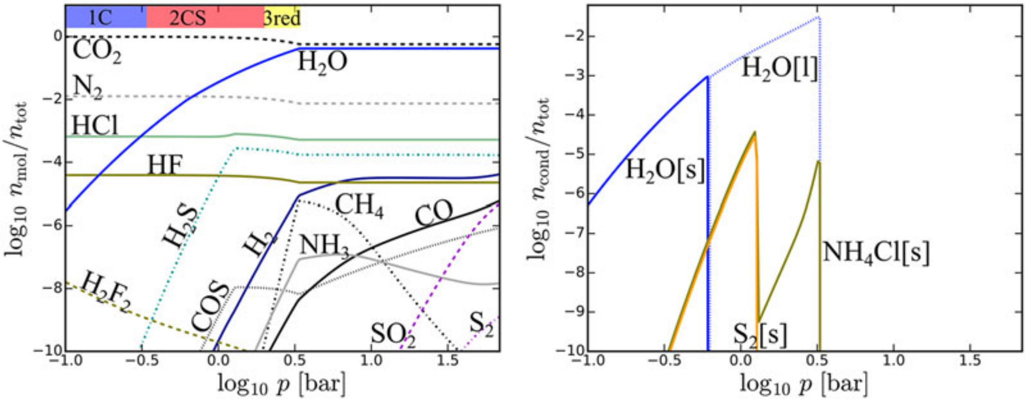

Figure 1. *Results of the 1D atmospheric model for CC total element abundances and surface conditions of* $T_{surf} = 700$ *K and* $p_{surf} = 70$ *bar.* ***Left panel:*** *Gas-phase composition of the model atmosphere with the indicated nutrient availability levels at the top.* ***Right panel:*** *Thermally stable cloud condensates in the model atmosphere.*

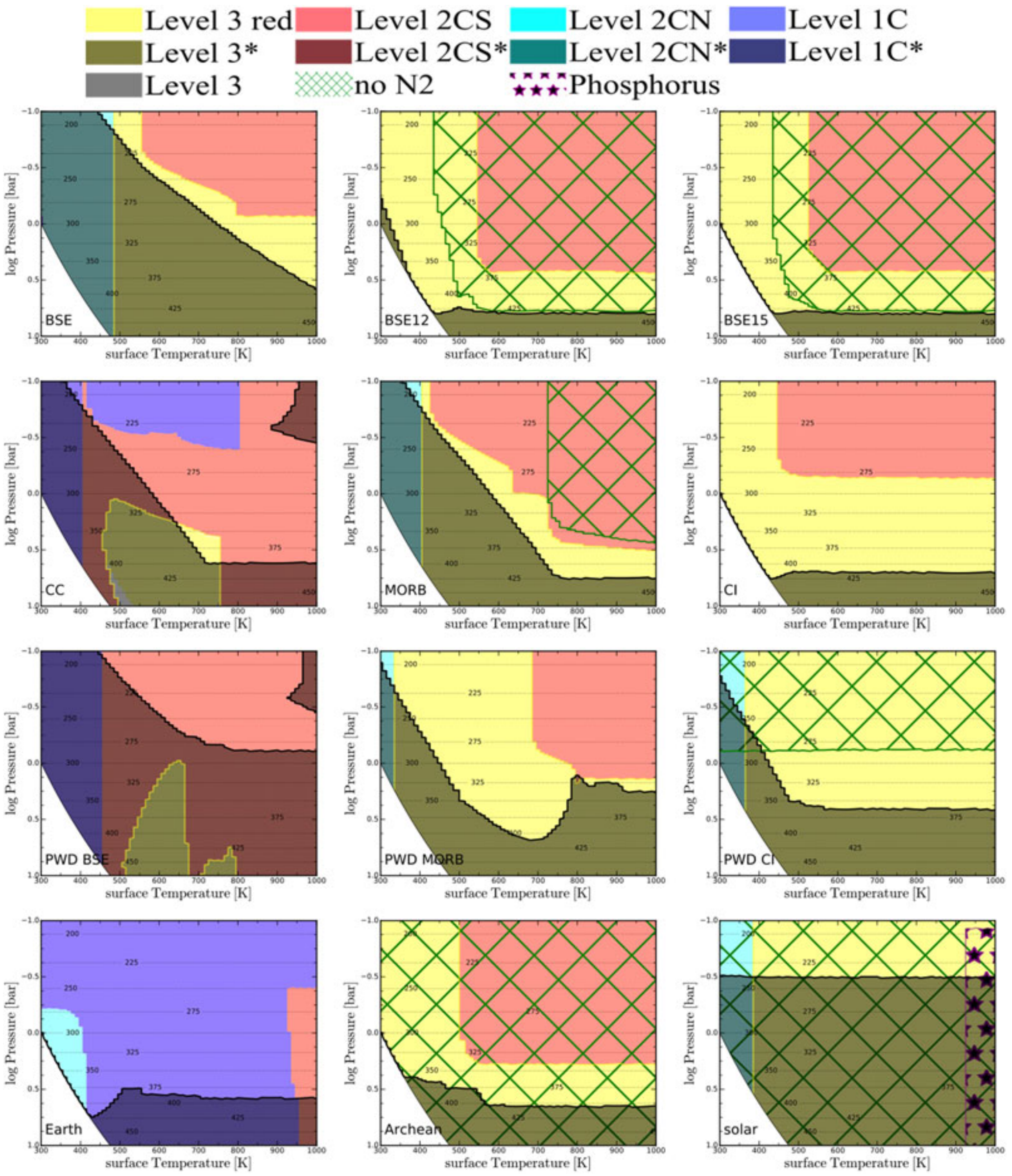

Figure 2. *Nutrient availability levels applied to a range of atmospheric models for different sets of total element abundances. The colouring of all present nutrient availability levels are given in the legend above. The solid black line provides the pressure level of the* H_2O*[l,s] cloud base in the respective models. The darker shade of the nutrient availability levels (indicated with * in the legend) shows the corresponding nutrient availability level of gas-phase composition although the primary assumption of the presence of* H_2O*[l,s] is not fulfilled. The green cross hatched region indicates* $N_2/n_{tot} < 10^{-9}$. *The horizontal dotted lines refer to the gas phase temperature* T_{gas}.

Results

In this section, we investigate nutrient availability levels in our model atmospheres with different sets of total element abundances and subsequently discuss the effects on the different elements individually.

First we illustrate the nutrient availability levels for a single model atmosphere for CC total element abundances and surface conditions of $T_{\mathrm{surf}} = 700$ K and $p_{\mathrm{surf}} = 70$ bar shown in Fig. 1. The nutrient availability levels are indicated at the top of the left panel. At the surface, the atmosphere is dominated by CO_2 and H_2O, with ten further molecules being present at concentrations higher than 10^{-9}. This includes H_2S, CO, SO_2, COS and S_2 as CHNOS bearing molecules. With decreasing pressure and temperature, CH_4 becomes the second most important carbon bearing species while CO and COS decrease in concentration. At $p_{\mathrm{gas}} \approx 3.5$ bar, H_2O[l] becomes a thermally stable condensate. Due to the presence of CH_4, NH_3 and H_2S, this part of the atmosphere is of nutrient availability level 3red. It is remarkable that C, N and S are all abundant in their reduced form, despite the most abundant species in the atmosphere being oxidized. The removal of H_2O[l] and NH_4Cl[s] condensates causes the depletion of H_2O, H_2, CH_4 and NH_3. The drop of NH_3 below a concentration of 10^{-9} causes the nutrient availability level to change to level 2CS. The removal of the thermally stable S_2[s] condensate causes the decrease in H_2S concentration, which drops below 10^{-9} leaving behind a level 1C atmosphere. The only molecules, which reach concentrations above 10^{-9} for $T_{\mathrm{gas}} < 450$ K and include further elements besides CHNOPS include fluorine or chlorine. The respective concentrations of HCl and HF remain roughly constant throughout the atmosphere. Similarly, N_2 does not show large changes in concentration.

Applying nutrient availability levels

The nutrient availability levels for the 12 different sets of total element abundances with varying surface conditions are shown in Fig. 2. Although these nutrient availability levels all require the presence of liquid water, the regions without water condensates are indicated similarly to the nutrient availability levels, but shaded in black. Of the 18 elements included in this model, only nine elements (C, H, N, O, P, S, Cl, F and Si) are present in the investigated atmospheric (p_{gas}, T_{gas}) space in the form of a gas-phase molecule with concentrations higher than $n_i/n_{\mathrm{tot}} > 10^{-9}$, where n_i is the number density of the molecule i and n_{tot} is the total number density. All 23 molecules that reach concentrations above 10^{-9} in the investigated part of the atmosphere are listed in Table 2 and sorted according to their peak concentration in any of the models.

Most atmospheres for the different sets of total element abundances are nutrient availability level 3red at the water cloud base. However, for $T_{\mathrm{surf}} \lesssim 400$ K, the concentrations of the sulphur bearing molecules are lower than 10^{-9} and the nutrient availability level drops to level 2CN or level 1C with additional N_2 being present. For these lower temperatures, the S abundance in the atmosphere is significantly lower, as more sulphur is incorporated into the crust (see Appendix).

The most dominant transition in the nutrient availability levels is the change from level 3red to level 2CS, which occurs in most of the models. This depletion in available nitrogen is a result of the condensation of NH_4Cl[l], which reduces the N element abundances. Especially for the BSE12, BSE15 and MORB sets of total element abundances, the condensation of NH_4Cl[s] depletes the N abundances such that also the N_2 concentration drops below 10^{-9}.

Besides H_2O[l,s] and NH_4Cl[s] a total of further 16 thermally stable condensates with normalized number densities of $n_{\mathrm{cond}}/n_{\mathrm{tot}} > 10^{-10}$ are present (see also Herbort *et al.*, 2022). These thermally stable cloud condensates can be seen in Fig. A3 in Appendix. For all sets of total element abundances NaCl[s] and KCl[s] are stable condensates for $T_{\mathrm{gas}} > 700$ K. Furthermore, every set of element abundances shows a (p_{gas}, T_{gas}) region, where H_2O[s] condenses. Only for the solar abundances there is no regime with H_2O[l] as a condensate. Another cloud condensate that is thermally stable in atmosphere of all element abundances, except solar, is the N bearing condensate NH_4Cl[s].

A more strict definition of the nutrient availability levels would include a concentration threshold of 10^{-6} instead of 10^{-9} for the relevant elements. The resulting nutrient availability levels for this are shown in Fig. A2. The parameter space that the level 3red nutrient availability regions are covering is significantly shrunken and mostly replaced by level 2CS. This result shows that the presence of

Table 2. *Gas-phase molecules according to their highest concentration in the models*

Element	>10%	>0.1%	$>10^{-6}$	$>10^{-9}$
C	CO_2, CH_4		CO, COS	
N	N_2	NH_3		HNO_3, NO_2 (Earth $T_{surf} < 400$ K)
P				P_4O_6(solar $T_{surf} > 920$ K)
S		H_2S, SO_2	COS, $[S]_x$	S_2O(PWD BSE), H_2SO_4(Earth $T_{surf} > 920$ K)
Cl		HCl, Cl_2(Earth)		OHCl(Earth $T_{gas} > 425$ K)
F		HF		SiF_4($T_{surf} > 900$ K)
Others	O_2, H_2, H_2O			

Notes: Some of the present molecules are only present in few atmospheric models. For these, the defining character of these models is added in brackets. Examples are a specific temperature range or a given total element abundance.

N can especially be a limiting factor if concentrations of the order of 10^{-8} for nitrogen bearing species is not sufficient.

In all models that reach nutrient availability level 3, the molecules are present in the reduced form. This is also true for the majority of the parts of the atmosphere, where water is not a stable condensate. The only model, where this is not the case is for the CC total element abundances with $T_{surf} \approx 500$ K and $T_{gas} > 425$ K.

Hydrogen, oxygen and water

The most fundamental molecule for the discussion of habitability is water, which is not only present as a condensate, but also as a dominant gas-phase molecule in many planetary atmosphere. Figure A9 in the Appendix shows the gas-phase concentrations of H_2O, H_2 and O_2.

The BSE and PWD BSE models are the models with the lowest H_2O concentration, due to the low hydrogen mass fraction in the total element abundances. H_2O does not reach concentrations above 10^{-6} for $T_{surf} \lesssim 430$ K and for $T_{surf} \lesssim 340$ K the H_2O concentration stays below 10^{-9}. The only other models, for which the H_2O concentrations drop below 10^{-9} are the sets of total element abundances of MORB and CC with $T_{surf} \lesssim 340$ K. For all other models, the H_2O concentrations are significantly higher and can become the most abundant gas-phase species, especially for high surface temperature models.

As noted above, for all sets of total element abundances, there exist atmospheric models, where H_2O[l,s] cloud condensates are stable and abundant. The H_2O gas-phase concentration above the cloud base is a result of the assumption of unity for the supersaturation, $S(H_2O[l,s]) = 1$. Therefore, every two models at the same (p_{gas}, T_{gas}) above the water cloud base show the same gas-phase concentration of H_2O. This confines the maximum H_2O concentration for the atmosphere above.

The only model which shows the presence of gaseous O_2 is the Earth model, as it is created to have an atmospheric composition similar to modern day Earth's atmosphere for $T_{surf} = 288$ K at the surface (Appendix A of Herbort *et al.*, 2022). For higher surface temperatures, the O_2 concentration reduces as the CO_2 concentration increases to become the most abundant gas-phase molecule, surpassing N_2. As the Earth model is the only model with a type B atmosphere (oxygen rich, see Woitke *et al.*, 2021; Herbort *et al.*, 2022), it is the only model to show O_2 as a gas species. For H_2 on the other hand, the picture is different, as the models for total element abundances of solar, Archean and CI composition result in atmospheres rich in H_2 for all surface temperatures (type A atmospheres Woitke *et al.*, 2021). For BSE12 and BSE15 total element abundances, H_2 is present for all T_{surf} where water is not condensing. For all other models, H_2 is only a trace gas.

Carbon

Of the CNS elements, carbon is the only element, for which there are always molecules present at concentrations higher than 10^{-9} for the entire parameter space of all sets of total element abundances studied in this paper. Carbon is bound in different oxidization states, ranging from CH_4 to CO_2, with some additional traces of CO and COS at concentrations of 10^{-6} (see Fig. A4). This coexistence of CH_4 and CO_2 is a fundamental result of chemical equilibrium for all atmospheres of atmospheric type C (Woitke *et al.*, 2021). For $T_{gas} \lesssim 350$ K, both redox end-members of carbon are present at concentrations higher than 0.1% in multiple models, while only PWD CI, Earth, Archean and Solar total element abundances show no coexistence of these redox states throughout the parameter space. The trace forms of carbon COS and CO are only present at concentrations up to 10^{-6}, if CO_2 is present at more than 0.1%. The importance of especially CO increases with higher gas temperature towards the lower parts of the model atmospheres. The significant presence of CO as a major carbon species can be seen in other works, such as Fegley *et al.* (1997); Visscher *et al.* (2006); Woitke *et al.* (2021) and others.

Throughout the atmospheres in this paper, the only form of carbon which is thermally stable as a condensate in the atmosphere is C[s] (graphite). It is only thermally stable in atmospheres where CH_4 and CO_2 coexist. In these cases, the atmospheric composition in the upper parts of the atmosphere evolves towards the dominance of the more abundant one of the two carbon molecules. The concentration of the respective other molecule decreases significantly as a result due to the ongoing depletion of carbon in the atmosphere. As discussed in Herbort *et al.* (2022), graphite is the only condensate which can be thermally stable for the entire investigated T_{gas} parameter space. The models based on CC total element abundances are the only ones for which CH_4 and CO_2 coexist in the atmosphere and C[s] does not condense.

For the carbon condensation, the different element abundances fall into three different regimes. For the element abundance without carbon condensation in the atmosphere and a hydrogen-rich atmosphere, also no carbon bearing condensate is stable as part of the crust. However, for the Earth model, where also no graphite condenses in the atmosphere, all non-gaseous carbon is bound in $CaCO_3$[s] and $CaMgC_2O_6$[s]. The other element abundances have either one or a multitude of carbon-bearing condensates stable at the crust.

For PWD BSE ($T_{surf} < 700$ K) and PWD MORB, the carbon condensates are $CaCO_3$[s], C[s] and $CaMgC_2O_6$[s]. These condensates are also stable for the model with the highest C abundance (3.5% mfrac for the CI), but the further condensates of $MgCO_3$[s], NCO_3[s], $FeCO_3$[s] and $CaFeC_2O_6$ are also stable. The carbon abundance in BSE, BSE12, BSE15 and MORB is more than two orders of magnitude lower than for CI or the PWD models. This results in a much simpler carbon condensate phase, with either C[s] or $CaCO_3$[s] being stable for the corresponding models. Similarly for the CC element abundance, carbon is bound in the crust in the form of $CaMgC_2O_6$[s] for $T_{surf} < 680$ K.

Nitrogen

The nitrogen in the gas phase can be found in the forms of N_2, NH_3, HNO_3 and NO_2 and their distribution is shown in Fig. A5. While there are no N bearing condensates in the crust for any model, NH_4Cl[s] and NH_4SH[s] are thermally stable condensates in the atmospheres (see Fig. A3).

For many sets of element abundances, N_2 is a main N carrier at concentrations over 1‰ for the entire investigated parameter space (BSE, CC, CI, Earth) or up to a certain threshold surface temperature ($T_{surf} < 450$ K: BSE12, BSE15; $T_{surf} < 650$ K: MORB, PWD BSE; $T_{surf} < 775$ K: PWD MORB). Otherwise, for H-rich atmospheres, NH_3 becomes the main N carrier for the entire parameter space of PWD CI, Archean and Solar, but also for $T_{surf} > 450$ K BSE12, BSE15. For the latter two, the dominance of N_2 over NH_3 is only present, when H_2O[l] is a stable crust condensate.

Although N is considered as an omnipresent gas species, we find the high atmosphere can be depleted in nitrogen for $T_{surf} \gtrsim 550$ K and therefore higher surface pressures. The depletion of N is caused by the condensation of NH_4Cl[s], which removes a considerable amount of N from the gas phase. This is the case for the the sets of total element abundances of BSE12, BSE15 and Archean. For MORB abundances, this temperature threshold is $T_{surf} > 725$ K. The condensation of NH_4Cl[s] is present in most models and coincides with the condensation of H_2O[l]. Therefore, the NH_4Cl[s] condensates are of additional interest for the discussion of habitability.

A further N condensate is NH_4SH[s], which is only thermally stable for $T_{gas} < 250$ K. This further depletes the N abundance for BSE12, BSE15, PWD MORB and PWD CI.

Sulphur

The most abundant sulphur bearing gas-phase molecule is H_2S. Only for high T_{gas} of CC and PWD BSE total element abundances SO_2 become more significant than H_2S, which remains present at concentrations of more than 10^{-6}. Furthermore, traces of COS, S_x, S_2O and H_2SO_4 can be present. In general, for higher T_{surf} a higher abundance of sulphur is found in our atmospheres (see also Fig. A6). The only model where no S bearing molecule is present at concentrations greater than 10^{-9} for $T_{surf} > 450$ K is the Earth model, for which only H_2SO_4 is present for $T_{surf} > 900$ K. This significantly depleted S abundance with respect to all of the other models is a result of the thermal stability of S condensates in oxidized environments. The investigation of S in CHNO+S atmospheres is further discussed in detail in Janssen *et al.* (subm). For the CC model with 400 K < T_{surf} < 800 K, the condensation of S_2[s] is sufficient enough to reduce the S abundance for $T_{gas} \lesssim 240$ K to below 10^{-9}. For all models, with $T_{surf} \gtrsim 450$ K H_2S is present with concentrations higher than 10^{-9} in the region of the H_2O[l] cloud base.

For most of the element abundances, the majority of the sulphur is kept in the crust as condensates. This explains the higher concentration of sulphur bearing molecules with higher temperatures, as the vapour pressure of the condensates increases with increasing temperature. Only for BSE12, BSE15 and Archean total element abundances and $T_{surf} > 700$ K no S condensate is stable at the crust. For most abundances and T_{surf} only one S condensate is thermally stable. These stable condensates are FeS for BSE, MORB, PWD MORB, PWD CI (all T_{surf}), BSE12, BSE15 (680 K < T_{surf} < 440 K), CI ($T_{surf} > 510$ K), Archean ($T_{surf} < 690$ K), solar ($T_{surf} < 670$ K); FeS_2 for BSE12, BSE15 ($T_{surf} < 430$ K), CC ($T_{surf} < 640$ K), CI ($T_{surf} < 500$ K), PWD BSE ($T_{surf} < 350$ K); $CaSO_4$[s] for CC ($T_{surf} > 650$ K), Earth (all T_{surf}) and MnS for solar ($T_{surf} > 680$ K). The PWD BSE total element abundance is the only model, where multiple S condensates are stable for the same surface condition. These stable condensates are FeS[s] and one of $CaSO_4$[s] and FeS_2[s] for $T_{surf} > 800$ K and 350 K < T_{surf} < 800 K, repetitively.

The sulphur-containing cloud condensates fall into three categories. First, at the highest temperatures, pure sulphur condensates of S2[s] and S[l] dominate. These can be present over a large range of surface conditions. In general, the S_2[s] cloud base lies at lower pressures than the H_2O[l] cloud bases. Second, H_2SO_4[s], which is only stable for models with high pressures at the surface and cold local temperatures of $T_{gas} \lesssim 300$ K. The H_2SO_4[s] and S_2[s] condensates can coexist in the one atmospheric model. Third, the NH_4SH[s] condensate is only stable for $T_{gas} \lesssim 250$ K. As NH_4SH[s] does not coexist in an atmosphere with either S_2[l], S[l] or H_2SO_4[s], these condensates could be used for the characterization of the atmospheric conditions.

Phosphorus

The rarest of the CHNOPS elements in our model atmospheres is phosphorus, which is for all non-solar total element abundances almost completely bound in the crust, in the form of hydroxyapatite and fluorapatite, $Ca_5P_3O_{13}H$[s] and $Ca_5P_3O_{12}F$[s], respectively. Apatite has been found to be the main phosphorus carrier for example on the Chelyabinsk meteorite (Walton *et al.*, 2021). This shows that P will be a limiting element for the formation and evolution of life in the atmospheres of almost all rocky exoplanets.

Only models for solar element abundances and $T_{surf} > 900$ K show any phosphorus-bearing molecule at concentrations higher than 10^{-9} in the atmosphere. At the crust–atmosphere interaction layer of these models, PH_3 is the dominant P carrier, with about 3 orders of magnitude higher concentration than P_4O_6. However, as virtually no Ca is present in the atmosphere, the remaining P cannot condense in the form of apatite and only condenses much higher in the atmosphere, when H_3PO_4[s]

becomes thermally stable at around $p = 10^{-0.5}$ bar and $T_{gas} = 250$ K. Despite the overall reducing atmospheric conditions in the H_2 dominated atmosphere, the most important P carrier changes close to the surface at $T_{gas} \approx 900$ K from PH_3 to P_4O_6. This change from the reduced to the oxidized form is consistent with previous models (e.g. Lodders and Fegley, 2002; Visscher *et al.*, 2006).

Chlorine and fluorine

The only molecules which reach concentrations above 10^{-9} for $T_{gas} < 450$ K and include further elements besides CHNOPS include the fluorine or chlorine. The gas phases in the relevant temperature regime are visualized in the Appendix in Figs. A7 and A8, respectively.

Most of the halides are condensed in the crust in the form of NaCl[s] and $Ca_5P_3O_{12}F$[s]. Additionally, MgF_2[s] tends to be stable towards higher temperatures, whereas CaF_2[s] is stable towards lower temperatures. Only for the CC total element abundances, the additional condensate of $KMg_2AlSi_3O_{10}F_2$[s] is thermally stable at the crust for $T_{surf} > 370$ K. The further condensate of $CaCl_2$[s] is only found stable in the model of BSE12 total element abundances for $T_{surf} \approx 590$ K.

The halides show increasing gas-phase concentrations for high surface temperatures, where residual Cl and F atoms remain in the gas phase. The most important species being HCl and HF for all atmospheres. In the atmospheres at $T_{gas} \gtrsim 700$ K, the condensation of KCl[s] and NaCl[s] occurs for every total element abundance. However, this condensation is limited by the abundance of K and Na, respectively. As a result, the concentration of HCl and HF reaches up to a few 10^{-3} in the atmospheres based on a high surface temperature. The presence of these hydrogen halides in coexistence with H_2O[l] can result in acidic environments, if they dissolve in the water droplets. Whether this acidity benefits or limits the formation of pre-biotic molecules is beyond the scope of this paper and is therefore not included in the nutrient availability levels as either helpful or harmful.

SiF_4 is the only CHNOPS bearing molecule present at concentrations higher than 10^{-9} in any model of our atmospheric models for $T_{gas} < 450$ K. It is only present for the CC and Earth total element abundances and $T_{surf} \gtrsim 900$ K.

Summary and discussion

In this paper, we defined nutrient availability levels based on the presence of water condensates and availability of nutrients. We used this concept to explore the potential for habitable atmospheres to form under equilibrium conditions in rocky exoplanets.

We find that for most atmospheres at (p_{gas}, T_{gas}) points, where liquid water is stable, CNS bearing molecules are present at concentrations above 10^{-9}. Carbon is generally present in every atmosphere, while the sulphur availability in the gas phase increases with increasing T_{surf}. For lower T_{surf} nitrogen in the form of either NH_3 or N_2 is present as a major contributor to the atmospheric composition for most atmospheres. However, for models with higher T_{surf}, the presence of gaseous nitrogen can be significantly depleted by the condensation of NH_4Cl[s], which leaves an upper atmosphere devoid of nitrogen. The limiting element of the CHNOPS elements is phosphorus, which is mostly bound in the planetary crust. This is consistent with the scarcity of phosphorus limiting the biosphere during at least some parts of Earth history (Syverson *et al.*, 2021).

Implications for surface biospheres

Similar to previous work, our models suggest that the limiting factor for habitability at the surface of a planet is the presence of liquid water. If water is present at the surface, CNS are available in the gas phase of the near-crust atmosphere. The only exception is the present day Earth model, which lacks the presence of gaseous S by construction.

The available elements for the equilibration of crust and atmosphere are dependent on the overall planetary composition and therefore formation history, as well as the tectonic regimes present (e.g. Tosi *et al.*, 2017; Dorn *et al.*, 2018; Kruijver *et al.*, 2021; Meier *et al.*, 2021; Höning *et al.*, 2021).

The evolution of a planet and its start also play a crucial role, as this determines the long-term temperature of the planet (Turbet *et al.*, 2021; Seales and Lenardic, 2021). The formation of hydrated minerals can also play an important role in the long-term stability of water condensates (Herbort *et al.*, 2020; Beck *et al.*, 2021). An example for this is the surface of Mars, which previously hosted liquid water, which is now bound in hydrated rocks (see e.g. Ehlmann *et al.*, 2010; Wernicke and Jakosky, 2021).

Even if (p_{surf}, T_{surf}) of a planet allow the formation of liquid water and water is present in the atmosphere, even in the form of clouds, our models show that no water ocean is to be implied from these (compare also Ding and Wordsworth, 2022). Especially if the surface pressure is not well constrained, the presence of water clouds is not conclusive for the presence of water at the surface of the respective planet. Therefore, additional detection methods of a surface ocean are necessary to confirm the existence (e.g. Williams and Gaidos, 2008; Lustig-Yaeger *et al.*, 2018; Ryan and Robinson, 2022)

If water is available at the surface, the elements not present in the gas phase are stored in the crust condensates. Processes such as chemical weathering or dissolution of condensates into water can make these elements available for life. This provides a pathway to overcome the lack of atmospheric phosphorus and metals, which are used in enzymes that drive many biological processes. However, there can be the difficulty in accumulating the pre-biotic molecules, if the water reservoir is too large and no exposed land is available (Noack *et al.*, 2016). This leads to Darwin's idea of the warm little pond with some dry–wet cycles (Follmann and Brownson, 2009; Pearce *et al.*, 2017). This hurdle also needs to be overcome for potential life on the icy moons in our solar system (see e.g. Vance *et al.*, 2007; Tjoa *et al.*, 2020; Taubner *et al.*, 2020). If indeed it can be shown that life can form in a water ocean without any exposed land, this constraint becomes weaker and the potential for the surface habitability becomes mainly a question of water stability.

Implications for aerial biospheres

Many of the models show the presence of a liquid water zone in the atmospheres, which is detached from the surface. These regions could be of interest for the formation of life in the forms of aerial biospheres.

Previous work by, for example, Seager *et al.* (2021b) focussing on sub Neptunes – which have thicker atmospheres than planets investigated in this work – has shown that aerial biospheres can be regions of nutrient scarcity. We show that some of the fundamental elements (CHNOS) are likely to occur in regions of most planetary atmospheres. These elements are mostly in a reduced state near the water cloud base, which is favourable for the formation of pre-biotic molecules. However, our models also show that phosphorus is unlikely to occur in planetary atmospheres, as it is bound in the crust in the form of apatite. A further hurdle for more complex life is the ability to use redox-sensitive transition metals as cofactors in enzymes. Our models show these metals are also absent in most atmospheres, as they are bound in the crust. Further cloud condensates to H_2O[l,s] like NH_4Cl[s] could play this role for life in such environments (see Seager *et al.*, 2021b). Our models show that these two cloud condensates coexist for most atmospheric models.

The lacking atoms of phosphorus and metals could in principle be provided by atmospheric updrafts induced by storms on the planet. The closer the liquid water zone is to the surface, the more likely an updraft of surface material would be. However, further cloud layers are expected at higher temperatures on planets with higher surface pressure (e.g. NaCl[s], KCl[s], FeS[s] or FeS_2[s]). These condensates could be delivered to the higher atmospheres by storms and accumulated by life in the higher atmosphere.

Implications for atmospheric biosignatures

The atmospheric composition in this work focussed on the pre-biotic atmospheric composition. The atmospheric composition after the emergence of life on a planet can deviate drastically from the pre-biotic atmospheric composition caused by biological activity itself (e.g. Holland, 2002; Izon *et al.*, 2017; Gregory *et al.*, 2021). Our approach does not directly aim for the understanding of biosignatures and atmospheres of planets, which are inhabited, but for the conditions in which pre-biotic chemistry can occur.

The results of our approach of a large variety of different atmospheric compositions in chemical equilibrium atmospheres can provide insights to which signatures of biology could also be produced by a simple chemical equilibrium atmosphere. We, for example, show that N_2 is a major atmospheric component for planets with $T_{surf} < 400$ K, whereas N in general can be lacking in atmospheres based on higher T_{surf}. Lammer *et al.* (2019) state that N_2 itself can be a sign of a biosignature or effective tectonic regimes. Our models of an equilibrated crust with the atmosphere can be seen as representative of active tectonic regimes as otherwise only a small portion of the crust is able to equilibrate with the atmosphere. Following up on this, Sproß *et al.* (2021) state that the coexistence of N_2 and O_2 is only a result of biology. We find that this coexistence – as well as the presence of O_2 in general – is only possible in a model with element abundances adopted to produce this exact atmospheric feature.

Another pair of atmospheric gas species that is discussed as a biosignature is CO_2 and CH_4 (e.g. Krissansen-Totton *et al.*, 2019; Wogan *et al.*, 2020; Mikal-Evans, 2021). However, as discussed in Woitke *et al.* (2021), it is possible to have these two molecules coexist in chemical equilibrium. In our atmospheres, we find this element pair coexisting at concentrations above 10^{-6} for many atmospheres with $T_{gas} > 275$ K. For a concentration greater than 1‰ of both, CO_2 and CH_4, $T_{gas} > 350$ K is necessary. Therefore, high concentrations of both molecules at cool temperatures could be explained by some out-of-equilibrium process such at biology. In the early Earth atmosphere during the Archean, the methane has been a product of biology (methanogenesis Ueno *et al.*, 2006). Similarly, it has been discussed that the presence of CH_4 in plumes of Enceladus could potentially be a result of methanogenesis (Affholder *et al.*, 2021).

HCN has been found, for example, on the surface of Titan (Lellouch *et al.*, 2017) and the comet Hale-Bopp (Jewitt *et al.*, 1997). Its production in planetary atmosphere is possible by lightning, especially if CH_4 and N_2 are present (Hodosan *et al.*, 2017; Pearce *et al.*, 2022). However, we do not find the presence of HCN in any of our models, underlining their non-equilibrium origin. With the presence of HCN, Rimmer *et al.* (2021) found that the production of HC_3N should occur on super-Earths rich in nitrogen and an overall reduced atmospheric composition by photochemistry.

Further molecules that we do not find to be present in any of our models are more complex hydrocarbons than CH_4. However, these have been found in various environments ranging from interstellar matter (e.g. Henning and Salama, 1998) to places in our solar system. Examples are the C_2H_4 haze on Triton (Ohno *et al.*, 2021) or various different molecules on comets such as 67P (Müller *et al.*, 2022).

Huang *et al.* (2021) found that the detection of NH_3 can be a biosignature on planets, where a deep atmosphere is excluded. We however find, that NH_3 can be present throughout the atmosphere of uninhabited planets with low pressure atmospheres. However, only if no water condensate is stable at the surface, we find that the NH_3 concentration actually exceeds 1‰. The reason for this difference can be that there is no photochemistry included in our model. Species such as NH_3 and CH_4 can be easily photodissociated, resulting in a significantly lower concentrations and the production of hazes.

By construction of the model, the influence of stellar irradiation to the planetary atmosphere is not discussed. A proper understanding of the influence of the host star to the atmosphere requires the upper atmosphere and is therefore beyond the scope of this paper. This can have a major factor for the atmospheric evolution (Chen *et al.*, 2020; Turbet and Selsis, 2021; Chebly *et al.*, 2021; Locci *et al.*, 2022; Teal *et al.*, 2022). The triggered photochemistry can not only destroy pre-biotic molecules, but also enhance the formation of these as many reactions need the input of some sort of energy, this can for example be provided by the stellar irradiation (in the form of UV radiation) or lightning discharges. Whether the strong UV radiation during flares of M dwarfs is harmful for the evolution of life has been a matter of discussion (Shields *et al.*, 2016; O'Malley-James and Kaltenegger, 2019).

Conclusions

The formation of life does not only require the presence of liquid water, but also nutrients, especially in the form of the elements CNS. This paper investigates the presence of these elements in aerial environments where water condensates are present. Therefore, we have introduced a new framework to extend the concept of habitability from the presence of liquid water to also include the presence and availability of nutrients. This new system of different nutrient availability levels includes the presence of water condensates and how many nutrients based on carbon, nitrogen and sulphur are available.

The application of these nutrient availability levels to 1D equilibrium chemistry atmospheric models which include the element depletion due to cloud formation shows that all three elements (CNS) are commonly present at the water cloud base. Our models also show that phosphorus is not commonly present in the atmosphere and could therefore be a limiting nutrient for the formation of life.

Supplementary material. The supplementary material for this article can be found at https://doi.org/10.1017/S1473550424000077

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