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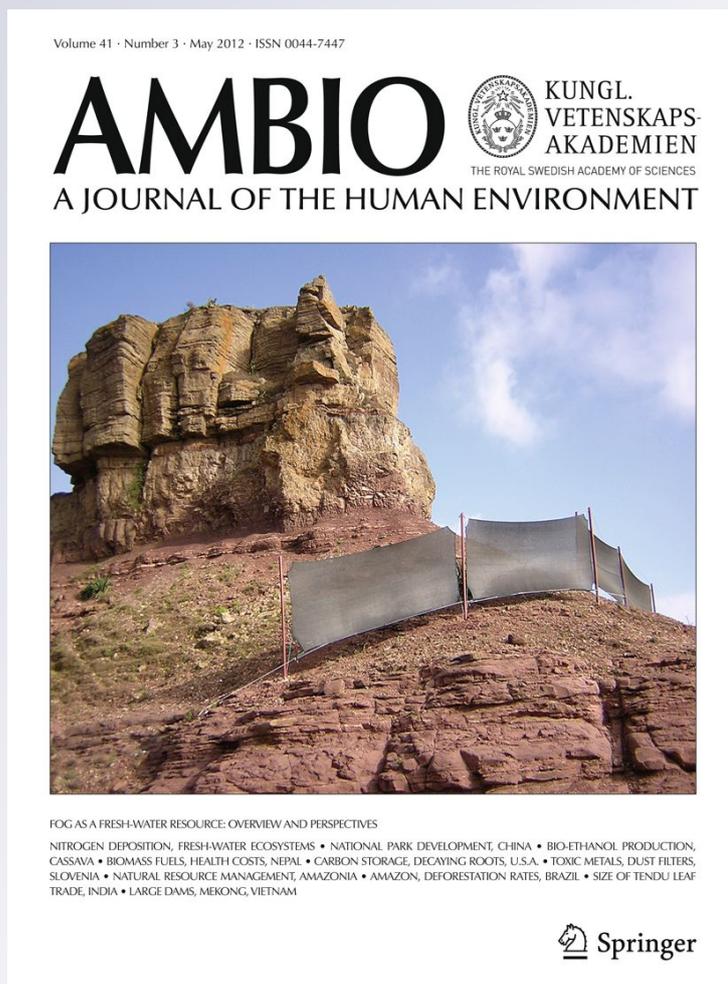
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INTRODUCTION

Humans became a global force in the chemical evolution with respect to climate change by interrupting naturally evolved biogeochemical cycles. However, humans also have all the facilities to turn the “chemical revolution” into a sustainable chemical evolution. I define a sustainable society as one able to balance the environment, other life forms, and human interactions over an indefinite time period. According to Steffen et al. (2007), “The Great Acceleration is reaching criticality. Whatever unfolds, the next few decades will surely be a tipping point in the evolution of the Anthropocene”. There is much discussion on “sustainable chemistry” (often called green chemistry), but, in my understanding, the basic principle, is to transfer matter for energetic and material use only within global cycles, without changing reservoir concentrations above a critical level, which is “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt 1988).

With respect to atmospheric pollution, the last unsolved issues (remaining pollutants) are “greenhouse” gases, namely CO₂, which contributes to about 70% of anthropogenically caused global warming (other important gases such as CH₄ and N₂O contribute to roughly 25% of warming; these gases are associated mainly with agricultural activities). The dilemma is given simply by time scales:

limits of the 2°C threshold by 2050 and drastic reduction in global CO₂ emission; that is, the *cumulative* CO₂ emissions determine atmospheric (and oceanic) CO₂ levels. Because of the large CO₂ residence time in natural reservoirs, in the order of 1000 years in the atmosphere and more (about 200 000 years for dissolved inorganic carbon—DIC) in surface seawater, humans now determine the still unknown relationships of possible climate recovery, irreversible climate change, and future abatement strategies (Solomon et al. 2009). Since the beginning of the (first) Industrial Revolution, the burning of fossil fuels caused about 330×10^9 t CO₂-C (Boden et al. 2009). In addition, about 150×10^9 t CO₂-C was released into the atmosphere through land-use change (Houghton 2005). Since 1950, the total CO₂ anthropogenic release has amounted to 350×10^9 t CO₂-C, of which about 50% has accumulated in the atmosphere (Prentice et al. 2001). The percentage not accumulated in the atmosphere must have been taken up by the ocean and terrestrial biosphere as well. Mining and the combustion of fossil fuels now results in the geological reservoir redistribution of carbon close to (or even surpassing) the “tipping point”. It is assumed that in the near future the acceleration of CO₂ release will increase as a result of economic growth. Due to the large CO₂ residence times in air and sea water, we are far away from reaching a steady state (global cycle in-time) and recovery (climate restoration), even after the complete cessation of fossil fuel use.

Therefore, forced by climate change and its uncertain, but very likely catastrophic impact after reaching the “tipping points” than fossil resource limits, we need to transfer into the “solar era” as soon as possible. Nuclear power may be considered as a “bridging technology” but the risks may not be longer accepted by society. Secondary “renewable” energy, that has already been in use for long

time, such as water and wind (and we should not forget that it was the only significant source of energy before the first Industrial Revolution), will probably never contribute on a global scale to fit the energy demand (this does not exclude national and regional solutions proposed nowadays for Germany). Hence only the direct use of solar energy as proposed, for example, by the *desertec* conception (<http://www.desertec.org/>), can realistically solve the global energy problem and fully replace fossil fuels. Without a doubt, electricity is the unique form of energy in the future and its direct application (also for mobility and heating) will increase, and will replace traditional fuels based on fossil resources to a large extent. The *desertec* technology is not unlikely to realize within the next few decades and can replace fossil fuels remarkable, if political (and thus financial) willingness is given. However, there are some open questions which have to be answered and transferred into technical solutions to establish the solar era:

- Electricity will not be produced constantly over time nor correlated with the demand for energy, hence it must be stored, likely best by transfer into “chemical energy”, to manage energy supply.
- For safety reasons, excess energy must be stored (for example, in water reservoirs, but this is limited). Again, the best way seems to transfer electricity into “chemical energy”.
- There are technical applications (for example, air traffic, long-distance street traffic, shipping, and metallurgy) where electricity cannot be taken directly from nets or storage units, and will be neither ecological nor economic.
- Humans always need synthetic organic materials (polymers, drugs, chemicals, etc.). These can be produced from the remaining fossil resources, but also from biomass, and from CO₂.

In this article, I put forward an option to create a global-closed anthropogenic carbon cycle using only solar energy to: (a) stop the further increase of CO₂ emissions, and to obtain a global zero-carbon budget; (b) solve the problem of electricity storage based on CO₂ utilization; (c) to provide carbon-based materials only from CO₂ utilization; and (d) use the infrastructure developed for the fossil fuel era. The specific approaches put together in this “CO₂ economy” are already known and/or have been proposed. However, to my knowledge, the creation of a man-made carbon cycle in such an integrative approach, and with such rigorousness in linking energy with material economy, adopting the principle of natural cycling but not copying natural processes,¹ as suggested here, is new and unique

¹ For illustration, some scientists dream of artificial leaves to transform CO₂ into (solar) fuels. Our approach consists of “secondary” use of solar energy in terms of electricity and heat in large industrial operational units, which are already known in principle.

worldwide, and even more complex than the “methanol economy”.

THE CARBON DIOXIDE ECONOMY

The SONNE (*SOL*ar-based maN-made carboN cyclE) concept (“*Sonne*” is the German word for sun) will link solar electricity ideas such as *desertec* with CO₂ utilization, to overcome the above mentioned open problems after the fossil fuel era. In other words, SONNE will build a man-made carbon (CO₂) cycle like the natural assimilation-respiration carbon cycle (Fig. 1). CO₂ is recycled within hybrid power plants (see Fig. 2) and captured from ambient air. It is changed from waste (emissions) to resource; process energy is taken from solar energy. CO₂ is unique²:

- as a final oxidation product of all organic matter and materials;
- because of its global cycling and homogeneous distribution in the atmosphere (but keeping a level before “tipping points”);
- as a resource for organic materials, concerning carriers of energy and functional materials;
- as the only element forming complex molecules and substances, and being within a global dynamic³ cycle and gaseous compounds in its lowest (CH₄) and highest oxidation states (CO₂);
- since the only environmental problem of CO₂ is its increase in the atmosphere (and seawater) with climatic implications; hence controlling its level to acceptable values will overcome the environmental problem.

It is evident that through the realization of these principles a CO₂ “zero-budget world” rather than a “CO₂ free world” can be achieved, because there is a closed anthropogenic carbon cycle (we call it CO₂ economy). Some CO₂ still emitted (e.g., from mobile and small equipment, where internal capture is unlikely) will be captured from air and cycled for reuse. I call this “Carbon Capture and Cycling” (CCC) technology. With this in mind, CCS technology (carbon capture and storage/sequestration) makes (more) sense, despite the controversial CO₂ storage problems, and provides considerable incentives because CO₂ storage is

² In a certain sense hydrogen (H₂) can also play the same role as energy carrier when we adopt the natural water splitting process, which was proposed as “hydrogen technology” in the early 1980s. However, there are several problems: (a) safety of storage and transport; (b) leakage and atmospheric implications; and (c) missing material supply. Water electrolysis will play an important role in SONNE for oxy-fuel combustion (O₂ supply) and CO₂ reduction (H₂ supply).

³ In (biogeochemical) cycles move all elements and their compounds, but often on a geological time scale (besides carbon only sulfur and nitrogen are in similar dynamic cycles).

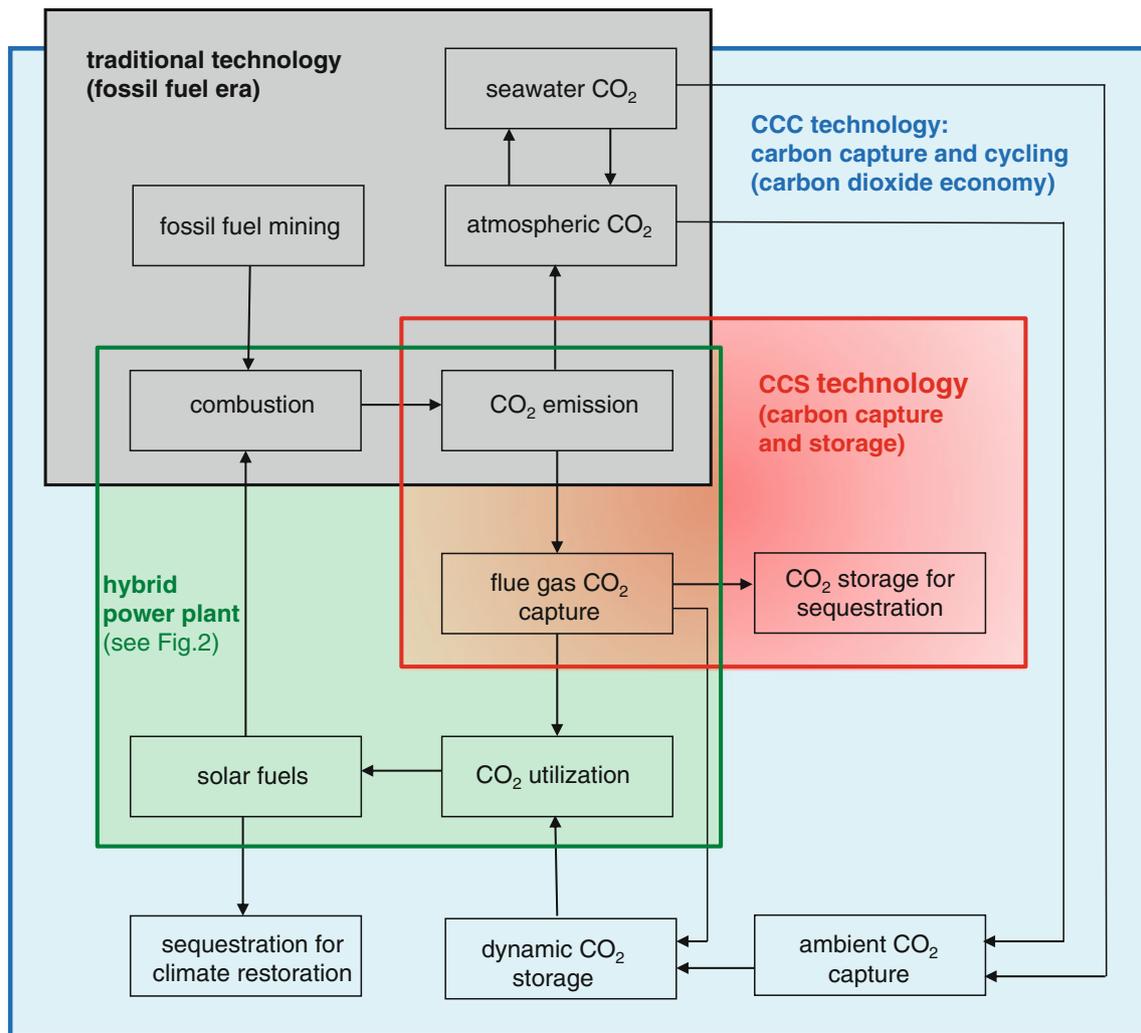


Fig. 1 Scheme of energy transition from fossil to solar era including CO₂ economy (SONNE concept). Three overlapping systems: fossil fuel burning without (gray box) and with carbon capture (red box), as well solar fuel production/use (green box), and global carbon cycling (blue box); the inner green box comprises a “hybrid power plant” (see Fig. 2). The blue box comprises all technologies into the carbon dioxide economy; providing electricity storage within usable carbon compounds (“solar fuels”), as well carbon materials produced from

CO₂ instead fossil fuels. Moreover, ambient CO₂ capture allows the global carbon budget to be “negative” through the sequestration of geological stable carbon (e.g., elemental carbon). The driving force is exclusively solar radiation; hence the CO₂ economy is interdependent with solar electricity conceptions such as *Desertec*. Elements of this concept can be introduced in parallel with the further use of fossil fuels aimed at their stepwise replacement

now only temporary (“dynamic”) until it is recycled from waste to feedstock. The proposed CCC technology allows a stepwise replacement of coal and other fossil fuels by solar fuels while keeping the carbon-based infrastructure, such as pipelines, tankers, storage facilities, and engines. It also allows the continuous use of other available technical applications developed within the last hundred and more years, but within a CO₂ neutral closed loop.

Closure of the carbon cycle, however, is only possible when CO₂ can be extracted from natural reservoirs, such as the atmosphere and seawater, because complete “industrial” CO₂ capture will be impossible with regard to many small

and mobile sources. The idea of air capture (CO₂ extraction from the air) as a climate control strategy is now accepted and considered in global ecological (Cao and Caldeira 2010) and economic models (Edenhofer et al. 2006). The large-scale scrubbing of CO₂ from ambient air was first suggested by Lackner et al. (1999) and Zeman and Lackner (2004). However, with the exception of CCS, which is presently transferred to larger technical equipment being tested in pilot plants, DAC (direct air capture) and CCU (carbon capture and utilization) still only exist within the laboratory or only on conceptual levels, characterized by different approaches. For example, our DAC approach is based on

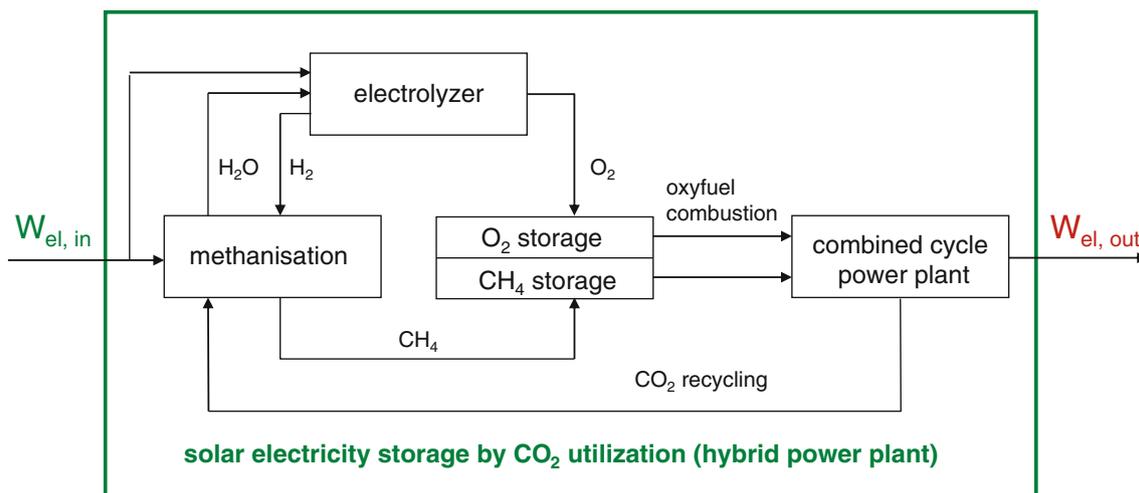


Fig. 2 Schema of a “hybrid power plant” (green box in Fig. 1): chemical storage of “renewable” energy (preferably solar electricity) by CO₂ utilization (likely best by methanization) and internal CO₂ recycling (likely best by oxy-fuel combustion); $W_{el, in}$ —direct solar

electricity, $W_{el, out}$ —indirect solar energy (electricity/heat) “on demand”. The energy efficiency is negative (for example, in the case of CH₄ production from CO₂, only 30% of electricity input can be reused)

CO₂ dry deposition above ponds with circulating solvents, where CO₂ desorption proceeds by ultrasonic stimulation at moderately increased temperatures up to 60°C.⁴

Principally, the SONNE concept is not aimed at the very near future, but for the solar era with “unlimited” access to useable solar energy, most likely after 2050. However, because CCS will be an essential technology within the “internal cycle” of hybrid-type power plants (Fig. 2), CCC could be introduced to some extent in parallel with the further use of fossil fuels, and stepwise replacement of them until fully establishing the SONNE cycle. The principal scheme of CO₂ use in solar electricity storage (valid also for other “renewable” energy such as wind) could be soon realized. So-called oxy-fuel combustion would provide high purity CO₂ as an exhaust gas which can be recycled without energy-intensive capture (Fig. 2). We can set ten mission statements or principles:

1. Further use of fossil fuel combustion in large stationary units, but only with CO₂ capture (CCS technology) until full transfer into the solar fuel world: capture CO₂ from combustion units as much as possible.
2. Replacement of fossil fuel use in small stationary and mobile units as far as possible (electricity-based and hybrid techniques): reduce carbon carriers as fuels as much as possible.
3. Sequestration of carbon (not CO₂) on medium and long-term scales, for buffering the further increase of

CO₂ emissions within the next decades, and for climate sanitation in the distant future.

4. Develop technologies for CO₂ extraction from natural reservoirs (ambient air, seawater), to achieve a global man-made carbon cycle, while allowing CO₂ emissions into the atmosphere from mobile and small sources: atmospheric CO₂ is considered as the only carbon reservoir for chemical CO₂ utilization (CCU).
5. Develop technologies for CO₂ reduction, but applications are only for renewable energy, namely, solar radiation (solar fuel production).
6. Introduction of large solar-thermal power plant units for electricity generation.
7. Develop technologies for electricity conversion into chemical energy carriers (solar fuels used in hybrid power plants).
8. Build up a solar fuel infrastructure (on the basis of the existing fossil fuel infrastructure).
9. Develop technologies for electricity conversion into large central heat storage units (based on molten minerals).
10. Economic paradigm change: solar energy is “in excess” (compared with global human demand) and is naturally dissipated in the atmosphere; hence, large energy consuming conversion processes and direct air capture can be carried out for resource generation and climate sustainability: a new economic thinking based on sustainability (or closed carbon cycle) is needed. In other terms, not energy but material efficiency becomes the key factor.

The idea of using CO₂ as a chemical raw material is not new (Aresta and Forti 1987; Edwards 1995; Aresta and

⁴ A critic of the American DAC Report comes also from the Climeworks Company which is doing solar-thermal CO₂ capture and conversion in cooperation with the Professorship of Renewable Energy Carriers, Institute of Energy Technology at ETH Zurich (Switzerland).

Aresta 2003; Park et al. 2004; Olah 2005; Aresta 2010). However, when using CO₂ from fossil-fuel gases, it is only climate-sustainable if the products are “sequestered”; for example, by long-term use in carbon materials such as polyurethanes. CO₂ captured from fossil-fired power plants and “utilized” for the storage of excess electricity (for example, from wind power) may help to improve energy efficiency (because the excess electricity cannot be used on demand), but will not solve the climate problem. Nevertheless, the results from many researchers are the basis for the utilization of CO₂, captured in the future from the environment. From the presentations given at the 11th International Conference on Carbon Dioxide Utilization, held in Dijon in June 2011, it seems to me that catalytic CO₂ hydrogenation and solid-state high-temperature electrolysis of CO₂ are favorites for the global industrial CO₂ economy, whereas photo-catalysis and artificial photosynthesis remain of academic interest only.

Within the last few years, considerable progress has been achieved in the catalytic hydrogenation of CO₂ (methanization). The possible synthesis of C₁-chemicals (CO, C, CH₄, CH₃OH, and HCHO) from CO₂, and further to C₃, analogous to the assimilation process (see also Möller 2010), leads to a variety of important basic chemicals being available for either direct combustion or material use (industrial synthesis in organic chemistry); we now call these *solar fuels*. However, a global CO₂ economy must not only provide chemicals in the order of a hundred million tons but also more gaseous and liquid fuels of 1–2 orders of magnitude. By using high-temperature chemical processes (which have been known for many years, but owing to the high energy consumption have hardly been mentioned before) based on solar-thermal energy it is also possible to remake “coal chemistry” (gasification and liquefaction) via CO₂ reduction. Namely, carbon monoxide (CO) and elemental carbon may be produced and transformed inversely. For example, elemental carbon could be stored better than carbon dioxide (sequestration), but could also be reused directly in an early stage of the CCC technology. It is known that in high-temperature processes of conversion, of carbon compounds to elemental carbon, the yield of polymeric carbon structure (fullerenes) results in large and unforeseen changes in creating new carbon materials. Human evolutionary responsibility should consider the retransfer of emitted CO₂ into geological stocks; for instance, as elemental carbon for safe sequestration and stepwise but long-lasting climate recovery.

Olah (2005) proposed a “methanol economy” but in the SONNE concept, CH₃OH is only one possible product among C₁ chemicals; the Fischer–Tropsch synthesis (from CO + H₂) basically offers a wide range of organics including liquid fuels. Our “CO₂ economy” includes the “CH₃OH economy”. Recently, it has been shown that the

energetic efficiency of the overall energy conversion–storage system (see Fig. 2), including CH₃OH as a storage medium, is only 17.6% in contrast to 29.7% for CH₄ (Rikho-Struckmann et al. 2010). However, taking into account ambient CO₂ capture, the overall energetic efficiency will lower drastically. As in nature, where the photosynthesis efficiency concerning solar light is only 2–3%, we realize a closed carbon loop only with large solar energy input; in other words, low energetic efficiency. However, the incoming solar radiation is roughly 1000 times higher than present global human energy demand. Still unanswered is the question, what are the limits of solar use without resulting in other climate implications?

It is remarkable to me that by establishing the SONNE conception (CO₂ economy), we first see that sun-belt countries, many of which are privileged with natural oil and gas reservoirs, will provide “solar sites” for electricity generation and probably CO₂ processing (Fig. 3). On the other hand, future use of fossil fuels is mainly in non-sun countries in the Northern hemisphere, which should become responsible for ambient CO₂ capture (there are good reasons, to establish DAC units more in the north because CO₂ absorption processes need low temperatures, and DIC in seawater is significant higher in cold areas). For example, Northern Europe will capture ambient CO₂ and transport it to Northern Africa as a “fuel feedstock” for solar processing (Fig. 3). Thus, a social win–win situation with many positive political and educational effects may be created.

At this point, I want to state that SONNE is based on ideas already known and investigated (for example, CCS, CCR,⁵ CCU, DAC) at many scientific institutions worldwide. As mentioned, a key idea of CCC technology is the capture of CO₂ from the atmosphere (and its dynamic storage) to close the man-made global carbon cycle analogous to the biosphere. The CO₂ economy is the adaption of the biospheres’ assimilation–respiration cycle by humans; the only long-term sustainable way of surviving. We also must learn (and accept) that permanent economic growth (stated by politicians as the solution to social problems) results in collapse when not reaching a steady-state condition. Our present socioeconomic approach must be replaced by a socio-ecological reference in the organization of society (note that ecology is the economy of Nature).

From today’s perspective it seems that as a result of the extremely low concentration of CO₂ in the air, the technical and economic solution of direct atmospheric CO₂ reuse is not very likely (DAC 2011). However, any technical solution in our concept is based on the paradigm change to establish a zero-carbon budget (not zero

⁵ R stands for recycling.

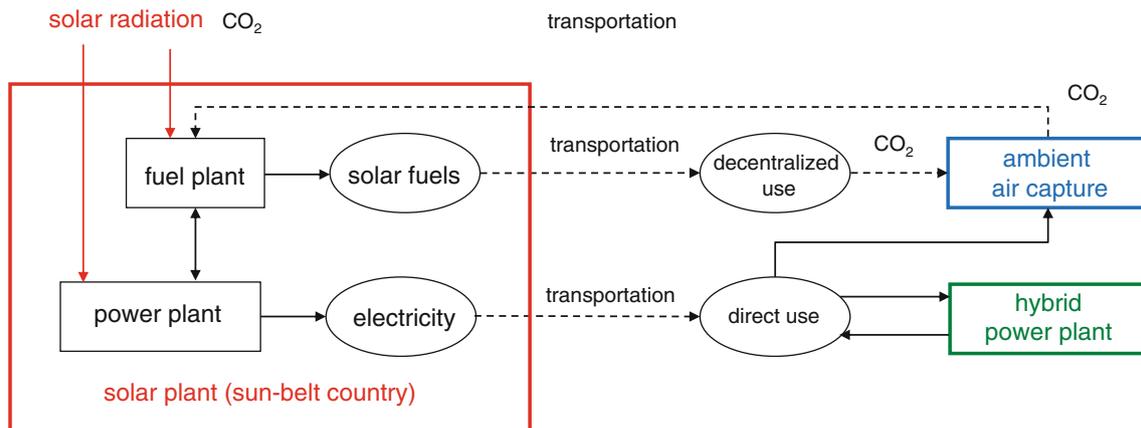


Fig. 3 Solar plant complex (solar-to-electricity and CO₂-to-fuel conversion in sun-belt countries), interlinked with transportation of fuels, chemicals and materials, and electricity to Northern hemisphere countries (to be used there), and back transport of CO₂ captured from ambient air to solar plants (achieving a geo-economic equilibrium and political interdependence as win-win-situation). Note that the whole

emissions!), and to no longer measure the effect on energy efficiency (solar energy is in “excess”) but on budget, with respect to climate sustainability. The “price” of CO₂ emitted from fossil fuels (and hence fossil fuel costs) must include climate change affects; this would encourage energy transition and also DAC technologies.

In the background of further—and likely globally increasing—fossil fuel use, carbon capture and storage (CCS) technology is considered to be the only practical solution in early CO₂ control. However, it will never reduce anthropogenic global CO₂ emissions, only smooth their further increase (Möller 2010). Acceptance of my SONNE concept, however, forces the introduction of CCS in the sense of a “bridging technology” and its later replacement by CCC, towards a solar-driven global complex and hierarchic CO₂ cycle.

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system dissipates solar energy as it happens into the climate system by natural processes. Therefore, energy efficiency plays a minor role (but should be maximized in single technical process); the key for a sustainable economy (socio-ecological system) is the closed cycling of matter (here carbon, but this is true for all elements to be used)

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