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Chemical synthesis of food from CO₂ for space missions and food resilience

Juan B. García Martínez^{a*}, Kyle A. Alvarado^{a,b}, Xenia Christodoulou^{a,c}, David C. Denkenberger^{a,b}

Affiliations:

- ^a Alliance to Feed the Earth in Disasters (ALLFED), Fairbanks, AK, USA
- ^b University of Alaska Fairbanks (Mechanical Engineering and Alaska Center for Energy and Power), Fairbanks, AK, USA 99775
- ° School of Business and Engineering Vaud (HEIG-VD), Avenue des Sports 20, 1401 Yverdon-les-Bains, Switzerland
- * Corresponding author: <u>juan@allfed.info</u>

Abstract

Recyclable food technologies are essential for long-term manned space missions. This research compares customary and alternative space foods to non-biological synthesis (NBS) systems using recycled CO₂. Using electrochemical conversion of CO₂ as a starting point, different carbohydrate synthesis routes are reviewed. Sugars and glycerol are considered as final products. Three roundtrip missions with 5 crew members and 3-year duration were analyzed: International Space Station, the Moon, and Mars. The equivalent system mass (ESM) technique was used to compare NBS systems to customarily storing prepackaged food, artificial-light grown Spirulina platensis, hydrogen-oxidizing bacteria (HOB), and microbial electrosynthesis (MES). This allows for a launch cost comparison of systems with different characteristics, including equipment mass, onboard volume, and power and heat rejection requirements. Power consumption was estimated via mass and energy balances using literature values. The Mars mission ESM of the NBS system is estimated within 10-30 tonnes. This was compared to an average of 65 t for Spirulina, 35 t for prepackaged food, 25 t for MES, and 11 t for HOB. NBS is estimated to be among the most energy efficient options, together with HOB and MES. Electricity-to-food conversion efficiencies of 10-21% and single-pass carbon yields up to ~70% are expected for an NBS system. Although NBS is not recommended over all alternatives (i.e., HOB), it is recommended over the prepackaged food and Spirulina benchmarks. These food production technologies could also help humanity survive extreme catastrophes.

Keywords

Equivalent system mass; Non-biological synthesis; CO₂ conversion; Formose reaction; Existential risk

Highlights

- A non-biological carbohydrate synthesis route from CO₂ is outlined using existing technologies.
- A closed-loop life support and food production system concept for space use is proposed.
- Electricity-to-food conversion efficiency of 10-21% appears achievable using glycerol.
- The non-biological system performs better than benchmarks, but is inferior to bacteria.
- These systems could contribute to increased food security and existential risk reduction.

Abbreviations

Acetic acid (AA)
Digestible sugars (DS)
Dihydroxyacetone (DHA)
Environmental Control and Life Support System (ECLSS)
Equivalent System Mass (ESM)
Formic acid (FA)
Formaldehyde (F)
Formose sugar mixture (FS)
Global catastrophic risk (GCR)
Glycerol (G)
Hydrogen oxidizing bacteria (HOB)
International Space Station (ISS)
Microbial electrosynthesis (MES)
Non-biological synthesis (NBS)
Single-cell protein (SCP)

1 Introduction

Food in space is currently delivered from the Earth requiring large amounts of fuel to transport. During missions on the International Space Station (ISS), fresh and prepackaged food is delivered regularly in the amount of 2,800 kcal/person/d [1]; or equivalent to about 0.70 kg dry carbohydrate/person/d. The cost of prepackaged food for manned space missions, such as to the Moon or Mars, would increase with mission duration since more food and therefore more fuel would be required. Since fuel is the dominant cost of a space mission [2], one method to reduce cost is producing food on location. This will become increasingly important as manned missions expand farther from the Earth [3] into the deep space region (over 2 x 10⁶ km away from Earth, such as Mars and beyond).

Artificial light grown vegetables are an apparent and popular space food solution [4–6]. However, phototrophs (light-consuming organisms), even if grown under ideal conditions in controlled growth chambers, are limited by many productivity factors. Such factors include photosynthetically active radiation relating to energy inefficiency, lack of gravity which may cause root, stem, or flowering complications, and ionizing radiation which could destroy cells or inhibit growth [3]. A similar argument can be made against much of the research in space food from photosynthetic organisms, as part of the biologically assisted life support system. For example, the Micro-Ecological Life Support System Alternative (MELiSSA) based on Spirulina platensis [7,8], managed by the European Space Agency. A recent study investigating food in space from hydrogen-oxidizing bacteria (HOB) [9] indicated that there are more promising alternatives than growing food with artificial light, which is not energy-efficient [10]. Space agencies such as the National Aeronautics and Space Administration (NASA) are looking for efficient food production methods to provide astronauts with caloric and nutritional requirements for long duration, deep space missions [11].

1.1 Non-biological synthesis of food

The aim of the present work is to characterize the potential of chemical synthesis, or more applicably non-biological synthesis (NBS), as a life support and food production system in space, and compare it to the customary system and other space food concepts. NBS is defined here as a process in which living organisms are not used in any way as a means to obtain the desired product. For example, neither fermentation as a unit operation nor the use of enzymes as reaction catalysts will be considered.

There are plenty of possible routes to obtain food compounds via NBS from only CO₂ and water (See Figure A1 for selected examples). We focus on three types of compounds that have been studied in the literature as potential NBS food products for space missions: sugars [12], glycerol (also known as glycerine), and fats [13,14]. Synthesis of sugars via formaldehyde using the formose reaction has been historically and extensively studied as a potential food synthesis method from CO₂ and water, with a particular interest in space applications [15]. Frankenfeld [14] studied the synthesis of fats from CO₂ and water (See Figure A1). He concluded that it appears technically feasible, but is too complex in comparison with alternatives as a food source for manned space missions. Thus, it will not be discussed further. He suggested the synthesis of glycerol as a much

simpler and likely more viable alternative. This was concurrently studied by Jagow [13], who concluded that the glycerol synthesis route proceeding via formaldehyde is more advantageous than the one proceeding via acetylene.

The NBS routes to food production covered in this work are summarized in Figure 1. The relevant synthesis routes for both sugars and glycerol proceed from a formaldehyde intermediate. Several methods are available for synthesis of formaldehyde from CO₂, some of which are listed in Figure A1. See [16,17] for more information. Electrochemical conversion of CO₂ to formaldehyde via formic acid is selected in this work, since formic acid synthesis from CO₂ has been proven to work efficiently at relevant scales for long time periods [18], and has recently become commercially viable [19].

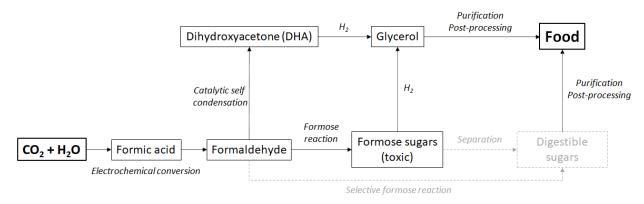


Figure 1. NBS routes for food production from CO_2 and water. The dashed grey lines represent currently unproven process steps.

1.2 Nutritional considerations

Sugars are an important and traditional source of energy in human diets, but glycerol has not been traditionally consumed in large quantities. However, it has been proven to be safe to eat. Glycerol is used extensively as a food ingredient thanks to several properties such as being nontoxic, digestible, environmentally safe, and providing a mild sweet taste, good flavour, and pleasant odor [20,21]. Glycerol is chemically a sugar alcohol, but is categorized by the U.S. Food and Drug Administration as a carbohydrate in terms of nutritional values [22,23]. It is processed by the human body via a different pathway than common sugar. Glycerol is first metabolized in the liver to glyceraldehyde 3-phosphate which can enter the same glycolytic pathway as the sugars. It reportedly boasts a calorie content of 4.31 kcal/g [24], slightly higher than the average carbohydrate, but the average caloric content for carbohydrates of 4 kcal/g was conservatively used.

Glycerol can be ingested in large amounts, and is known to be safe when administered in doses of <5 g/kg body weight/day [25]. This dosage is approximately equivalent to fulfilling ~70% of the daily caloric requirements of the average human or ~50% of those of an astronaut. Furthermore, no experiment has shown any nutritional dose-related adverse effects, with experts concluding that a numerical 'acceptable daily intake' value for glycerol is superfluous [21]. Indeed, it has been fed to levels up to 41% without apparent negative effects [26]. However, a small number of subjects do

show adverse effects to glycerol consumption [27]. Overall, glycerol seems safe to consume as a calorie source to satisfy a large fraction of the total caloric intake. It also is highly stable in the long term under ordinary conditions of storage and use, remaining free from objectionable color, odor or taste with the passage of time [21].

Naturally, the use of one compound as a single source of calories is not feasible for humans. This preliminary analysis is simply based upon setting a fixed amount of calories as a production target for the proposed technologies. Further research on combining various food production systems such as single cell protein (SCP), microbial electrosynthesis (MES) or NBS as part of the life support systems for a healthy diet can be important future research, but is beyond the scope of this analysis.

1.3 Aims and applications

This study is part of a series of works on the topic of life support and food systems for manned space missions. The previous research was based on identical premises to those proposed here using biological systems instead: food in space from HOB [9] and MES (Alvarado et al., to be published). This is essentially a comparison study of different technologies to the same end; overall results of the three works are compared here for all five space food options that have been reviewed, including prepackaged food and *Spirulina*. Regenerative life support and food systems based on technologies like these could not only reduce the cost, but also significantly increase the potential duration of manned space missions.

The food production methods from CO₂ proposed in these studies do not depend on agricultural inputs. For this reason, they are resilient to global catastrophic risks (GCRs) that inhibit agriculture, which can include supervolcanic eruptions, abrupt climate changes or a nuclear winter [28]. GCRs are described as posing a threat to humanity's well-being and potentially even to civilization's existence [29]. A parallel series of works covers the potential of these space food technologies as food sources during food-related GCRs [30,31]. Many other resilient alternative food production methods for GCRs are currently being investigated, including low-tech greenhouses [32], ramping up global seaweed production, producing sugar from lignocellulosic biomass [33], producing SCP from methane [34], leaf-protein concentrate production [35], or mushrooms and insects [36], among others.

Several of these alternative foods are already in use, or have been proposed as promising future foods for humanity. In fact, Dinger and Platt [37] have made the case for earthly production of carbohydrates via NBS on the grounds of sustainability. They concluded that NBS could eventually enable CO₂ capture and utilization at a global scale while producing carbohydrates at a lower environmental cost than traditional agricultural sources [37]. By virtue of consuming CO₂ as an input, these space food technologies could conceivably be applied for environmentally-friendly food production on Earth by acting as negative emissions technologies. Currently food systems contribute 19%–29% of global anthropogenic greenhouse gas emissions [38]. The food solutions proposed here could potentially reduce the amount of CO₂ that ends up in the atmosphere while also enabling humanity to feed more people around the world while using less water and land than conventional agriculture.

In addition, a closed-loop food production system with recycling of metabolic waste could be applicable to surface-independent refuges [39] on Earth or in space. These have been proposed as a means to increase resilience to extreme GCRs that could potentially threaten the survival and future of humanity, constituting an existential risk.

2 Methods

2.1 Calculation of equivalent system mass

Since the dominant cost of a space mission is fuel, and mass required for a mission is directly proportional to fuel requirement, space agencies including NASA employ an equivalent system mass (ESM) equation to compare between competing subsystem alternatives. Lower ESM values indicate lower overall launch costs. ESM is estimated using Equation 1, originally from [2] and adapted by Alvarado et al. [9] to fit a time-independent study.

$$ESM = L_{eq} * [M_I + (P * P_{eq}) + (C * C_{eq}) + (V_I * V_{eq})]$$
 (1)

The initial apparent mass M_{ll} represents the physical mass of the food production subsystem (i.e., reactor, reaction medium, separation equipment, water, pumps, piping, power supply, and other auxiliary equipment) at the beginning of the mission. The subsystem's power requirement P and cooling (or heat rejection) requirement C are the electrical and thermal demands, considering a closed system analysis. The initial pressurized volume requirement V_I represents the onboard volume occupied by the subsystem's apparent mass. Mass equivalency factors for power Peq, heat rejection C_{eq}, and volume V_{eq} represent the mass per unit provided by the parameter and originate from NASA's Life Support Baseline Values and Assumptions Document [1]. For example, the nuclear reactor selected as the power source for NBS measures 76 kg for every kWelectrical provided. The product of the power requirement P (in $kW_{electrical}$) and the equivalency factor P_{eq} (in $kg/kW_{electrical}$) is kilograms, such that all parameters are converted to mass units for the final summation. The location factor L_{eq} is the final step and is multiplied by the summation of parameters. It is dependent on vehicle and location and is equated relative to the fuel requirement needed to launch from the Earth's surface to Low Earth Orbit [1]. Location factors were derived for the ISS mission, the Moon mission, and the Mars mission distinctly, being 1.0, 16.6 and 14.1 respectively [1]. All ESM values were estimated for 5 crew members and a duration of 3 years.

2.2 Non-biological synthesis scheme and energy efficiency

Power consumption and cooling requirements are major contributors to the ESM of the combined food and life support system. These will depend on the electricity-to-food conversion efficiency of the system, defined as the amount of electrical energy needed to be invested in the overall process to obtain a unit of food energy. This in turn depends on the overall reaction yield and energy consumption per product unit of each chemical conversion step. Additionally, separation processes such as product purification and capture of CO₂ from the spacecraft's atmosphere must also be accounted for. The carbon yield is defined as the share of carbon mass present in the

carbon-containing reactant that will become part of the target reaction product for a given conversion step or series of steps.

To the best of our knowledge, there are no reports of industrial or pilot systems that have non-biologically produced digestible material from CO₂, although Air Company claims to have achieved this [40,41]. There is significant uncertainty about the performance that such a system would have, which should be accounted for. To this end, two scenarios are proposed in this analysis. First, a "base case" scenario aiming to conservatively characterize the current state of the technologies involved in each step as shown in Figure 1. Second, a "high end" scenario describing reasonable upper bounds of the potential of current or conceivable future developments.

A literature review was conducted to find the expected carbon yield and associated energy consumption values of the chemical process steps involved in the proposed food production routes, as summarized in Tables 1-2 and detailed in Section 3.1. These were used as the basis for a mass balance analysis (see Table A1) and to finally estimate the energy consumption of each route. All food production routes use the CO₂ captured from the spacecraft's atmosphere as a starting material for synthesis of the formaldehyde intermediate. CO₂ is obtained via the CO₂ removal assembly present as part of the Environmental Control and Life Support System (ECLSS). The energy consumption of CO₂ capture is estimated based on a 20% thermodynamic efficiency from the ISS CO₂ capture system [42].

Table 1. Expected overall reaction yields of the chemical conversion steps involved in the proposed CO_2 to food schemes. Where: FA refers to formic acid, F to formaldehyde, FS to formose sugars, DS to digestible sugars, DHA to dihydroxyacetone, G to glycerol, X to conversion, and S to selectivity (defined as the amount of desired product formed per unit of reactant consumed).

Process		Carbon yield (overall)					
step	Symbol	Base case	High end	Discussion			
$CO_2 \rightarrow FA$	Yfa	90%	100%	Based on high reaction selectivity and high CO ₂ recyclability. Base case value based on somewhat imperfect selectivity and/or CO ₂ recycling.			
$FA \rightarrow F$	Уг	51%	80%	Base case [43]: $X_{\rm FA}$ = 64% (based on single pass conversion), $S_{\rm F}$ = 80%. High end: $X_{\rm FA}$ = 100% (complete overall conversion), $S_{\rm F}$ = 80%.			
$F \rightarrow FS$	Yfs	60%	96%	Base case: [12]. High end: [44].			

FS → DS	Yds	35%	100%	Base case: complete separation of the digestible sugars from the formose mixture. High end: perfect selectivity towards digestible sugars.
$FS \rightarrow G$	YG,s	30%	70%	Base case [45]: X_{FS} = 90%, $S_{G,S}$ = 33%. High end [46]: X_{FS} = 70%, $S_{G,S}$ = 100%.
F→ DHA	У DHA	85%	95%	Base case [44]: $\gamma_{DHA} = 85\%$. High end [44]: $X_F = 99\%$, $S_{DHA} = 96\%$.
DHA → G	Y _{G,DHA}	90%	90%	[47].
$CO_2 \rightarrow DS$	Yfa Yf Y	10%	77%	Base case assumes a selective separation of digestible sugars from the mixture, which is not straightforward. High end assumes a yet unproven selective reaction.
$CO_2 \rightarrow FS \rightarrow$ G	Уға ⁻ Уғ ⁻ Уғѕ - У _{G,} ѕ	8%	54%	Circumvents challenges of obtaining digestible material from formose sugars by converting them to glycerol.
$CO_2 \rightarrow DHA$ $\rightarrow G$	Yfa Yf Yd ha Yg,dha	35%	68%	More efficiently circumvents challenges of the first route by selectively targeting glycerol.

Table 2. Breakdown of energy consumption values by step. Energy consumption is given per unit of reaction product for the relevant conversion step. Some values differ for the base case and high end scenarios.

Step	Energy consumption (kWh/kg step product)			
CO ₂ capture	0.7			
$CO_2 \rightarrow FA$	5.4			
$FA \rightarrow F$	4.1			
$F \rightarrow DS$	6.8-13.6			
$F \rightarrow G$	4.2-17.9			

3 Results and discussion

3.1 Non-biological synthesis routes and power requirements

Three NBS food production routes from CO₂ via formaldehyde are reviewed here: A) formose sugar synthesis and purification to obtain digestible sugars, B) hydrogenation of a formose sugar mixture to glycerol, and C) selective formation of glycerol via DHA. Base case and high end of performance scenarios are proposed for all three routes. A simplified block diagram of the three proposed NBS food production routes is found in Figure 2.

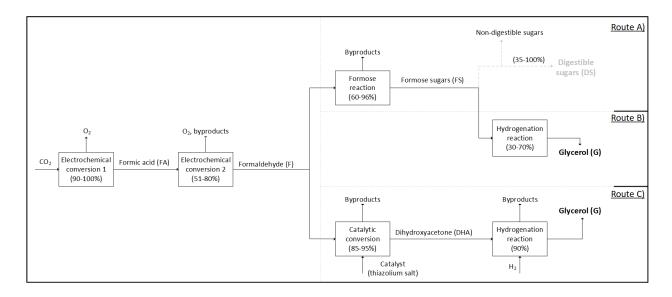


Figure 2. Simplified block diagram of the proposed NBS routes for food production from CO_2 . Separation steps, recycles and reaction media are not shown. The percentage values inside the blocks express the expected carbon yields. The bolded words indicate target food products. The dashed grey lines represent currently unproven process steps.

Results of the analysis of the expected performance of NBS systems are shown in Figures 3-4. Figure 3 shows the breakdown of the resulting energy consumption values, and Figure 4 shows the overall electricity-to-food efficiency of each route. We do not ultimately choose a specific route towards food compounds, rather we use the ones described in this section to illustrate the range of key performance values that can be expected from an NBS system. Results (see Figure 4) suggest that an electricity-to-food efficiency of \sim 10% is achievable via NBS for the current state of the technology (based on the base case for glycerol via DHA). We expect electricity-to-food efficiency values of \sim 21% may be achievable via either optimal designs with current technology or via application of conceivable future technologies (based on the higher ends). Note how synthesizing formaldehyde from CO_2 is the largest contribution to energy consumption. This serves as a

depiction of the potential of NBS for food production based on the literature, it does not ensure that these levels of performance will be achieved.

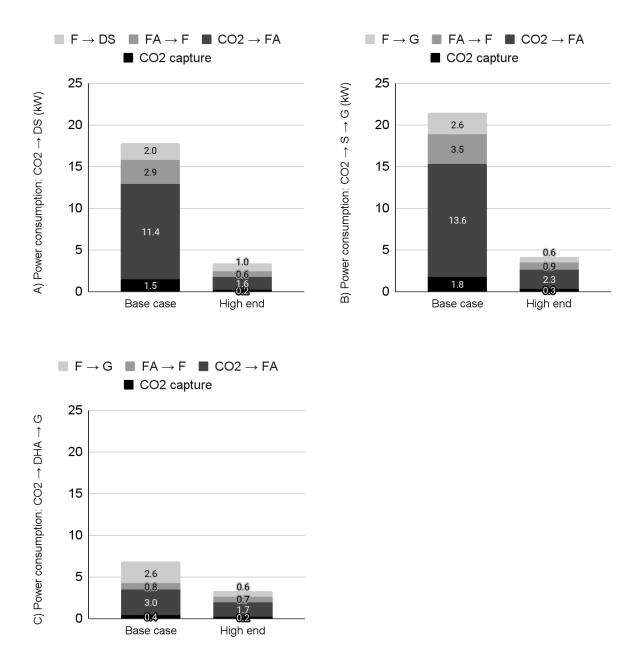


Figure 3. Expected power consumption of the three proposed routes from CO_2 to food for continuously feeding a crew of 5. All values given in kW, for fulfilling a required production of 0.70 kg dry carbohydrate/person/d. From left to right the graphs represent the following routes: A) CO_2 to sugars, B) CO_2 to glycerol via formose sugars, C) CO_2 to glycerol via DHA.

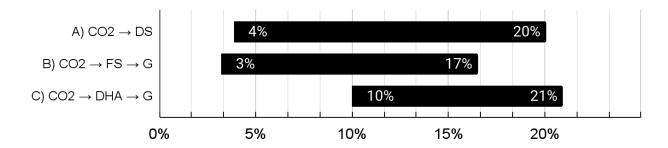


Figure 4. Estimated values of expected electricity-to-food efficiency for the three proposed routes from CO_2 to food. The right side represents the high end while the left side represents the base case or low end of efficiency.

The electrochemical conversion of CO₂ to formaldehyde (F) as proposed here proceeds via formic acid (FA). Single pass yield values of electrochemical conversion of CO₂ to FA close to 30% have been found in experimental literature [18]. Presumably, when recycling unreacted CO₂, conversion would be greatly boosted. Since this is a selective process, we state a high value of the overall yield between 90-100% for this step. An energy consumption of 5.4 kWh/kg FA is expected [18]. State-of-the-art systems have demonstrated commercially relevant activity and stability up to 1,000 hours, but catalytic stability still requires further improvement [19].

Regarding synthesis of F from FA, Masel et al. [43] described an example of a system converting FA to F with 64% conversion and 80% selectivity, which we use as base case. For the high end, we propose that conversion could conceivably be boosted up to 100% by using a technology allowing for higher conversion and/or a selective separation with subsequent recycling of FA for an 80% yield. The energy consumption of this step is estimated based on an expected hydrogen requirement of 1.25 mol H₂/mol F and a typical water electrolysis electrical efficiency of 80%.

3.1.1 Historical NBS route: synthesis of sugars via the formose reaction

Route A, from formaldehyde to sugars, is based on the formose reaction, which has historically been studied as a sugar production method for space missions [15]. The formose reaction is expected to produce sugars with four to seven carbon atoms [48], comprising a notoriously complex racemic mixture [49]. A racemic mixture contains approximately equal amounts of D and L sugar enantiomers (mirror molecules), of which only the former, known as biotic enantiomers, is digestible by humans. In addition, this formose mixture potentially contains all possible sugar stereoisomers (3-dimensional configurations) [37]. This causes significant challenges regarding its use as a food production method: a) abiotic enantiomers such as L enantiomers (expected to comprise ~50% of the mixture) appear to be non-digestible by humans, b) some of these, such as L-glucose, are known to produce strong purgative effects in the human digestive system [50], which is counterproductive to our purposes. The resulting formose sugar mixture (named FS from here on) reportedly led to linear weight loss and eventually premature death in animal testing, whether it comprised 100% of the diet [12] or 50% [48]. This is consistent with the purgative effect of L-glucose, although Akerlof [12] hypothesized that it could be caused by the presence of

aldehydes and alcohols in the FS. The presence of branched sugars has also been suggested as potentially problematic [48].

Two possibilities have been identified in literature to circumvent these challenges: (1) targeted stereoisomer synthesis towards digestible sugars (DS) and (2) optimization of process conditions followed by subsequent separation and reprocessing of the undesired stereoisomers [37]. Regarding (1), the consensus seems to be that it is extremely difficult to synthetize DS with acceptable selectivity, see [15,51]. However, Air Company claims to have recently achieved DS synthesis via a new chiral catalyst that "takes advantage of the isomerization of formaldehyde" to produce a mixture of "primarily d-glucose and d-ribose along with d-fructose as a minor component" [40,41]. Regarding (2), a proposed separation route is chromatographic separation of a racemic mixture of selected isomers from the FS followed by an enantiomeric separation of the biotic D enantiomers [52]. However, no enantiomeric separation process has yet been proven to produce digestible material from a formose mixture based on current research. Further discussion of the challenges involved with the formose reaction as a food production method can be found in [37]. Regardless of the challenges of this route for food production, its potential has been estimated for comparison with the other routes proposed.

Experimental studies on the formose reaction reported yields of 60% [12], while some researchers claim up to 96% [44]. Based on [12], we estimate a typical formose mixture to contain 35% as many calories as a typical sugar compound containing 4 kcal/g (see Table A2). This stems from the presence of nondigestible and low-calorie sugar compounds. A subsequent selective separation of the digestible sugars is assumed. The high end assumes a (currently nonexistent) method to selectively produce digestible, high calorie sugars from the formose reaction.

No published values for the energy consumption of the separation of digestible sugars from a formose sugar mixture were found in literature. Instead, the higher end of energy consumption proposed here is based on the experimental result of Akerlof [12] for production and purification of FS (including non-digestible sugars). The lower end assumes half as much energy consumption, since selectively converting formaldehyde to calorie-rich sugars requires minor or no purification. The resulting energy consumption values can be found in Figure 3A. The resulting range of expected electricity-to-food values is 3.8%-20.1%. For comparison, the experiments of [12] took 14.3 kW to produce 3.5 kg/day of FS destined to feed a crew of 5. Based on the proposed FS caloric content of 1.39 kcal/g we estimated from these results, this would be equivalent to at most a \sim 1.7% electricity-to-food conversion efficiency. The difference compared to the expected values is not surprising given how technologies have improved in the last decades, for example the multiple new technologies and efficiency improvements for conversion of CO₂ to formaldehyde in the last decade [17–19,43,53]. The single-pass carbon yield to DS was 10.5%, similar to the base case of route A.

3.1.2 Alternative NBS routes to glycerol

Route B proceeds to glycerol (G) via hydrogenation of a formose sugar mixture. It can be considered a way to food production that circumvents the absence of formose synthesis methods selective towards DS, as well as the need for complex separation schemes to obtain the DS from the FS for a non-selective formose reaction. The base case yield comes from [45], who reports >90%

conversion of a sugar mixture with a 33% selectivity to glycerol for a 30% yield. The high end yield is based on [46] reports of up to 70% conversion of pure sorbitol to primarily glycerol.

Routes A and B could theoretically be combined, by hydrogenating any non-digestible sugars separated from FS to glycerol. The electricity-to-food efficiency value of this combined route would necessarily be within the interval estimated for the first route, since it cannot surpass the efficiency of selective formation of DS nor underpass the efficiency of the base case since it does not include any byproduct reuse.

Route C aims to characterize a selective route from formaldehyde to glycerol which appears feasible with current technology. It can be seen as a way to overcome the obstacles of route A in a more efficient way than route B. The formose reaction can be leveraged for targeted synthesis of DHA, which can be a valuable intermediate for selective synthesis of glycerol. Experiments have shown a DHA yield of 85% with a formaldehyde conversion of 99%. Under optimized conditions, the production of DHA from formaldehyde has a selectivity of 96% [44]. Thus, an overall yield between 85-95% is expected. Gracey et al. [47] reported maximum values of 100% conversion and 90% selectivity for DHA to glycerol.

The energy consumption of synthesizing glycerol from formaldehyde is considered similar for the second and third routes. The lower end is based on the energy requirement of glycerol synthesis from sugars proposed by Frankenfeld [14]. The higher end is conservatively estimated by adding the experimental formose reaction energy consumption of Akerlof [12] to the previous value. The resulting energy consumption values estimated for routes B and C can be found in Figures 3B and 3C, respectively.

3.2 Carbon cycling

The spacecraft is a closed system, and any unwanted reaction byproducts would need to be either ejected, similar to the methane byproduct of the Sabatier system in the ISS [54], or recycled. If choosing to eject the byproduct mass, that would require additional water and carbon mass at launch to compensate for the mass losses, for example in the form of prepackaged food. We do not calculate the ESM for this possibility, since this is a comparison study and mixed options would not make for a fair comparison. We leave possible combinations of the different space food systems for further research. Thermochemical processes (i.e., combustion, gasification or pyrolysis) could be a way to recycle the organic byproducts of the NBS system. Gasification or pyrolysis would mainly produce CO and H₂, which could be useful resources. However, these processes typically involve complex product profiles (potentially including e.g. methane or solid carbon residue), making the resulting stream difficult to recycle completely.

For simplicity, we estimate the ESM for an option in which byproducts are subjected to complete combustion, yielding only water and CO_2 by using a large enough excess of O_2 . This would be followed by condensation of the water and capture of the CO_2 which gets recycled to the feed of the NBS system. The unreacted excess O_2 gets recycled back into the combustion chamber to keep the combustion going. If feasible, this could allow for complete carbon and water recycling, which would make for a closer comparison with the microbial food production systems (Alvarado et al., to be published), [9]. The energy produced by combustion is conservatively neglected. Figure 5

presents a diagram illustrating the carbon recycling concept for route C. Note that the mass flows are shown in proportional terms, making the diagram applicable to the concept at any scale. For example, a large scale production system on Earth or another planet. Equation 2 shows the overall chemical reaction from CO₂ and water to glycerol via DHA (route C), disregarding side reactions.

$$3CO_2 + 4H_2O \rightarrow C_3H_8O_3 + 3.5O_2$$
 (2)

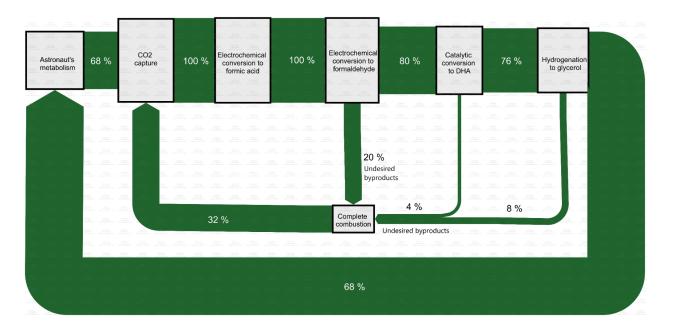


Figure 5. Carbon flows of the proposed closed-loop system for food production using NBS of glycerol via DHA (Route C, with high end carbon yields). Note: complete carbon recovery from metabolic waste is presumed.

Stable food production over time can be achieved with complete carbon recycling; only a small amount of raw materials would need to be included at the start of a mission. The vast majority of the carbon consumed by the astronaut's metabolism is exhaled in the form of CO₂ [55], and is captured by the spacecraft's air purification system. If total recycling efficiency of carbon from CO₂ and other metabolic waste is achieved, then all carbon produced by the astronauts' metabolism could be recycled by the NBS system, resulting in a closed-loop carbon cycle.

Complete water recycling could similarly be achieved by recovering the water produced by the astronauts' metabolism and the combustion system. CO₂ and water may also be available from local mission sources, particularly on Mars, which could be used as supply to counter any mass losses if the loop is not completely closed.

3.3 Other NBS process considerations

This work is based on synthesis of formaldehyde via electrochemical conversion, but other routes from CO₂ to formaldehyde could also be promising for space applications. Examples of recent advances include a method for selective low-temperature one-pot catalytic conversion [56] and a new catalyst for synthesis under ambient conditions [57]. CO₂ photoreduction to formaldehyde is well studied in literature [17] compared to the proposed route, but was not selected due to its dependence on sunlight which could be problematic in dark periods. Keeping the system independent of sunlight allows for continuous oxygen production, making it easier to integrate with life support systems.

The final food product must be safe to eat, virtually free of toxic material. For example, the presence of homogeneous catalysts in the food must be minimized. Selective conversion of formaldehyde to DHA is based on the use of thiazolium salts as homogeneous catalysts. Heterogeneous catalysts are preferable for easier separation, but experiments with immobilized thiazolium salts resulted in notably lower efficiency and selectivity [51]. Further research is needed on the topic of separation and purification methods for the proposed NBS systems. In-depth engineering design of the system would be required to ascertain the actual energy usage.

A potential alternative way to overcome the challenges of non-biologically producing safer and more nutritious food could be growing microorganisms which efficiently assimilate the NBS products for food production as [13] suggested, but it is outside the scope of this NBS-focused analysis.

3.4 Equivalent system mass contributions

Given that the proposed NBS systems have not been modelled in-depth in this work, no design parameters are available for the system. We have chosen to represent ranges of values considered reasonable in the ESM calculation: a) between 10-21% electricity-to-food efficiency, b) 200-1,000 kg of setup mass and c) 0.2-1 m³ of volume (corresponding to an overall system density of 1,000 kg/m³). The amount of additional starting material such as catalysts that need to be packed from the start of the mission would depend on the separation and recycling efficiencies of the steps in which they would be involved, but can be considered negligible in comparison to the rest of the contributions. Operation, maintenance, and cleaning of the NBS system are also neglected.

Table 3 contains the ESM values estimated for these proposed values of electricity-to-food efficiency, setup mass, and volume. Energy use is the major contributor to total ESM, at 43-73% (includes power consumption and heat rejection requirements). The actual mass and volume of the equipment required by a given NBS life support and food system would depend on the number and type of unit operations required, their efficiencies, and the amount and types of reaction byproducts and waste products. Further research is needed on these aspects for precise estimations of NBS ESM, in particular engineering design.

Table 3. NBS ESM values estimated for both ranges of the uncertainty intervals in the three different space mission scenarios reviewed.

Apparent mass (kg)	System volume (m³)	Electricity-to- calories efficiency	ISS ESM (kg)	Moon ESM (kg)	Mars ESM (kg)
200	0.2	20%	775	11,221	9,688
1000	1	10%	2,244	33,940	30,267

3.5 Alternatives comparison

In order to characterize the potential of NBS as a life support and food system for manned space missions, the results of this work are compared in this section to other similar concepts based on different technologies for the same mission scenarios based on the ESM methodology. The relevant ESM values for the benchmark options (prepackaged food, *Spirulina*) and for HOB-SCP were estimated in [9]; the ESM of MES-AA in (Alvarado et al., to be published). ESM is not the exclusive metric for a comparison study since it lacks considering reliability, safety, and performance [58]; however, it is pivotal for evaluating cost.

Given the major contribution of power consumption and heat rejection to the system ESM, it is worth comparing the food production systems in terms of electricity-to-food efficiency alone. Figure 6 shows how NBS is expected to be on average less efficient than HOB-SCP and MES-AA, while all are vastly more efficient than artificial-light-based *Spirulina*. As long as the energy consumption of formaldehyde synthesis from CO₂ stays high, HOB-SCP and MES-AA will likely dominate in terms of energy efficiency. It is worth noting that the expected efficiency of MES-AA is 19.8%, higher than the average value shown in Figure 6 (Alvarado et al., to be published). The other major ESM contribution besides energy is the system mass. The contribution of system volume is minor (1-10% for all alternatives and mission types).

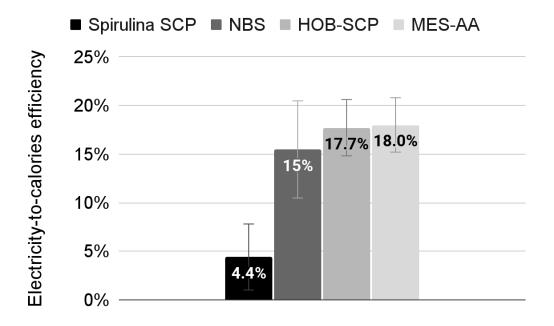


Figure 6. Comparison of expected electricity-to-food efficiency values of NBS with various food and life support options previously assessed in literature.

Regardless of the actual final values of mass and volume of the NBS systems, some conclusions can be derived from the estimated range of electricity-to-food efficiency values. In terms of ESM for the proposed mission scenarios, NBS compares unfavorably to HOB-SCP, since the expected ESM is higher except for the most optimistic scenario of highest efficiency and lowest system mass and volume. It compares very favorably with the prepackaged food and Spirulina benchmarks. Comparisons of NBS with MES are within the bounds of uncertainty. Figure 7 shows the ESM values estimated for the three space mission scenarios proposed, while Figure 8 presents a breakdown of the expected ESM contributions for the Mars mission. Both figures present ESM results estimated from the mean values of the uncertainty ranges for all five alternatives. The order of preference between alternatives is the same for the different missions in terms of ESM, as the ESM comparison between alternatives is similar for all three proposed scenarios.

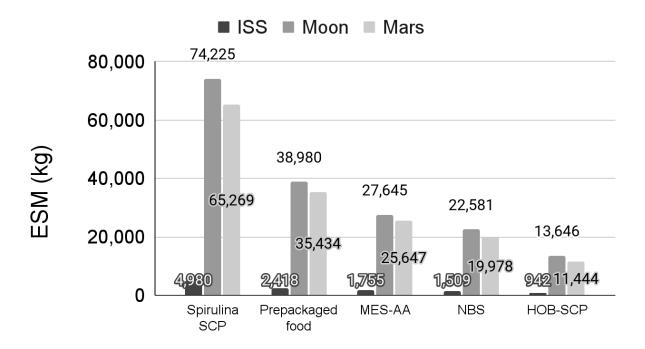


Figure 7. ESM comparison of NBS with various life support and food systems previously assessed in literature for continuously feeding a crew of 5 during a 3-year mission. Results are shown for the average values of the ESM uncertainty ranges.

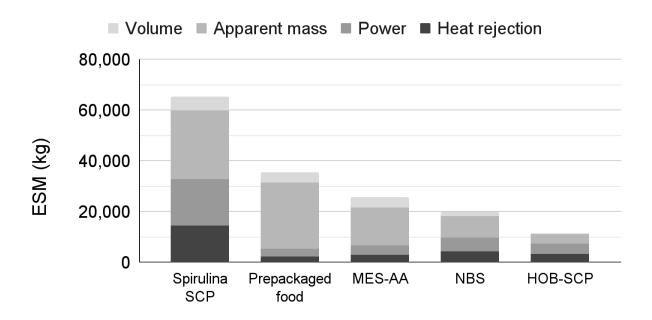


Figure 8. Breakdown of the average ESM contributions in the Mars mission scenario for the various food and life support options previously assessed in literature for continuously feeding a crew of 5 during a 3-year mission. Results are shown for the average values of the uncertainty ranges.

Both HOB-SCP and Spirulina SCP are more nutritionally complete food options compared to NBS-based glycerol, but can be complementary. These microbial food products contain valuable assortments of macronutrients and micronutrients [59,60], while glycerol acts nutritionally as a caloric contribution (see Section 1.2). A constant stream of nutrients is needed to keep the microbial cultures productive, while NBS requires catalytic materials. An advantage of NBS systems could be increased resilience or even immunity to contamination from stowaway microorganisms, which is a known concern [61].

It is worth noting that the reason why ESM results are higher for the Moon mission compared to the Mars mission is because the latter has a lower location factor for ESM estimation, as found in NASA's Life Support Baseline Values and Assumptions Document [1]. The location factors for each mission originate from different shuttle systems, propulsion types and transportation history. This being a comparative analysis, the accuracy of individual location factors for each mission does not impact results significantly, since they are consistent between food alternatives. However pivotal ESM is as a cost metric, a more in-depth comparison study would include other reliability, safety, and performance metrics.

3.6 Significance and future work

This comparison analysis can help inform future decisions on life support and food system concepts for long-term manned space missions. The availability of multiple food options can contribute to diet diversity in space. The potential of combining different options for increased nutritional quality and lower cost could be an attractive avenue for future research on the topic. Nonbiological synthesis of carbohydrates could complement the low-carbohydrate HOB-SCP option. Currently, there is greater uncertainty on the performance of the proposed NBS life support and food systems compared to the microbial systems. Front-end engineering design would allow for estimating NBS key performance values with more certainty. Then, laboratory and field testing would be required to establish feasibility prior to use in deep space missions. Further fundamental research on NBS of food could make it more competitive than the microbial food options. In particular, reducing the energy consumption of converting CO₂ to formaldehyde could make NBS the best alternative in terms of ESM and thus cost, given how this is the most energy-intensive part of the process. For example, slashing the energy consumption of this step by half could potentially bring the upper-bound estimate of the electricity-to-food efficiency from ~21% up to ~33%. This is an active research area with significant advances taking place [19,56,53], which foreshadows a promising future for the NBS food option.

This work's results are also relevant outside of the topic of life support. The intermediate and final products and byproducts could be potentially used as chemical building blocks in space for chemical synthesis or biomanufacturing of other useful resources. While not yet applicable, further research on NBS of carbohydrates, whether for use in space missions or as a sustainable food production method, could enable its application in food-related GCRs on Earth. Additionally,

research on integration of closed-loop food production systems with surface-independent refuges could contribute to existential risk reduction.

4 Conclusions

A practicable route towards synthesis of carbohydrates from CO₂ has been proposed, primarily for use as part of a life support and food system for long-term manned space missions. A closed-loop food production system based on glycerol appears viable using existing technologies. electricity-to-food conversion efficiencies of 10-21% seem achievable, similar to other promising alternative space foods such as HOB-SCP or MES-AA.

This comparison analysis can help inform future decisions on life support and food system concepts. The ESM analysis indicates that NBS would beat the benchmark space food production technologies in terms of launch cost; namely prepackaged food and *Spirulina*. HOB is superior to NBS in terms of ESM and nutritional quality, while MES-AA performs similarly to NBS on these criteria. Future research on combining these options could contribute to achieving nutritional quality and diet diversity in space at a lower cost than the current space food standards.

These concepts for food production from CO₂ could also contribute to sustainability, to food security during extreme global food catastrophes, and to societal resilience against existential risk if applied to isolated refuges.

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Appendix A) Expanded NBS scheme

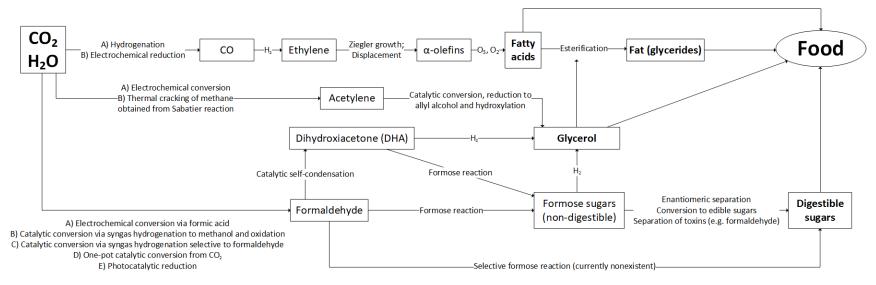


Figure A1. Expanded scheme including the different types of proposed NBS foods for space missions found in literature and a selection of their possible synthesis routes, as well as several alternative options to obtain formaldehyde from CO_2 apart from the electrochemical route followed in this work.

Appendix B) Mass balances

Table A1. Mass balances of the three proposed reaction routes and the base case and high end scenarios. Mass flow rates given in kg/day. Where: FA refers to formic acid, F to formaldehyde, FS to formose sugars, DS to digestible sugars, DHA to dihydroxyacetone and G to glycerol.

Reaction route	A) CO₂ → Dig sugars	gestible gestible		·	B) CO₂ → Formose sugars → Glycerol			C) $CO_2 \rightarrow DHA \rightarrow Glycerol$	
Scenario	Base case High end		Base case	High end			Base case	High end	
Digestible									
sugars	3.5	3.5	Glycerol	3.5	3.5		Glycerol	3.5	3.5
Formose sugars	10.1	3.5	Formose sug	gars 12.1	5.2		DHA	3.8	3.8
Formaldehyde	16.9	3.7	Formaldehyd	de 20.1	5.4		Formaldehyde	4.5	4.0
Formic acid	50.6	7.0	Formic acid	60.3	10.3		Formic acid	13.5	7.7
CO ₂	53.7	6.7	CO ₂	64.1	9.8		CO ₂	14.4	7.4

Appendix C) Estimation of the nutritional value contained in a formose sugar mixture

The caloric content of a formose sugar mixture was estimated at 1.39 kcal/g. First, the caloric content of each component was found in literature. Then, the weighted calorie content was calculated by multiplying the caloric content of each component by its weight concentration in the mixture and by a factor of 50% to account for the indigestibility of L-enantiomers (See Section 3.1.1). The sum total of the weighted calorie content results in 1.39 kcal/g of mixture, or 35% as many calories as a typical pure hexose sugar of 4 kcal/g.

Table A2. Composition of a formose sugar mixture (adapted from [12]) and expected caloric content of the components.

Component number	Weight (%)	Туре	Possible identity	Caloric content (kcal/g compound)	Weighted calorie content (kcal/g)
1	0.4		glycollic aldehyde	0	О
2	1.1		glyceraldehyde	0	0
3	1.8	triose	dihydroxyacetone	0	0
4	3.2	tetrose	erythrose, threose,erythrulose	0	0
5	2.5		xylulose	0	0
6	6.5	pentose	ribose, dendroketose	4	0.13
7	17.2	pentose	xylose	2.4	0.2064
8	16.5	pentose	fructose, mannose	4	0.33
9	17.5	hexose	sorbose, arabinose	1.52	0.133
10	16.8	hexose	glucose	4	0.336
11	8.5	hexose	galactose	4	0.17
12	4.4	hexose	unidentified	4	0.088
13	2.0	hexose	heptulose	0	0

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