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Atmospheric chemistry – Bridging the chemical air composition with the climate

Detlev Möller

Chair for Atmospheric Chemistry and Air Pollution Control,
Brandenburg Technical University, P.O. Box 10 13 44, D-03013 Cottbus, Germany
Volmerstr. 13, D-12489 Berlin, Germany
E-mail: moe@btu-lc.fta-berlin.de

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Abstract—Studying the environment is an extremely complex issue, comprising all natural sciences and regarding the “key” compartments water, air, and soil, often defined as the climate system. Without any doubt, the atmosphere is the most important one, it is highly dynamic and globally interlinking the biosphere. Nowadays, the humans are shortly before reaching the “tipping point” between moving into the (climate) catastrophe or the sustainable development. This paper introduces a new discipline, the “chemistry of the climate system”. Atmospheric chemistry, without using this term before the 1950s, was a fundamental approach in the beginning of modern chemistry (air analysis and understanding combustion) some 200 years ago, and it is now enlarging to the interfaces (“interfacial chemistry”) with the biosphere and hence our climate system. Humans created a new dimension (the “anthroposphere”) by globally modified biogeochemical cycles. Climate control means learning from nature and creating closed man-made material cycles, first of all that of carbon. Our environmental problem is not the limited energy but creating unbalanced reservoir distributions of substances with different characteristic timescales out of steady-state conditions.

Key words: atmospheric chemistry, air pollution, climate, climate change, chemical climate, sustainable chemistry, human evolution, anthroposphere, biogeochemistry

1. Introduction

Out of the recognized complexity of nature, the three basic sciences arose: physics, chemistry, and biology. Further progress in the understanding of natural processes created numerous sub-disciplines and cross-disciplines, termed with a variety of prefixes and combinations, often creating misunderstandings unless careful definitions are used. In order to overcome disciplinary borders, a new super-science was established,the earth system science, to study the earth as a system, with an emphasis on observing, understanding, and predicting global environmental changes involving interactions between land, atmosphere, water, ice, biosphere, societies, technologies, and economies. Humans – by decoupling their life cycle from natural conditions – have altered “natural” biogeochemical cycles. The Russian geochemist, *Vladimir Ivanovich Vernadsky*, understood a new dimension of the biosphere called noosphere (or anthroposphere by *Paul Crutzen*) a new dimension of the biosphere, developing under the evolutionary influence of humans on natural processes (*Vernadsky*, 1926).

We will define air (or atmospheric) chemistry as the discipline dealing with the origin, distribution, transformation, and deposition of gaseous, dissolved, and solid substances in air. This chain of matter provides the atmospheric part of the biogeochemical cycles. A more general definition, but one that is appealing as a wonderful phrase, was given by the German air chemist, *Christian Junge* “Air chemistry is defined ... as the branch of atmospheric science concerned with the constituents and chemical processes of the atmosphere ...” (*Junge*, 1963). In other words, air chemistry is the science concerned with the origin and fate of the components in air. The origin of air constituents concerns all source and formation processes, the chemicals of air itself, but also emissions by natural and man-made processes into the atmosphere. The fate of air constituents includes distribution (which is the main task of meteorology), chemical conversion, phase transfers and partitioning (reservoir distribution), and deposition of species. Deposition is going on via different mechanisms from gas, particulate, and droplet phases to the earth’s ground surface, including uptake by plants, animals, and humans. Removal from the atmosphere is the input of matter to another sphere (see *Fig. 1*).

The term “atmospheric chemistry” appears to have been used for the first time in German by *Hans Cauer* in 1949 (*Cauer*, 1949). It was soon used as the label for a new discipline. The first monograph in the field of this new discipline was written by *Junge*, entitled “Air Chemistry and Radioactivity” (New York and London, 1963), soon after he had published a first long chapter entitled “Atmospheric Chemistry” in a book in 1958 (*Junge*, 1958). This clear term identifies a sub-discipline of chemistry and not meteorology or physics. The “discipline” was called “chemical meteorology” before that time. However, before the 1950s, chemical meteorology was mainly looking for the relationship between condensation nuclei, its chemical composition, and the formation of clouds and rain.

Basically, there is no need to prefix the sciences (environmental, ecological, geo-, air, hydro-, etc.). Other sciences such as geology and meteorology, for example, can be reduced to the natural sciences. It is worth noting the language roots of “geo” and “meteor”. In Greek, γή (ge) means earth, land, and ground (poetically γαία (gaia))[1]. In the composition of words, Greek γέα (gea), γης

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[1] In Latin, the word for earth, land, and ground is “tellūs” (in sense of the celestial body) and “terra” (in sense of matter).

(ges), γην (gen), and γεω (geo) occur. Already before the year 600 B.C., the Greek word μετέωρος (metéoros) was in use, meaning "a thing in the air, altitude, or above ground"[2]. Until the end of the eighteenth century, μετέωρος denoted all celestial phenomena: aqueous, vaporous, solid, and light. *Aristotle*'s "Meteorologia" is a book on natural philosophy (in modern terms: earth science). Hence, in a more narrow (historical) sense, geosciences (geology, geography, geochemistry, geophysics, etc.) deal with the subject of the solid earth (the geosphere). Geochemistry studies the composition and alterations of the solid matter of the earth; geology is the scientific study of the origin, history, and structure of the earth; geography studies the earth and its features and the distribution of life on the earth, including human life and the effects of human activity. Geography (which is an old discipline, founded as a modern science by Humboldt) is thus the science of the earth's surface – the physical and human landscapes, the processes that affect them, how and why they change over time, and how and why they vary spatially. In other words, geography is an interfacial science between the solid and the gaseous earth (the atmosphere); indeed some geographers consider climatology to be a sub-discipline of geography (and vice versa, some meteorologists do not include climatology in meteorology). Finally, the liquid earth (the hydrosphere) is the subject of hydrology[3]. It is important to note that gaseous water (vapor) in the atmosphere is not considered to be an object in hydrology, but liquid water in the atmosphere is. Logically we now can state that the science of studying the "gaseous earth" (atmosphere) is meteorology. Older (synonymous) terms for the science of the study of earth's atmosphere are aerology and atmospherology. For example, meteorology has been defined as the physics and chemistry of the atmosphere (*Scharnow et al.*, 1965) but also reduced to just the physics of the atmosphere (*Liljequist* and *Cehak*, 1984). Some definitions focus meteorology on weather processes and forecasting, which surely was the beginning of that science. A satisfactory definition is "the study dealing with the phenomena of the atmosphere, including physics, chemistry, and dynamics, extending to the effects of the atmosphere on the earth's surface and the oceans". Hence, meteorology is more than "only" the physics and chemistry of the atmosphere. Nowadays, the term "atmospheric sciences" is also used to summarize all the sub-disciplines needed to explain atmospheric phenomena and processes. To return to chemistry, (atmospheric)

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[2] μετέωρολογια [metéorologia] means the "science of celestial, heavenly things" but also "vague talk" and "philosophical shenanigans" (*Benselers* Greek-German School Dictionary (*Griechisch-Deutsches Schulwörterbuch*, Leipzig and Berlin 1911).

[3] In Greek ϋδωρ (idor) means originally rain water and generally water; it appears in composite words as ύδρο (idro). This is the derivation of the prefix "hydro" (the pronunciation of Greek letter ϋ and ύ is like "hy"). In Latin, water as a substance is "aqua", but natural waters are referred to as "unda", derived from the Greek "hy-dor" (ϋδωρ); ϋδρα (Greek), and hydra (Latin) is the many-headed water snake in Greek mythology. From Gothic "vato" and Old High German "waz-ar", are clearly seen the roots of English "water" and German "Wasser".

chemistry is just one of the sciences to understand the (chemical) processes in the atmosphere (see above for definition of atmospheric chemistry).

Antoine Lavoisier, who revolutionized the science of chemistry in the eighteenth century and replaced the mythical "phlogiston" with the term and concept of oxygen, clearly understood the importance of accurate definitions. In his words: "We cannot improve the language of any science without at the same time improving the science itself; nor can we, on the other hand, improve a science without improving the language or nomenclature" (*Lavoisier*, 1789). *Imre Lakatos* (1981) wrote "Philosophy of science without history is empty; history of science without philosophy is blind".

2. Air and atmosphere – a multiphase and multi-component system

The typical dictionary definition of atmosphere is "the mixture of gases surrounding the earth and other planets" or "the whole mass of an aeriform fluid surrounding the earth". The terms air and atmosphere are widely used as synonyms. The word "air" derives from Greek άήρ and Latin aer or ær. The term "atmosphere", however, originated from the Greek ατμόσ (= vapor) and σφαιρα (= sphere), and was not regularly used before the beginning of the nineteenth century. The Dutch astronomer and mathematician *Willebrord van Roijen Snell* translated the term "damphooghde" into Latin "atmosphæra" in 1608. *Otto von Guericke*, who invented the air pump and worked on his famous experiments concerning the physics of the air in the 1650s, used the term "ærea sphæra" (*Guericke*, 1672). In addition, in the nineteenth century the term "Air Ocean" was also used, in analogy to the sea.

It makes even more sense to define the atmosphere as being the reservoir (space) surrounding our (and any) planet, and air to be the mixture of substances filling the atmospheric space. With this in mind, the term air chemistry is more adequate than atmospheric chemistry. From a chemical point of view, it is possible to say that air is the substrate with which the atmosphere is filled. This is in analogy to the hydrosphere where water is the substance. Furthermore, air is an atmospheric suspension containing different gaseous, liquid (water droplets), and solid (dust particles) substances, and therefore, it provides a multiphase and multi-component chemical system. Solar radiation is the sole primary driving force in creating gradients in pressure, temperature, and concentration which result in transport, phase transfer, and chemical processes (*Fig. 1*).

The physical and chemical status of the atmosphere is called climate (see Section 4 for details). Supposing that the incoming solar radiation shows no trend over several hundred years, and accepting that natural biogenic and geogenic processes vary but also do not show trends on these time scales, it is only mankind's influence on land use and emissions into the atmosphere that changes air chemical composition, and thus, the climate. Human activities have an influence on natural processes (biological, such as plant growth and diversity, and physical, such as radiation budget), resulting in a cascade of consequent physical and chemical developments (feedback).

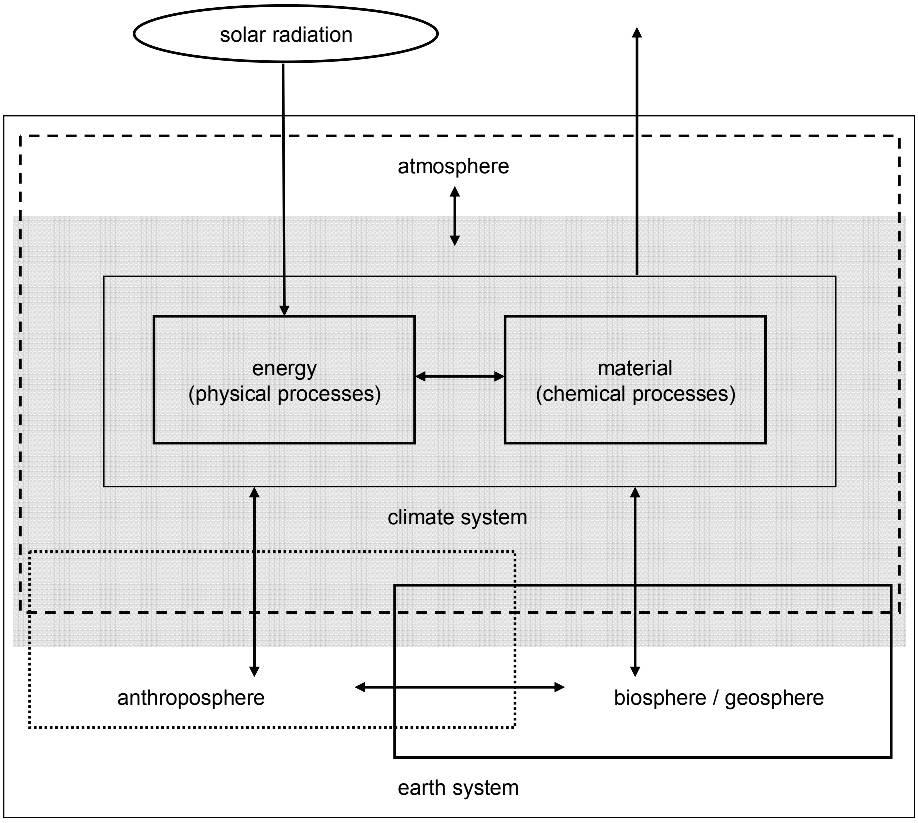

Fig. 1: Scheme of the physico-chemical interactions between the atmosphere, biosphere, and anthroposphere (the climate system).

We will initially set the climate system in a chain of subsystems:

Cosmic system → solar system → earth system → climate system →
→ sub-systems (e.g., atmosphere).

In other words, each system to be defined lies in another "mother" system, the surrounding or environment where an exchange of energy and material is realized via the interfaces. Consequently, there are no fully closed systems in our world.

The chemical composition of air depends on the natural and man-made sources of the constituents (their distribution and source strength in time and space) as well as on the physical (e.g., radiation, temperature, humidity, wind) and chemical conditions (other trace species), which determine transportation and transformation. Thus, atmospheric chemistry is not a pure chemistry but it also includes other disciplines which are important for describing the interactions between atmosphere and other surrounding reservoirs (biosphere, hydrosphere, etc.). Measurements of chemical and physical parameters in air will always contain a "geographical" component, i.e., the particularities of the locality. That is why the terms "chemical weather" and "chemical climate" have been introduced. For example, diurnal variation of the concentration of a substance may occur for different reasons. Therefore, general conclusions or transfer of results to other sites should be done with care. On the other hand, the basic task in atmospheric chemistry is not only to present local results of chemical composition and its variation in time, but also to find general relationships between trace species and their behavior under different conditions.

Without discussing the biological and physical processes within the climate system in any details, the chemical composition of the atmosphere and its variation in time and space, as well as its trends, are essential for an understanding of climate change. Atmospheric substances with their physical and chemical properties will have many effects in the climate system; we list the most important ones together with their impacts (there are many more impacts, parallel and synergistic effects):

- formation of cloud condensation nuclei (CCN) and subsequent cloud droplets: hydrological cycle;
- greenhouse gases (GHG): radiative interaction (warming the atmosphere);
- ozone-depleting substances (ODS): radiative interaction (increasing UV radiation penetration into the lower troposphere);
- formation of atmospheric aerosol: radiative interaction (cooling the atmosphere);
- oxidation capacity: lifetime of pollutants;
- acidity: chemical weathering;
- toxicity: poisoning the environment (affecting life functions and biodiversity);
- nutrition: bioavailability of compounds essential for life but also eutrophication.

We see that the climate system has physical and chemical components, interacting and (at least partly) determining each other. Physical quantities in the climate system show strong influences on chemical processes:

- temperature and pressure: reactions, rates, and (chemical and phase) equilibria;
- radiation (wavelength and intensity): photochemistry,
- motion: fluxes of matter (bringing substance together for chemical reactions).

Let us now turn to the atmosphere as a multiphase system. While gases and particles (from molecules via molecular clusters to nano- and micro-particles) are always present in the air, although with changing concentrations, condensed water (hydrometeors) is occasionally present in air, depending on the presence of so-called cloud condensation nuclei (CCN) and water vapor supersaturation at the site of fog and cloud formation. With the formation, transportation, and evaporation of clouds, huge amounts of atmospheric energy are transferred. This results in changing radiation transfer and thus "makes" the weather, and, on a long-term scale, the climate. Furthermore, clouds provide an effective "chemical reactor" and transport medium and cause redistribution of trace species after evaporation. When precipitating, clouds remove trace substances from the air (we term it wet deposition) to the earth's surface. As a consequence, beside the continuous process of dry deposition, clouds may occasionally lead to large inputs of trace substances into ecosystems. The amount of condensed water in clouds and fog is very small, with around 1 g/m^3 air or, in the dimensionless term, liquid water content $1 \cdot 10^{-6}$ (identical with 1 ppb). Thus, 99.99% or more of total atmospheric water remains in the gaseous phase of an air parcel. Hydrometeors may be solid (ice crystals in different shapes and forms) or liquid (droplets ranging from a few μm up to some tens of μm). We distinguish the phenomenon of hydrometeors into clouds, fog, and precipitation (rain, drizzle, snow, hail, etc.). This atmospheric water is always a chemical aqueous solution where the concentration of dissolved trace matter (related to the bulk quantity of water) is up to several orders of magnitude higher than in the gaseous state of air. This analytical fact is the simple explanation why collection and chemical analysis of hydrometeors began much earlier than gas-phase measurements.

Due to permanent motion, namely advection and turbulent diffusion, having stochastic characteristics on different time and spatial scales, it is extremely complicated to model chemistry and transport (so-called chemistry-transport models, CTM, which are also a basis for climate modeling) in space and time. At the earth-air interface, exchange of matter occurs, emission as well as deposition.

3. Changes in air chemical composition: air pollution

Changes in the chemical composition of air caused by humans are termed air pollution. The terms air pollution and pollutant need some comments. To start with, the term pollutant should be used only for man-made (anthropogenic) emitted substances, despite the fact that most of them are also of natural origin. Air pollution represents a deviation from a natural chemical composition of air (providing a reference level) at a given site. Depending on the residence time of the pollutant, we can characterize the scale of pollution from local via regional to global. Air pollution nowadays is a global phenomenon because long-lived pollutants can be found to be increasing at any sites of the globe. Remote air just means that the site is located far away from the sources of emissions and, consequently, this air has lower concentrations of short-lived (reactive) substances compared with sites close to sources of pollutants. Although polluted air is human-influenced air, clean air is not synonymous with natural air. The natural atmosphere no longer exists; it was the chemical composition of air without man-made influences. However, this definition is also not exact, because humans are part of nature. In nature situations may occur, such as volcanic eruptions, sand storms and biomass burning, where the air is being "polluted" (rendered unwholesome by contaminants) or in other words, concentrations of substances of natural origin are increased. Therefore, the reference state of natural air is a climatological figure where a mean value with its variation must be considered. The term clean air is also used politically in air pollution control as a target, i.e., to make our air cleaner (or less polluted) in the sense of pollutant abatement. A clean atmosphere is a political target, it represents an air chemical composition (defined in time and scale) which should permit sustainable development. The largest difficulty, however, lies in the definition of what sustainable means. This term comprises the whole range of categories from simple scientific questions (for example, impact threshold) to political decisions (global ecomanagement) and also to philosophical questions (for example, what human life needs).

Therefore, air pollution in terms of the changing chemical composition of the atmosphere must be identified through a problem, not simply by measured concentrations. The problem lies between "dangerous" and "acceptable" climate impact, a definition that is beyond the direct role of the scientific community despite the fact that scientists have many ideas about it (*Schneider*, 2006).

Doubtless, the air of settlements and towns was extremely polluted in the past. Heavy metals have been found in Greenland ice cores dating back to the Roman Empire; thus demonstrating that metallurgical operations of immense volume took place in that era. The modern quality of air pollution, since the industrial revolution of the nineteenth century, is best characterized by a continuous worldwide increase in emissions as the era of fossil fuel combustion began. In the last 150 years, serious air pollution problems have been described, analyzed, and solved (to an extent, by end-of-pipe technologies), such as soot, dust and smoke plagues, winter and summer smog, and acid rain. The chemicals (or emitted compounds) behind these phenomena are soot, sulfur dioxide (SO_2), nitrogen oxides (NO_x), and volatile organic compounds (VOC) – all of them short-lived species, but because of global use of fossil fuels, the pollution problem is distributed globally. Some air pollution problems connected with long-lived compounds – now in terms of persistent compounds, such as agricultural chemicals (food chain accumulation) and halogenated organic compounds (ozone layer destruction) – have been solved by legislation banning their use. Unfortunately, some long-lived emitted compounds, the so-called greenhouse gases, have increased significantly in global mean concentration (*Table 1*).

Table 1. Increase of some climate relevant gases in air (in ppb)

Substance	**1850**	**2008**	**Increase (in ppb)**	**Increase (in %)**
Carbon dioxide CO_2	285,000	383,000	98,000	34
Methane CH_4	700	1700	1000	143
Nitrous oxide N_2O	300	320	20	7
Ozone O_3	10	20	10	100

Ozone is not among long-lived species but globally it is a secondary product from methane oxidation. CH_4 and N_2O (mainly byproducts of agriculture) as well as CO_2 (a byproduct of fossil fuel combustion) are coupled with the two columns of human existence; food and energy. There is no doubt that the CO_2 problem can be solved only through sustainable technology change. It should be noted that these three pollutants (CO_2, CH_4, and N_2O), even with much higher atmospheric concentrations, are harmless for life. They are in fact key substances in biogeochemical cycles, but there is also no doubt that these three substances contribute to about 90% of global warming. Besides, N_2O acts as an ozone-depleting substance in the stratosphere, while CH_4 contributes to O_3 formation in the troposphere and is probably responsible for up to 80% of the global O_3 increase. Future air pollution control is synonymous with climate control and must be focused on CO_2, CH_4, and N_2O.

4. *Climate and the climate system*

The physical and chemical status of the atmosphere on a medium-range timescale (by definition 30 years) is called climate. The expression climate is currently often used in connection with climate change (a term widely popularized in recent years). Synonymously, the expression climate alteration (a more scientific term) is used. In the United Nations Framework Agreement on Climate Change, which was signed by 34 countries and was approved in New York on May 9, 1992, climate change is defined as “changes in climate, that are directly or indirectly attributable to anthropogenic activities, which change the composition of the atmosphere and which add up to the natural climatic changes observed over comparable periods of time”. This definition is circumscribed, as natural processes are included in the sense of climate change but not in the sense of climate alteration. However, it can only be observed that climate change is due to all factors. Possible anthropogenic factors having an effect on climate (and, therefore, on its alteration) are manifold and stretch from an alteration in the use of land (altered surface properties) to emissions, whereas direct energetic influences (e.g., warmth islands) are at most (first) of local importance. Thus, a climatic change is the difference between two states of climate. A state of climate is described through the static condition of the climatic system. A climatic fluctuation can therefore be described as a periodic climatic change, independent of the respective scale of time. Climate is a function of space and time – and is continuously changing (*Schneider-Carius*, 1961).

The climate system is too complex to be clearly mathematically described. In all likelihood, this will be valid in the future as well. Our knowledge about many single processes in this system is still imperfect. Nevertheless, immense progress has been made in the past twenty years and the description of the principal processes is considered as assured (*Hansen et al.*, 2007). It is all the more surprising, that a (quasi)-linear relationship between the GMT (global mean temperature) and the cumulative CO_2 emission was found; on the other hand, the atmospheric CO_2 concentration also shows a strong linear relation with the cumulative CO_2 emission since about 1910 (*Möller*, 2010).

Both climate researchers and historians of climate science have conceived of climate as a stable and well defined category, but such a conception is flawed. Of particular interest becomes the impact of climate on human beings and the environment. In modern climate research, at the close of the twentieth century, the concept of climate lost its temporal stability. Instead, climate change has become a core feature of the understanding of climate and a focus of research interest. Climate has also lost its immediate association with specific geographical places and become global. Interest is now focused on the impact of human beings on climate (*Heymann*, 2009).

Alexander von Humboldt's often cited definition of climate shows three remarkable particularities: the relation of changes instead of a mean status (as defined later in the nineteenth century), the inclusion of the chemical status of the atmosphere (by using the terms cleanness and pollution), and the restriction on parameters affecting human organisms but also the whole biosphere:

> "The term climate, taken in its most general sense, indicates all the changes in the atmosphere which sensibly affect our organs, as temperature, humidity, variations in the barometric pressure, the calm state of the air or the action of opposite winds, the amount of electric tension, the purity of the atmosphere or its admixture with more or less noxious gaseous exhalations, and, finally the degree of ordinary transparency and clearness of the sky, which is not only important with respect to the increased radiation from the earth, the organic development of plants, and the ripening of fruits, but also with reference to its influence on the feeling and mental condition of men." (*Humboldt*, 1850–1852)

The father of the physical definition of climate is *Julius Hann* who defined the climate as "the entirety of meteorological phenomena which describe the mean status of the atmosphere at any given point of the earth's surface"[4] (*Hann*,

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[4] " …Gesamtheit meteorologischer Erscheinungen, welche den mittleren Zustand der Atmosphäre an irgendeiner Stelle der Erdoberfläche charakterisieren."

1883). He further stated that “climate is the entirety of the weathers[5] of a longer or shorter period as occurring on average at a given time of the year”. The aim of climatology is to establish the mean states of the atmosphere over different parts of the earth’s surface, including describing its variations (anomalies) within longer periods for the same location (*Hann*, 1883). *Hann* also introduced the term climatic element emphasizing that measurements must characterize these through numerical values. He also freed the term “climate” from the close relation to humans and plants, and related it to the time before life appeared on the earth (without using the term palaeoclimate).

Köppen (1906) adopted *Hann*’s definition (“mean weather at a given location”) but stated in contrast to *Hann* that it is meaningless to define a climate without focus on human beings; hence only factors (elements) influencing “organic life” should be considered. *Köppen* explicitly presents a “second definition” of climate “… as the entirety of atmospheric conditions, which make a location on earth more or less habitable for men, animals and plants” (*Köppen*, 1906). He wrote that “with the progress in knowledge new subjects will be included in the number of climatic elements when their geographical characteristics are unveiled” (*Köppen*, 1923). During the last 60 years the idea of climate has broadened in so far that, in the definition of climate, apart from the mean value, higher statistical moments are included. According to the new definition, climate describes the “statistical behavior of the atmosphere, which is characteristic for a relatively large temporal order of magnitude” (*Hantel et al.*, 1987).

For an understanding of the dynamics of climate, that is, the processes that determine the average state and variability of the atmosphere over longer periods, the meteorological definition is inadequate, as over longer periods, changes in the atmosphere are considerably affected by interdependencies of the atmosphere, the ocean, vegetation and ice masses (*Claußen*, 2006). For this reason, in climate dynamics, climate is defined by the state and the statistical behavior of the climatic system, as can be read in modern textbooks on meteorology and climate physics (e.g., *Peixoto* and *Oort*, 1992; *Kraus*, 2004; *Lutgens et al.*, 2009). *Claußen* (2006) distinguishes between a meteorological and a system-analytical definition. Note that it is essential to include the statistics into the idea of climate, i.e., climate means not simply the “mean weather”. It follows that

- climate is a function of space and time;
- climate cannot be described as a single unit.

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[5] There is no direct English translation of German Witterung. The translation as “weather” (German Wetter) is not fully correct because Witterung denotes short-term averaged weather (a weather period).

In the history of mankind and the exploration of air and atmosphere, the concept of climate has been subject to change, but also various descriptions have existed at the same time. It is beyond the scope of this paper to address them here. Here, one can conclude that different definitions of climate are also in use. A priori, this is a contradiction, as there is only one climate system on the Earth. Obviously, this results from a pragmatic approach to the cognition and description of the climatic system by

- diverse disciplinary points of view,
- different objectives (e.g., description of subsystems), and/or
- differentiated knowledge of the system relationships.

The climate system can be described (and widely quantified through measurements) by the

- natural energy system,
- hydrologic cycle,
- carbon cycle and
- other biogeochemical cycles.

The subsystems are linked to each other through flows of energy, impulse, and matter. To the flows of matter, the transport of chemical substances and the processes of their transformation need to be added, as far as these substances, e.g., greenhouse gases or nutrients of the biosphere are directly or indirectly related to the energy budget. The definition of the climate system is not derived from superior principles, but is a pragmatic restriction of the subject to be examined by classification in subsystems and interpretation of the respective system environment. The separation of the climate system from its environment is carried out in that way, as no significant flow of matter between the system and its environment occurs on timescales relevant for examination.

In a broader sense, the climate system can be seen as an interlayer within the earth system, buffering a habitable zone from uninhabitable physical and chemical conditions in altitude (upper atmosphere) and depth (deep lithosphere); see *Fig. 2*. In a more narrow sense, the human-habitable zone is limited to the gas-solid interface (earth-surface/atmosphere) with a very small extension of a few tens of meters. The anthroposphere (or noosphere), however, is permanently expending in space – also out of the climate system – due to the fact that humans are creating inhabited and uninhabited closed habitable systems in an uninhabitable surrounding.

In the literature, the total of climate system and anthroposphere is defined as the earth system (*Schellnhuber* and *Wenzel*, 1998; *Schellnhuber*, 1999; *Claußen*, 1998, 2001). *Hantel* (2001) presents a very pregnant definition "The climate is not a subject-matter but a property. Its carrier is the climate system. The climate is the entirety of the properties of the climate system."

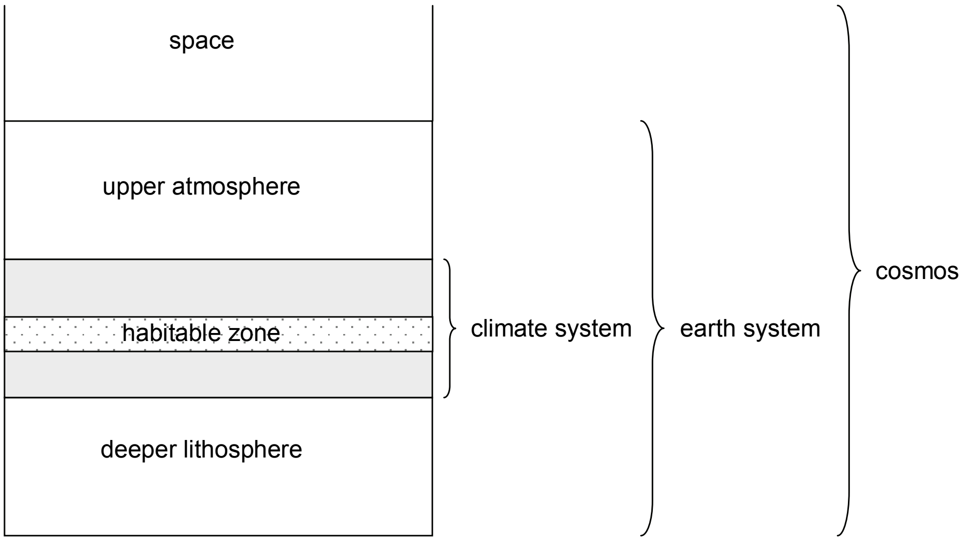

Fig. 2. Layer structure of the climate system.

According to the definition of "chemical weather" given by *Lawrence et al.* (2005), we arrive at a formal definition of chemical climate as: "The synthesis of chemical weather conditions in a given area, characterized by long-term statistics (mean values, variances, probabilities of extreme values, etc.) of the chemical substances in that area."

Instead of "meteorological elements", we have "chemical substances". When – as implicitly mentioned earlier – meteorology feels responsible as a discipline for the description of the chemical state of the atmosphere, the list of "meteorological elements" can simply be expanded to accommodate all relevant chemical variables as well. So as not to raise misunderstandings, "chemical aspects" and not "chemistry" are dealt with the extension of the term climate, since climatology is not a subdiscipline of physics.

I would like to define climate in a general sense as follows:

> Climate describes the mean status of the atmosphere at a given site of the earth's surface, represented by the statistical total properties (mean values, frequencies, durations etc.) of a long enough time period.

It is understood, and, therefore, no differentiation between a meteorological and a system-related definition of climate according to *Claußen* shall be undertaken, that the atmosphere is only one part of the climatic system, and therefore, climate can only correctly be described in a physical-chemical manner taking into consideration material and energetic interdependencies with the other subsystems. Advantageously, a climatic state (i.e., the scientific, broadly mathematical description of the climate) can be defined:

> A climate state is given by the whole description of the statistical status of the internal climate system.

The conclusion that the climate is permanently but slowly changing is true. Now, however, we have the likely situation of man-made abrupt climate change. An abrupt climate change occurs when the climate system is forced into transition to a new state at a rate that is determined by the climate system itself, and which is more rapid than the rate of change of the external forcing. Moreover, with this background we understand climate change better, because it means that one or more of the earth-system determining climate elements is changing by a qualitative jump. In nature there is no jump, as nature itself composes of jumps. The idea of interactive changes from quality to quantity and vice versa is a main principle of dialectics. The fact, that our subjective thinking and the objective world underlies the same laws, and therefore, cannot be contradictory in their results, but correspond, rules our complete theoretical thinking. It is an unconscious and unconditional premise. The materialism of the eighteenth century studied this premise according to its vitally metaphysical character on its content only. It limited itself to the proof that the content of all thinking and knowledge descends from sensual experience, and reconstructed the sentence: *Nihil est in intellectu, quod non fuerit in sensu.* (Nothing is in the mind, which has not been in the senses before). Dialectics as the science of the common laws of all movement was first scientifically postulated by *Friedrich Engels*. Despite the linguistic simplification, we should not forget the complexity of the system: even the most advanced mathematical models running on the biggest (and networked) computers are still unable to produce a true picture of the climate system sufficient to draw reliable conclusions. Experimental validation of the model outputs, however, is only possible after climate change – that is the dilemma! Parts of the model (or the system) can be validated with relevant experimental approaches, for example, cooling of the atmosphere by particulate matter after large volcanic eruptions.

5. Chemical evolution and the role of humans

The term evolution was used first in the field of biology at the end of the nineteenth century. In the context of biology, evolution is simply the genetic change in populations of organisms over successive generations. Evolution is widely understood as a process that results in greater quality or complexity (a process in which something passes by degrees to a different stage, especially a more advanced or mature stage). However, depending on the situation, the complexity of organisms can increase, decrease, or stay the same, and all three of these trends have been observed in biological evolution. Nowadays, the word has a number of different meanings in different fields. The term chemical evolution is not well-defined and used in different senses.

Chemical evolution is not simply the change and transformation of chemical elements, molecules, and compounds as it is often asserted – that is the nature of chemistry itself. It is essentially the process by which increasingly complex elements, molecules, and compounds develop from the simpler chemical elements that were created in the Big Bang. The chemical history of the universe began with the generation of simple chemicals in the Big Bang. Depending on the size and density of the star, the fusion reactions can end with the formation of carbon or they can continue to form all the elements up to iron.

The origin of life is a necessary precursor for biological evolution, but understanding that evolution occurred once organisms appeared and investigating how this happens do not depend on understanding exactly how life began. The current scientific consensus is that the complex biochemistry that makes up life came from simpler chemical reactions, but it is unclear how this occurred. Not much is certain about the earliest developments in life, the structure of the first living things, or the identity and nature of any last universal common ancestor or ancestral gene pool. Consequently, there is no scientific consensus on how life began, but proposals include self-replicating molecules such as RNA, and the assembly of simple cells. Astronomers have recently discovered the existence of complex organic molecules in space. Small organic molecules were found to have evolved into complex aromatic molecules over a period of several thousand years. Chemical evolution is an exciting topic of study, because it yields insight into the processes which lead to the generation of the chemical materials essential for the development of life. If the chemical evolution of organic molecules is a universal process, life is unlikely a uniquely terrestrial phenomenon, instead, it is likely to be found wherever the essential chemical ingredients occur.

In colloquial contexts, evolution usually refers to development over a long time scale, and the question is not important whether evolution tends toward more complexity. Many definitions tend to postulate or assume that complexity expresses a condition of numerous elements in a system and numerous forms of relationships among the elements. At the same time, deciding on what is complex and what is simple is relative and changes with time.

A modern understanding of evolution includes continuous development, but also leaps (catastrophes). This is referred to as “transformation of quantity into quality” (dialectic leap) and may characterize the current discussion on the impacts of climate change. However, it is hard to envisage a physical situation in which a quantifiable parameter can increase indefinitely without a critical condition occurring. Physical processes – starting with the Big Bang – created the first atoms (which form chemical elements) and physical conditions, permanently affecting the subsequent chemical and biological evolution. Compared with the Big Bang as the beginning of physical evolution, the creation of molecules and life can be referred to as the starting point of a chemical and biological evolution, respectively. Life became a geological force with oxygenic photosynthesis and created an interactive feedback with chemical and physical evolution. After forming the geosphere and the first atmosphere in the sense of a potentially habitable system, and later the biosphere with the modern atmosphere, a habitable climate system evolved.

If water was as common in the solar system as it is implied by the facts, that would suggest that there were many environments in the solar system where the conditions were right for the development of life. What is life? For instance, *Lynn Margulis* (quoted by *Horgan*, 1997) has stated that to proceed "…from a bacterium to people is less of a step than to go from a mixture of amino acids to that bacterium". At the beginning of the seventeenth century, the ultimate origin of life was considered to be primarily a theological issue. However, it was thought possible that small creatures, such as maggots and even mice, could arise from non-living material by spontaneous generation, a theory first propounded by Aristotle. Today, there is no doubt that bacterial life is created, exists, and survives in space (*Maurette*, 2006). But, what is life? Where did we come from? These two fundamental questions remain (still) unanswered in science. The existence of humans (and all animals) depends on free oxygen in the atmosphere, and this compound is almost completely produced from oceanic cyanobacteria. Hence, the origin of life lies in the darkness of the evolution of molecules in structured systems (a chemical plant that we call a cell) to provide work-sharing synthesis via non-equilibrium electron transfer processes (in other terms, redox processes). Cells represent a dissipative structure, whose organization and stability is provided by irreversible processes running far from equilibrium. *Falkowski* and *Godfrey* (2008) state that the question posed above reflects our ignorance of basic chemistry of the electron transfers, that bring the ensemble of molecules in cells to "life".

But life created a further dimension, human intelligence, which becomes another geological force (human evolution – today approaching a critical condition which we call crisis). Human intelligence disengaged humankind from the rigorous necessities of nature and provided unlimited scope for reproduction (at least in the past). Man, in all his activities and social organizations is part of, and cannot stand in opposition to or be a detached or external observer of, the nature. However, the new dimension (or quality) of human intelligence as a result of biological evolution – without some global ecomanagement – could change the climate system in a direction not providing the internal principle of self-preservation. Mankind converts the biosphere into a noosphere. Chemical evolution is now interloped with human evolution. Changing fluxes and concentrations of chemicals in bio- (or rather noo-) geochemical cycles with a subsequent changing climate system seems to be the creation of a human-chemical evolution.

Vernadsky was the first to propose the idea of biogeochemical cycling (having asked: What is the impact of life on geology and chemistry of the earth?). *Vernadsky*, who had met *Suess* in 1911, popularized the term biosphere in his book The Biosphere (first published in Russian in 1926, not translated into a full English version until more than 60 years later), hypothesizing that life is the geological force that shapes the earth (*Vernadsky*, 1926; 1944; 1945). *Vernadsky* first took the term “noosphere” in 1931, as a new dimension of the biosphere under the evolutionary influence of humankind (*Vernadsky*, 1944). He wrote: “The Noosphere is the last of many stages in the evolution of the biosphere in geological history” (*Vernadsky*, 1945). The biosphere became a real geological force that is changing the face of the earth, and the biosphere is changing into the noosphere. In Vernadsky’s interpretation, the noosphere is a new evolutionary stage of the biosphere, when human reason will provide further sustainable development both of humanity and the global environment

> “In our century the biosphere has acquired an entirely new meaning; it is being revealed as a planetary phenomenon of cosmic character... In the twentieth century, man, for the first time in the history of earth, knew and embraced the whole biosphere, completed the geographic map of the planet earth, and colonized its whole surface. Mankind became a single totality in the life on earth... The noosphere is the last of many stages in the evolution of the biosphere in geological history.” (*Vernadsky*, 1945).

Today the term “anthroposphere” is also used. The idea of a close interrelation between the humans and the biosphere is topical in understanding the “earth system”, i.e., the climate change, and is used by *Schellnhuber* (1999) with the terminology “global mind” and by *Crutzen* and *Stoermer* (2000) with “anthropocene” to characterize the present epoch[6].

We also may state that the climate is first a result of geophysical and chemical processes, and that evolving life adapts to these conditions. We know that oxygen is definitively a result of the photosynthesis of plants, hence it is of biological origin. Without any doubt we can state that life did change the climate – with “sense” or without –, and that feedback did influence the evolution of life. What is life or a living thing? Each living thing is composed of “lifeless” molecules, independently of its dimension and complexity, which are subject to the physical and chemical laws that are characteristic of inanimate bodies. A living thing (to avoid the definition of “life”) has certain characteristics which are common to living matter but not found in nonliving objects, such as:

- the capacity for self-replication (they grow and reproduce in forms identical in mass, shape, and internal structure),
- the ability to extract, transform, and use energy from their environment (in the form of nutrients and sunlight), and
- an organized structure, where each component unit has a specific purpose or function.

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[6] *Zalasiewicz et al.* (2008) published the first proposal for the formal adoption of the Anthropocene epoch by geologists, and this adoption is now pending.

All these characteristics result in non-equilibrium with themselves and with their environment. Non-living things tend to exist in equilibrium with their surroundings. An earth-like planet, not developing life, would therefore be oxidized with aging. Only photolytic dissociation and thermal degradation would occur, depending on incoming radiation (distance from the Sun) and available thermal heat (planetary size). At a final stage, the atmosphere would be composed solely of oxides and acids. The large CO_2 content would increase the atmospheric temperature. Missing free oxygen (because it is fixed in oxides, volatile and non-volatile) in the atmosphere (and subsequent ozone) would prevent a UV-absorbing layer and, therefore, allow almost all photodissociations close to the planetary surface. With time, all water would disappear due to photolytic splitting into hydrogen, which escapes into space. The oxygen from water splitting cannot accumulate until all primordial reduced atoms are oxidized. Finally, free oxygen could be possible in the case of an excess over the equivalent of atoms in reduced state. Conversely, no free oxygen would occur when the reduction equivalent exceeds that of oxygen. The planet becomes irreversibly uninhabitable, especially because of absence of water. There is no doubt that this process would occur over a long time, potentially over the planetary lifetime.

The non-living world tends to dissipate structures and, therefore, to increase entropy. However, because of its huge energy pools, the earth's internal geothermal heat and the Sun's radiation, both are likely to remain over the entire expected lifetime of the earth, provide gradients to force geochemical cycling with the irreversible direction of oxidation and acidification (*Fig. 3*).

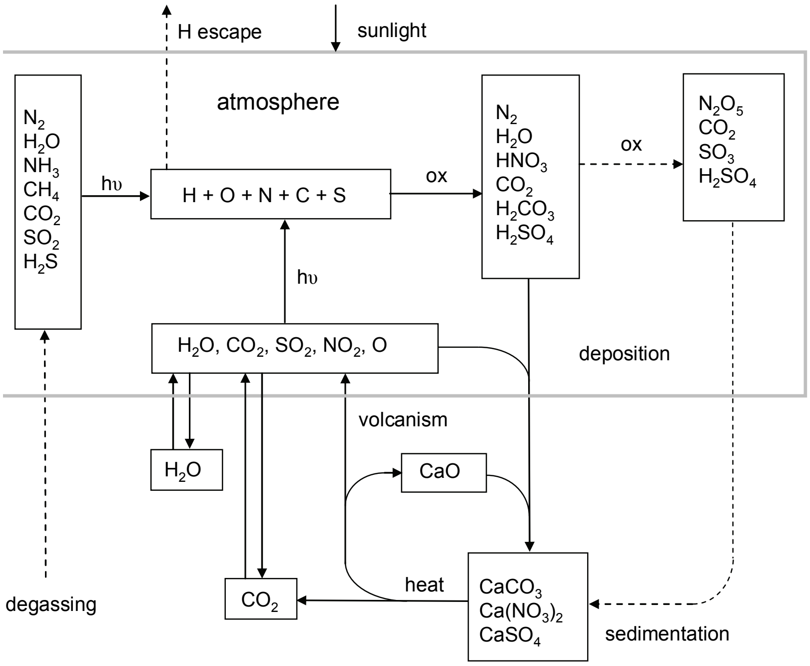

Fig. 3. Scheme of geochemical cycling over geological epochs.

Only the living world is able to reduce entropy by creating structures, or in other words, to move the system back from equilibrium. Indeed, the central role of life (more exactly, autotrophic organisms) is to maintain the cycle shown in *Fig. 4*, starting with light-induced electrolytic water splitting, and subsequent parallel – but, important to note, in separated organs – reduction of CO_2 into hydrocarbons and the oxidation of them back to CO_2. Globally, this cycle represents a dynamic equilibrium. Consequently, there are established stationary concentrations in the climate system.

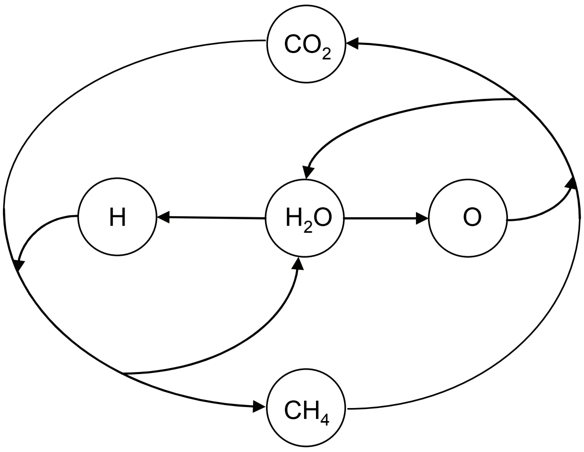

Fig. 4. Scheme of the water-carbon interlinked cycle.

Under the evolution of the earth and the climate system, we will simply understand the historical development from earliest times until the present. Theories for how the atmosphere and ocean formed must begin with an idea of how the Earth itself originated (*Kasting*, 1993). An understanding of our atmosphere and climate system is incomplete without going into the past. "The farther backward you can look the farther forward you can see." (*Winston Churchill*).

6. Conclusion: toward sustainability

In recent decades, humans have become a very important force in the earth system, demonstrating that emissions and land use changes are the cause of many of our environmental issues. These emissions are responsible for the major global reorganizations of biogeochemical cycles. With humans as part of nature and the evolution of a man-made changed earth's system, we also have to accept that we are unable to remove the present system into a preindustrial or even prehuman state, because this means disestablishing humans. The key question is which parameters of the climate system allow the existence of humans (and how many) under which specific conditions.

The chemical composition of air is now contributed by both natural and man-made sources. The chemical composition of air has been changing since the settlement of humans. In addition to the scale problem (from local to global), we have to regard the time scale. Natural climate variations (e.g., due to ice ages) had a minimum time scale of 10.000 years. The man-made changes in our atmosphere over the last 2000 years were relatively small before the 1850s. In the past 150 years (but almost all after 1950), however, the chemical composition has changed drastically. For many atmospheric compounds anthropogenic emissions have grown to the same or even larger order of magnitude than natural ones. Because of the huge population density, the need (or consumption)[7] of materials and energy has drastically forced the earth's system. “The Great Acceleration is reaching criticality. Whatever unfolds, the next few decades will surely be a tipping point in the evolution of the Anthropocene” write *Steffen et al*. (2007).

The time scale of the adaptation and restoration of natural systems is much larger than the time scale of man-made stresses (or changes) to the climate system. We should not forget that “nature” cannot assess its own condition. In other words, the biosphere will accept all chemical and physical conditions, even worse (catastrophic) ones. Only humans possess the facility to evaluate the situation, accepting it or not, and coming to the conclusion of making it sustainable. But humans also do have all the facilities to turn the “chemical revolution” into a sustainable chemical evolution. That does not mean “back to nature”.

Let us define a sustainable society as one that balances the environment, other life forms, and human interactions over an indefinite time period. A global sustainable chemistry first needs a paradigm change, namely the awareness that growth drives each system towards a catastrophe. The basic principle of global sustainable chemistry, however, 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).

The value in identifying current trends and viewing them in a historical light is that the results can be used to inform ongoing policy and investment decisions. The planet could not support the six to seven billion people that exist today without the commercialization of first coal, and then oil and gas. Whereas historical increases in energy consumption had been gradual, once indust–rialization occurred, the rate of consumption increased dramatically over a

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[7] This is an interesting question: do we need all this consumption? What consumption do we need to realize a *cultural* life? Of course we move from natural (earth sciences) to a social and political dimension (life sciences) in answering these questions. But there is a huge potential to economize and save resources in answering these questions and implementing it. *Karl Marx* wrote “The philosophers have only managed to interpret the world in various ways. The point is to change it”. (This is still fixed in the main hall of the central building of the Humboldt University in Berlin, Germany). However, the key point is how and in which direction we have to change the world to receive sustainability.

period of just a few generations. In fact, the rate of energy use from all sources has been growing even faster than, the world population growth. Thus, from 1970 to 1995, energy use increased at a rate of 2.5% per year (doubling in every 30 years) compared with a worldwide population growth of 1.7% per year (doubling in every 40 years). During the next 20 years, energy use is projected to increase at a rate of 4.5% per year (doubling in every 16 years) compared with a population growth rate of 1.3% per year (doubling in every 54 years). Although about 50% of all the solar energy captured by worldwide photosynthesis is used by humans, this amount is still inadequate to meet all human needs for food and other purposes (*Pimentel* and *Pimentel*, 2007). Hence, only the direct conversion of solar energy (heat and radiation) into electricity and chemically stored energy can overcome the future gap.

A global sustainable chemistry first needs a paradigm change, namely the awareness that growth drives each system towards a catastrophe. Sustainable chemistry, also known as green chemistry, is a chemical philosophy encouraging the design of products and processes that reduce or eliminate the use and generation of hazardous substances.

Permanent growth – as stated by politicians – will not solve life problems such as employment; this is a question of reorganizing society. In nature, many processes follow a simple law Eq. (1), which expresses that the change of a quantity N (for example population, mass, energy) is proportional to the quantity itself. In other words, exponential growth occurs when some quantity regularly increases by a fixed percentage. The proportionality coefficient λ characterizes the process (biology, chemistry, physics, economy, etc.) as follows:

$$\frac{dN}{dt} = \lambda N = F, \qquad N(t) = N_o \exp(\lambda t). \tag{1}$$

We see that dN/dt denotes a flux F; according to the sign, it could result in a growth (positive sign) or decline (negative sign). It is clearly seen that a negative flux will end with $N(t)=0$ with $t \to \infty$ when there is no permanent source (positive flux) of N to maintain a pool of this quantity. After productivity (expressed as constant annual turnover) satisfies social consumption needs, stationary conditions are then achievable, i.e., λ becomes zero in Eq. (1). Naturally, the human population will (and must) tend to a constant number. This limitation process is likely to go on over the next 200 years. Another limitation must be set through per capita consumption to provide social and cultural standards. The growth, however, is going on this century. Without revolutionary technological changes, the climate becomes out of control. As stated above, the atmospheric CO_2 increase must be stopped within the next few decades. There are several ways, simultaneously linked with the solution of the energy problem:

- reducing fossil fuel combustion and (drastically) replaceing by solar energy (e.g., desertec conception),
- CO_2 capture from exhaust gases and storage (CCS technology) – likely for future reuse and cycling (CCC technology), and
- CO_2 capture from ambient air and recycling (or sequestration achieving a negative flux) to establish a global carbon/CO_2 man-made cycle.

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