BIOLOGICAL PROCESSES WITHIN ATMOSPHERIC AEROSOLS v1.0

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ABSTRACT

A surprisingly high fraction of sunlight is absorbed during transit of the Earth's lower atmosphere. One third to one half of this absorption may be unexplained, referred to as the 'atmospheric absorption anomaly' or 'cloud absorption paradox'. This quantity is comparable to the total amount of sunlight falling on known photosynthesising organisms.

This paper sets out the hypothesis that the anomalous absorption is a consequence of biological activity within atmospheric aerosol droplets. Bacterial spores and the recently discovered vesicles emitted in huge numbers by marine bacteria are possible candidates for growth and maybe even reproduction within such droplets. Ways in which, within a droplet, a microorganism can harvest energy while avoiding any significant UV dose are described.

While at present hypothetical, the processes described could resolve several current anomalies, including a major puzzle concerning the origin of life. Practical implications could be relevant to anthropogenic global warming, attempted geoengineering, and photosynthetic energy harvesting. The hypothesis could be tested at low cost.

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1. INTRODUCTION: THE BASIC PHYSICS

Given the transparency of air and water to visible light, a surprisingly high fraction of sunlight is absorbed during transit of the Earth's lower atmosphere^[1], as shown in Table 1. This is sometimes described as the 'atmospheric absorption anomaly' or 'cloud absorption paradox'. While the issue remains highly controversial and has generated an extensive literature, the discrepancy may be considerable, $\sim 25 \text{ W/m}^2$ of the Earth's surface.^[2] At ~13 PW, this quantity is 200× greater than the net carbonfixing output of photosynthesising organisms: comparable to the total amount of sunlight falling on all surface level vegetation.

Table 1	Table 1Sunlight energy averaged over whole Earth surface			
		W/m2	%	Total PW
Incoming sunlight from space		340	100	173
Reflected into space		99	29	50
Absorbed	by ozone layer	15	4.5	8
Absorbed in lower atmosphere		63	18.5	32
(of which a	anomalous)	~25	~7	~13
Absorbed at surface		163	48	83
Photoauto	trophic carbon output	0.24	.07	0.12
(Photoautotrophic carbon output is 100-115 billion tonnes/C annum				
$\sim 3.5 \times 10^6$ kg/sec @ 33 MJ/kg => 115 TW output energy.)				

Atmospheric water (vapor plus droplets plus ice) is equivalent to a layer of water about 1 inch thick covering the Earth's surface, 12.5 trillion tonnes. This is an order of magnitude greater than the total of all known biomass, which is of the order of one trillion tonnes. At any given time, a

substantial proportion of this water is in the form of liquid droplets of size broadly comparable to bacteria, with diameters in the range 1-100 microns.

Such droplets normally form around airborne particles called CCNs, Cloud Condensation Nuclei. These are typically ~0.2 microns diameter, whereas a typical droplet is ~20 microns diameter. Some biogenic particles act particularly efficiently as CCNs,^[3] so will have a strong tendency to become encased in a water droplet.

A wide variety of biological entities can readily become airborne and find themselves in the interior of a water droplet. As explained in section 3 below, while airborne particles on their own suffer intense sunlight including UV, within a water droplet there are locations where this flux is comparatively negligible.

This paper sets out the hypothesis that the anomalous absorption is a consequence of biological activity within atmospheric water droplets.

2. AEROSOL SEEDING CANDIDATES

One likely candidate for colonising an atmospheric droplet is a bacterial spore. These vastly numerous but poorly characterised entities range in shape and size from ~ 1 micron diameter spheres to $\sim 0.5 \times 2$ micron ellipsoids. An outstanding characteristic of bacterial spores is that, in contrast to most other living material, they are highly resistant to ultraviolet light, so could survive being airborne on their own until they encounter a droplet or one forms around them.

A more speculative but very promising candidate is the 'vesicles' which have recently been discovered to be emitted in vast numbers from marine photoautotrophic cyanobacteria.^[4] With a diameter of ~0.1 microns, these vesicles appear to be the right size to be efficient at droplet nucleation, while also just large enough to contain both the bacterium's complete DNA and a supply of elements essential for growth which would not be available in an airborne water droplet of marine origin. Thus the vesicle could in effect be a minimal size spore. The vesicle would not survive unprotected ultraviolet exposure: however it will become airborne in the

fully saturated air layer just above the sea surface, likely in a droplet of sea spray, so can be housed in a protective droplet for all or almost all of its flight.

3. AEROSOL DROPLET INTERIOR

It is normally assumed that airborne spores must remain spores until they alight somewhere more hospitable. However this ignores the internal optics of aerosol droplets. The ray paths are sketched in Figure 1. As water has refractive index 1.33, the front surface of the droplet acts as a lens with focal length $\sim 3 \times$ the droplet radius, and internal reflection from the back surface follows.

Figure 1



Two regions are of particular interest.

- 1. In the toroidal 'safe zone' where sunlight intensity including UV is negligible, a spore can safely unpack itself and become active.
- 2. By contrast the central high-intensity region marked with a red spot will act as a kind of 'stomach': organic materials of non-trivial

molecular weight will tend to have their chemical bonds rearranged, and often be broken down into smaller components.

Consider a spore which has acted as a nucleation point and formed such a water droplet around it. Forces potentially acting on the spore include surface tension, modified by any temperature and chemical gradients present, light pressure and electric fields. If the spore can position itself within the safe zone, either by harnessing these forces to remain there at all times, or simply by moving rapidly whenever it is in high sunlight intensity, its survival time is greatly increased.

Within a droplet, a spore has access to unlimited carbon, hydrogen, oxygen and nitrogen, from water and dissolved atmospheric gases. A droplet of marine origin will also contain significant amounts of the other elements most necessary for life, with the exception of phosphorus.

As the refractive index of water is higher for the more damaging UV than for visible light, the spore may be able to position itself at the edge of the safe zone so as to receive a benign spectral distribution, permitting photosynthesis without excessive UV.

The stomach zone can also be used to harvest energy cyclically. Suppose the spore obtains energy by joining components of low molecular weight into a product of higher molecular weight. In the stomach, the product of high molecular weight (which might be relatively opaque) gets broken down again: the cycle can harvest energy indefinitely using the same material.

(Note that thermal harvesting as described in my related paper^[5] would be relatively ineffective: the thermal capacity of the droplet is tiny compared to the total sunlight gathered by its surface area during the diurnal cycle.)

In effect, the droplet becomes an 'extended phenotype' outer cell for its passenger. There is a resemblance to an eukaryotic cell containing symbiotes such as chloroplasts or mitochondria.

A fascinating possibility is that if two or more spores share a droplet, the one which can occupy the safe zone while pushing the other into the stomach zone can kill its rival and generate food for its own use.

4. DROPLET CONTROL

The fate of the droplet itself can to an extent be controlled by its passenger. Production of even a molecule-thick layer of oil to coat the droplet surface will slow evaporation. If the spore partially protrudes through the droplet wall, it may be able to facilitate the escape of water and other molecules, possibly also leaving the droplet with a net electrical charge.

Evidently, the spore can benefit from its time aloft. It can harvest matter and energy and grow, bulking up and even dividing. It also benefits from the 'information processing time' which is the main resource provided by an externally laid egg such as a bird egg.

Ultimately, by forcing aggregation with other droplets, e.g. using the fact that opposite charges attract, it can control the formation of a raindrop to fall at a favourable location. Environmental parameters it might respond to could include air temperature, sunlight intensity, and (as the droplet can act as a crude focusing eye) possibly even the colour of light and quantity of IR from the surface below.

In many circumstances (high altitude / high latitude / local winter) the droplet may freeze. Trace constituents can control the temperature at which it freezes, the shape of the ice crystal formed, and perhaps any subsequent tendency to aggregate with other crystals. In sunlight the solid state greenhouse effect (clear ice transmits visible light readily, but not infrared) could cause an opaque bacterium to occupy a blob of salty water within an otherwise frozen agglomeration of water crystals.

This is all hypothetical, but it could explain at least two puzzles:

1. Absorption Anomaly: if something tends to force it toward the optimum location within the droplet, the 'stomach', a small quantity of light absorbing material, organic or otherwise, can intercept tens

of times the sunlight that it could either floating at a random position within the droplet, or floating in the atmosphere on its own. This could explain the large and rapidly changing amount of sunlight absorbed by clouds.

2. The high concentration of bacteria frequently found in raindrops and snowflakes.

The hypothesis might also help to resolve longstanding puzzles concerning the processes which take place in thunderstorms.

It would be fascinating to discover an additional association with such curiosities as frost flowers growing on sea ice.

Testing the hypothesis is fairly easy: an initial test simply involves bringing natural cloud aerosol droplets detected to contain organic chemicals within the field of view of a confocal microscope and observing how light-absorbing items within the droplet reposition themselves with respect to incoming sunlight, as the droplet rotates or the direction of the sunlight is artificially changed.

If life does make use of this resource, the theoretical and practical consequences will be enormous. The sunlight energy harvested is potentially of the same order as the total used for photoautrophy by currently known organisms. If some aerosol droplets are functioning as a kind of meta-eukaryotic cell, the total mass of these cells could be greater than the total mass of known single-celled organisms.

5. ORIGIN OF LIFE

It has been suggested that airborne water droplets may have constituted the protocells within which life began.^[6,7] While there are many uncertainties concerning the primordial atmosphere before the advent of life, the mass of gas was probably comparable to today's atmosphere, the solar energy entering it ~70% of today's value, yet it contained liquid water (the faint young sun paradox^[8]). Today there are roughly as many atmospheric aerosol droplets of a given size as there are bacteria of comparable size, and the number then was likely of the same order of magnitude. Looking at Titan's present atmosphere, Earth's primordial atmosphere probably contained no free oxygen, but water and large quantities of hydrocarbons, including tholins: long-chain heteropolymers formed by nonbiological processes.

There is an overwhelming statistical case for life arising in primordial atmospheric aerosol droplets. As today, the droplets would have been continually churning about in atmospheric turbulence, potentially interacting with one another, the population turning over as new droplets come into existence while others evaporate. The richness in terms of number of chemical reaction vessels and number of approaches between neighbours with the potential for interaction dwarfs anything which could be provided by other environments, for example rock pores in the vicinity of black smokers as sometimes suggested.

A unique asset is the 'stomach' I have identified. Initially, it acts as a Darwinian processor: irrelevant large molecules get broken down and recycled within milliseconds. Put another way, it destroys both unwanted information and physical debris, which – as Szilard's disproof of Maxwell's Demon, and the necessity for efficient rubbish collection services within cities, indicate – are key if often neglected processes in physics and biology. In practical terms, it avoids the obstacle so often described by those who try to recreate the origin of life in test tubes: the black goo problem, everything just gets clogged up with gunk. Here, uninteresting gunk is almost instantly recycled.

How could autocatalytic cycles^[9] *not* be rapidly discovered by decillions of such machines being created and destroyed each day? How could the successful mixtures *not* have some influence on the fate of their containing droplets, thus starting the next level of evolution?

Later, the stomach becomes an agent of creation. In biology, unlike most human engineering, the strategy of trying out vast numbers of combinations in order to immediately throw away all but a tiny fraction is a ubiquitous strategy: from the level of creating long strand molecules in the ribosomal complex, to emplacing neurones and their connections in the developing brain, to creating vast numbers of tiny swimmers only a

tiny fraction of which will reach adulthood. It is very challenging for a simple proto-lifeform to manufacture everything it needs. But if it merely clings on to what it needs, and everything else drifts into a nearby stomach, to be instantly reordered into random chemicals, what it needs is likely to come by soon enough, even a chemical which forms only once in a billion random recombinations.

Physically, even very small quantities of material can control the size, surface tension, electric charge, and other properties of the droplet. When the surrounding temperature drops below zero, the material can select the freezing temperature (by acting as an antifreeze) and shape (by acting as a crystal seed) of the ice crystal formed.

So after a first level of intra-droplet competition – which remains ongoing, of course, just as our selfish genes compete with one another within our bodies – the droplets will compete with one another, in due course 'learning' the value of protective membranes with machinery for passing only wanted materials through them, and becoming the kind of cells we recognise today.

It is usually assumed that origin-of-life theories will remain untestable, the original proto-organisms long ago eaten. However there are environments which are intrinsically unsuitable for DNA or RNA-based life, or indeed any based on molecules of very high molecular weight. These include the smallest aerosol droplets, due to the limited safe space available within, and high altitudes, due to the high level of UV radiation present.

It is possible that if we look, we will find that in such niches what is sometimes called 'metabolic life', autocatalytic collections of smaller molecules, can still be discovered. An especially promising location would be the human-created ozone holes at the north and south poles. It would be a wonderful irony if our tendency to wreak ecological devastation has recreated environments where the earliest life of all can again become numerous.

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