Redistribution of CO_2 between atmospheric layers as a means to mitigate global warming – two thought experiments

John R Porter
Professor D.Sc.
Faculty of Sciences
University of Copenhagen
Denmark
jrp@plen.ku.dk

Summary: Climate change mitigation and negative emission technologies need to consider options for redistributing CO_2 between different layers of the atmosphere, which may raise the global albedo as well as mitigate global warming. This short paper is an attempt to stimulate thinking about how this might be done. I am convinced that radical technical solutions will be required to limit global warming.

Negative greenhouse emission technologies (NETs) attempt to redistribute CO_2 between atmospheric, land, ocean and geological reservoirs to reduce the concentration of greenhouse gases in the atmospheric reservoir. The biophysical and economic limits to a range of such processes were recently presented[1], with the conclusion that 3.3 Gt C is the level of NETs needed for a possible 2^{o}C future, in the absence of drastic fossil fuel emissions. Given the current lack of international urgency to ratify agreements made at the COP21 meeting in December 2015, which could be considered modest at best and extremely unlikely to hold warming below 2^{o}C about pre-industrial – it is likely that 'technical fixes' involving large scale geo-engineering will perhaps be the only way to avoid lethal threats to the planet from high levels of warming. Finding reservoirs for greenhouse gases may mean that perhaps we need to look upwards, as well as downwards from the Earth surface, for other possible reservoirs[2] for CO_2 and consider how it might be possible to remove CO_2 from the global atmosphere and out into near-space. The atmosphere is structured with the three lowest layers being the troposphere (up to about 10km above the land surface), followed by the stratosphere (up to about 50km above the land surface). Above these two is the mesosphere and there is a boundary layer between this and the lower layers such that the CO_2 concentration (ca. 150 ppmv CO_2) in the mesosphere is less than half that in the lower layers.

The key property to enable the biological storage of C is the fact that the terrestrial ecosystem is composed of strata - for example above- and below-ground reservoirs of C. These strata have different residence times for C, caused by the degree and types of chemical linkages of C with other elements, particularly H. At one extreme there is oxygenated C, as CO_2, and at the other extreme are hydrocarbons of one form or another giving chemically reduced C. The terrestrial C strata are a reflection of the different forms of C. On the other hand the global atmosphere has an almost complete predominance of C as the CO_2 form and thus the question arises as to what is the basis of the atmospheric stratification into the troposphere, stratosphere and mesosphere (ignoring the thermosphere – or near-space layer at about 110 km height above the Earth surface). In the atmosphere, stratification is caused (according to the people who know about these things) by vertical differences in O_3 amounts and concentrations but much more importantly by the spectral properties of CO_2 – which absorbs long-wave radiation but is transparent to short wave-lengths. Planetary stratification of C depends, in the biosphere, on the chemical form of C, but in the atmosphere on the physical radiative preferences of gaseous CO_2; the generic element being that both planetary chemistry and physics generate 'boxes or reservoirs' in which C can be stored. Technologies and policies, such as climate-smart agriculture and forest preservation, are being used in order to redistribute C from atmosphere to biological sinks to offset the warming effects of burning C from historical reservoirs, with chemistry being the driving '*metier*'. But can the physics of the atmosphere be used to alleviate tropospheric warming by altering the strata-based distribution of C in the planetary atmosphere? We will examine two ideas.

The mesosphere is characterized by two important features – it harbours so-called noctilucent clouds formed by ice crystals and, whereas in the lower atmosphere CO_2 warms the planetary surface by absorbing infrared radiation radiated by the earth's surface; in the mesosphere CO_2 cools the atmosphere by radiating heat into space[3]. Noctilucent clouds are formed by ice crystals from water vapour and seeded by small dust particles with sizes of about 40-100nm and are thought to have a role as indicators of climate change[4]. In addition, the mesosphere temperature is about 10-40^{o}C colder than the STP sublimation temperature of CO2 (-75^{o}C at 100kPa or 1bar(B), decreasing to -140^{o}C at 100Pa or 1mB), at which point CO_2 changes directly from a gas to a solid and, in the mesosphere, changes to white crystals. Thus at first glance, if more CO_2 could be injected into the mesosphere via geostationary machine and what might be called 'atmospheric stomata' (to borrow an analogy from leaves) then this would reduce levels in the lower layers and at the same time raise the global albedo via the presence of reflective crystals in the mesosphere, thus having a solar radiation management effect on the stratrosphere[4]. The main physics questions would be what is the effect of moving CO_2 upwards, how much needs to be moved, how long is its residence time in the mesosphere and what effect would an increase of CO^2 in the mesosphere have on the global albedo.

However and unfortunately, this scenario is not realistic for two decisive reasons; the first is shown from the CO_2 phase diagram (Figure 1) that the sublimation temperature decreases to about -140^{o}C for the kind of extremely low pressures that exist in the mesosphere, which is to all intents a vacuum. Such extremely low temperatures are only found occasionally in the mesosphere and only at high latitudes. The second issue would be the engineering aspect that would be very difficult, as it is for all NETs. The main challenge is how to concentrate the CO_2 so that one does not have to move large volumes of air. There are technologies on the horizon that might help here; catalytically enhanced sorption-desorption[5], nano-sieve membrane separation[6] or cyclonic separation technologies[7], that are capable of separating CO_2 from air and are currently being developed. However, overcoming technical and engineering barriers is a feature common to many forms of NETs. The energy aspect of such technologies is not a concern as solar energy at almost the power density of the solar constant would be available to drive the required processes.

Initial calculations indicate that about 3.3 Gt C per year would be a reasonable aspirational target for atmospheric stomata, but in concert with other NETs, a level of 1 Gt C of NETs removed via atmospheric stomata is another option. What is needed is an effort to make some models of the processes involved and work on possible designs of machines for sorption-desorption , membrane separation or cyclonic separation that could be used to efficiently concentrate and transport CO2 from the stratosphere to the mesosphere. It is impossible to disagree with Smith *et al.'*s conclusion that the preferred route to a livable planet in the future is via a rapid transfer from fossil to non-fossil based fuels, but there is likely to be a need for new thinking in terms of NETs in the future. Combination of the removal of CO2 together with an increase in the atmospheric albedo may offer one such novel route.

If the mesosphere as an enhanced reservoir for CO_2 has to be excluded for the reasons given above, then is there any mileage in considering the next level down - the stratosphere, which extends from about 10km to 50km above the Earth's surface. The pressure-temperature relationship of the stratosphere is almost as unconducive to sublimation of CO_2 as in the mesosphere but there is evidence that increasing levels of CO_2 in the stratosphere does lead to cooling of this layer[10]. The reasons for this are unclear, but has to do with the differential emissivity and absorption responses of the troposphere and stratosphere in the presence of CO_2. We know that emissivity of long-wave radiation from the troposphere to the stratosphere declines as CO_2 increases (as it is doing via fossil fuel burning); however emissivity of the stratosphere either remains constant or slightly increases with higher levels of CO_2 (ozone in this layer also plays a minor role in the stratospheric short-wave radiation emission and energy balance). The net effect is that emissivity increases relative to absorption in the stratosphere with increased CO_2 in the troposphere and thus the stratosphere paradoxically cools with increased levels of tropospheric CO_2. The big question is – can this understanding be used to cool the global atmosphere by physically pumping CO_2 from the troposphere to the stratosphere? At first sight this would have the effect of reducing warming in the troposphere whilst increasing cooling in the stratosphere – and thereby contribute to a reduction in global warming. There are no doubt many who will say that the engineering challenges of doing such changes of carbon pools in the atmosphere are prohibitive – but are they really more challenging as physics than the chemical challenges of converting CO_2 into more reduced terrestrial forms and trying to store them in global ecosystems, including agriculture. We need more research on the physical and chemical properties of the higher atmosphere and to think 'big' about how we are to avoid the pitfalls and Earth system shocks of an enhanced Anthropocene, which is the direction in which humans are moving at ever faster speeds.

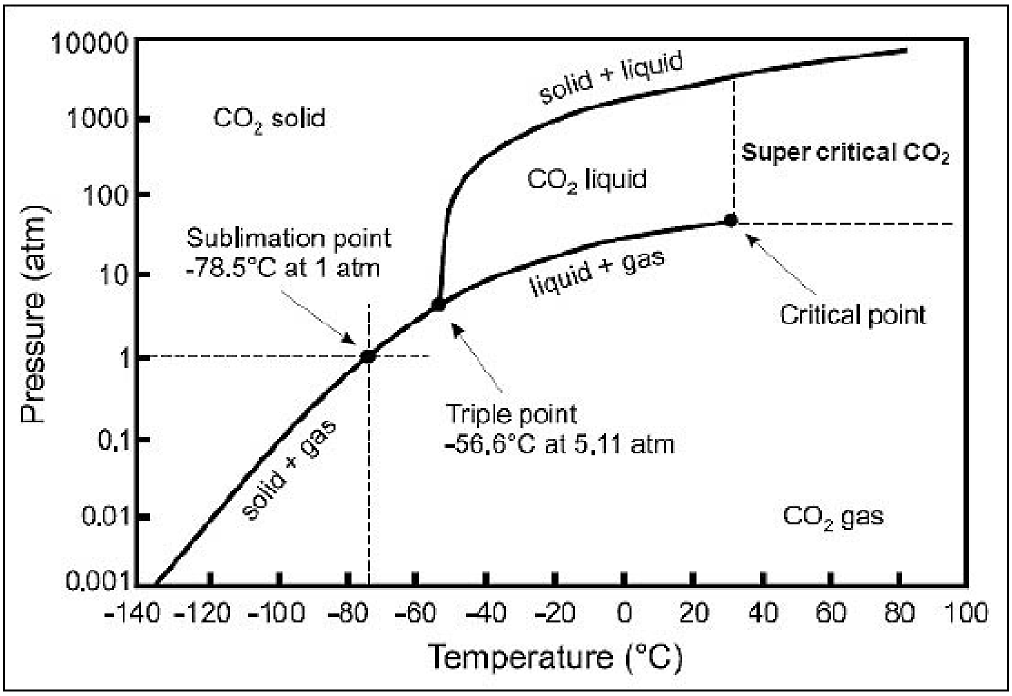

Figure 1. The Temperature-Pressure Phase Diagram for CO_2 showing how the sublimation temperature of CO_2 decreases with decreasing pressure. http://hub.globalccsinstitute.com.

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Short information on the author:

Consultant Professor, Supagro Montpellier, France
Emeritus Professor of Climate and Food Security, University of Copenhagen (UC), Denmark
Emeritus Professor of Agriculture and Climate Change, Natural Resources Institute, University of Greenwich, UK
Honorary Professor, Lincoln University, New Zealand
Honorary Fellow, University of Bangor, UK

Career Summary

Porter is an internationally known agro-ecological scientist with an expertise in ecosystem services in agro-ecosystems, including agro-ecology, simulation modelling and food system ecology. His main contribution has been multi-disciplinary and collaborative experimental and modelling work in the response of arable crops, energy crops and complex agro-ecosystems to their environment with an emphasis on climate change, ecosystem services and food systems. Porter has published 145 papers in peer-reviewed journals out of a total of about 350 publications. On average, his peer-reviewed papers have been cited more than 100 times each. He has personally received three international prizes for his research and teaching and two others jointly with his research group. His career H index is 57 and with 129 papers receiving over 10 citations.

From 2011 to 2014 he led the writing of the critically important chapter for the IPCC 5th Assessment in Working Group 2 on food production systems and food security, including fisheries and livestock. This chapter was one of the most cited from the IPCC 5th Assessment and formed an important scientific bedrock of the COP21 agreement in Paris in 2015.