Journal of Natural Sciences Research
ISSN 2222-XXXX(Online)
Vol. X, No.X, 2026

On the Physical Chemistry of the Greenhouse Effect

Helmut Ullmann[1], Martin Bülow[2*]

1. D-01705 Freital, Burgwartstrasse 13a, Germany
2. Leibniz Soziietät der Wissenschaften zu Berlin e.V., Langenbeck-Virchow-Haus, Luisenstraße 58/59, Germany

* E-mail of the corresponding author: mertnblw@gmail.com

Abstract

In the present, a looming catastrophe is feared due to climate change. People are looking for ways to prevent the anticipated warming of the climate. The primary approach promoted is the reduction of carbon dioxide, CO_2, emissions from human activities. This would require a major transformation of existing ways of life and production. There are legitimate doubts about the reasoning that human-generated CO_2 is the main cause of global warming.

We examined the physico-chemical properties of CO_2 and the so-called greenhouse gases to determine their influence on the heat exchange between the Earth's atmosphere and space. Their role as converters of infrared radiation from the sun into heat is a fact; conversely, they are equally indispensable as transmitters of heat from the Earth's atmosphere into infrared radiation to balance the Earth's radiation equilibrium. The balances of CO_2 exchange with the plant world and the oceans, in comparison with anthropogenic emissions, are critically assessed. The anthropogenic energy flow is four orders of magnitude smaller than that of daily solar radiation. The planetary regulatory system of ice ages and warm periods, which has been operating for hundreds of thousands of years, is being discussed. It must be taken into account in the climate discussion of the past centuries.

Keywords: Greenhouse gases, heat capacity, energy transmitters and emitters, radiation balance, exchange of energy and CO_2

1. Introduction

No other chemical compound is currently attributed with as much harmful impact as carbon dioxide. The basis of climate predictions is the correlation between CO_2 concetration and global temperature rise, which has been observed since the beginning of the Industrial Revolution around 1850 (IPCC 2014 & WMO 2024). During this period, CO_2 levels increased from about 280 to 480 vppm (volume parts per million). By extrapolating this relationship, a climate catastrophe is predicted. Climate research recognizes numerous sources and sinks for CO_2 as well as amplifications or reductions of solar radiation. Some of them can be quantified, others cannot, and additional as-yet-unknown influences are suspected. Climate predictions are evaluated more or less critically. However, the sensitivity of the climate to anthropogenic CO_2 is excluded from any doubt. The significant climate impact of water in its three states of matter in the atmosphere usually goes unmentioned, because it cannot be influenced by humans.

However, a good correlation does not necessarily imply causality. Causal relationships could only be deduced from results of the exact natural sciences. Studies of the physico-chemical properties of CO_2, which would explain its high climate sensitivity, have so far been patchy; existing partial results tend to contradict this.

The Swedish chemist and physicist Svante Arrhenius was the first to suspect an effect of atmospheric CO_2 levels on global warming (Arrhenius 1896). He assumed that the ice ages were primarily controlled by CO_2 concentrations. No mention is made of measurements by the Swiss scientist N. de Saussure (De Saussure 1830), which he carried out in the city of Geneva and its surroundings before the beginning of the Industrial Revolution, from 1827 to 1829. The numerous measurements, day and night, at various wind speeds, before and after rainfall, resulted in atmospheric CO_2 levels of 400 to 500 ppm (Marten 2025). CO_2 levels have been measured automatically since 1960 at a measuring station on the Mauna Loa volcano of Hawaii; previous measurements were taken from ice cores (Grassl 1993). The accuracy of recent CO_2 measurements and global temperature reported by climate researchers is rightly criticized: CO_2 is distributed very differently regionally. Estimating a global temperature to within tenths of a degree is physically questionable given the enormous temperature differences that exist on planet Earth (Ullmann *et al.* 2024).

Atmosphere CO_2 concentrations have a long history. In the Earth's prehistoric era, which was characterized by intense volcanism, the atmosphere likely consisted of a high proportion of CO_2. At the time of the dinosaurs (approximately 200 to 100 million years ago), its concentration was still a few thousand vppm (Rae *et al*. 2021 & Hönisch *et al*. 2023). During the Pleistocene (ca. 1 million years ago), CO_2 levels were about 180 vppm, and during the interglacial periods, approximately 300 vppm. The difference between ice ages and interglacial periods is explained by outgassing from the oceans, which can occur with a delay of centuries.

2. Physico-chemical Properties of CO_2

2.1 Interaction of photons with gas molecules

According to quantum theory, during absorption and emission of a photon in a molecule, the entire energy of the quantum, $\varepsilon = h\nu$, is transferred or released to the molecule. The molecule receives an energy pulse of the same magnitude in the direction of the incident quantum upon absorption, and in the opposite direction upon emission (Einstein 1917). The fundamentals of photon absorption in atmospheric gas molecules describe which photon wavelengths resonate with which rotational and vibrational degrees of freedom of the molecules (Schrader 1995 & Hug 2000).

Heat is the kinetic energy of gas molecules. The molecules are set in motion by collisions with other gas molecules, by contact with the heated Earth's surface, or by radiation. The second law of thermodynamics determines the direction of heat flow from warmer to colder molecules. This results in a dispersion of energy, which leads to the *Gaussian* bell curve of temperature distribution among all molecules in the atmosphere. Heat from gas molecules is composed of translational, rotational, and vibrational energy, depending on the structure of the molecules:

$$E_{kin} = E_{trans} + E_{rot} + E_{swing}. \qquad (1)$$

Thermal energy corresponds to the energy of photons in the infrared, IR, range of electromagnetic radiation. Only greenhouse gases possess the ability to absorb radiation in the IR range, of which water vapor has the highest concentration, followed by CO_2.

2.2 Greenhouse gases as heat storage medium

The measure of the maximum absorbable heat energy of a gas is its specific or molar heat capacity, Cp. It has been recorded in tables for decades (Kortüm 1981 & WIKIBOOKS 2023). If one compares the Cp values of monatomic gases with those of diatomic and triatomic gases found in the atmosphere, the Cp values for monatomic gases - helium, neon, argon - and diatomic ones - N_2, O_2, NO, CO - are the same, regardless of their atomic or molecular weight (Table 1).

Table 1. Molar heat capacities of gases.

Type of gases	Cp (J/mol K)
(1) Monoatomic gases (He, Ne, Ar)	20,8 ± 0,2
(2) Diatomic gases (H_2, N_2, O_2, NO)	29,3 ± 0,5
(3) Threeatomic gases (H_2O, N_2O, CO_2)	35 ± 2

Diatomic gases have a heat capacity about 40% higher than monatomic gases. The heat capacities of triatomic gases such as H_2O, and trace gases CO_2, CH_4, N_2O, and SO_2 are approximately 20% higher than those of diatomic gases (Ullmann *et al.* 2023). The increase in heat capacity in this order is explained by the larger number of energy-absorbing degrees of freedom according to equation (1). Monatomic gases possess only translational degrees of freedom for absorbing energy. In the case of diatomic, doubly- or triple-bonded O_2 and N_2 molecules, rotational and oscillatory degrees of freedom are added to the translational degrees of freedom. However, their excitation requires higher-energy radiation outside the IR range. Polyatomic gases can absorb further energy and convert it into heat by exciting stretching and deformation vibrations with IR photons, which led to the designation ”greenhouse gases”.

The molar heat capacities of gases show that the majority of the energy in the atmosphere is translational energy. The heat in the “greenhouse Earth” is stored by all gas components according to their molar heat capacities multiplied with their concentrations
in the atmosphere. The heat is located primarily in its main components, nitrogen and oxygen. Viewing the CO_2 molecule as an energy storage medium cannot be justified based on the measured molar heat values. Only water vapor can make a significant contribution at the appropriate concentration and taking into account the phase transition enthalpies.

2.3 Greenhouse gases as converters between electromagnetic radiation and heat

The heat energy bound to gas molecules cannot be released into space due to the force of gravity to which the molecules are subject. This can only occur in the form of electromagnetic radiation (IR photons). For this to happen, the heat energy of the atmosphere must be converted through collisions into vibrations of the greenhouse gas molecules and ultimately into IR photons, as shown in equation (2):

$$E_{trans} \quad \leftrightarrow \quad E_{swing} \quad \leftrightarrow \quad E_{phot} \tag{2}$$

Water vapor and carbon dioxide, as well as other trace gases, would then act as transmitters and photon emitters, without which the process of energy exchange between the Earth's atmosphere and space would not be possible in both directions. Only in this function can trace gases have a significant effect, even in low concentrations (Hönisch *et* al.2023).

Whether this energy conversion can be fully reversible would depend on the residence time of the photon energy in the molecule. So far little is known about residence times.

Greenhouse gases absorb a portion of the IR radiation from the sun, thus generating the life-sustaining greenhouse effect in the Earth's atmosphere. The energy radiated in the IR range by the heated Earth's surface is also absorbed by greenhouse gases. The subsequent radiation is re-radiated in all spatial directions. The assumption is that the same radiation intensities are radiated both toward space and back to Earth, which should lead to a heat build-up near the Earth's surface. However, with increasing altitude, the density of the atmosphere decreases according to the barometric altitude formula, and thus also decreases the number of IR-absorbing CO_2 molecules. As a result, the “mean free path” of the IR quanta toward space would always be greater than toward the Earth's surface. With increasing altitude and decreasing temperature, water vapor also ceases to act as an IR absorber due to condensation. The exit for the IR radiation toward space thus remains open for Earth's radiation.

For the absorption of electromagnetic radiation of a certain wavelength by absorbing molecules, the following relationship applies:

$$log\ (I_o / I_i) = \varepsilon\ c\ d \qquad (3)$$

ε - absorption coefficient, c - molar concentration, d - layer thickness.

The intensity of irradiation I_0 to the intensity I_1 after that, the layer thickness d, and, thus, the absorbed energy, increases logarithmically with concentration. In the case of CO_2 in the atmosphere, this relationship must be considered in a differentiated manner: In the main absorption range, the 15-µm-band, saturation already occurs after a small layer thickness at high absorption coefficients. In the adjacent regions of shorter and longer wavelengths, the layer remains permeable at low energy absorption coefficients. Thus, even with saturation in one part of the spectrum, there is still permeability for other IR wavelengths with low energy input. However, the argument is used to question the continued use of fossil fuels.

3. Balances of CO_2 and Energy at the Earth's Surface

3.1 CO_2 emissions and exchange processes

Carbon dioxide is subject to exchange processes between the hydrosphere, the atmosphere, and the living and fossil biospheres. These exchange processes lead to a fractionation between C^{13}-CO_2 and C^{12}-CO_2, with the lighter is preferentially released from the oceans and also preferentially absorbed into the living biosphere. The gradation of the concentration ratio of these isotopes has been demonstrated; it is present in the least amount in fossil biomass (Koutsoyiannis 2024). Extensive studies (Resplandy *et al*. 2018, Friedlingstein 2019 & IPCC 2021) showed the difficulties of quantifying the contributions of individual sources and sinks. The amounts vary like crop yields from year to year. Estimates of annual CO2 emissions and exchange quantities of the year 2021 can be found in Table 2 (IPCC 2021).

The majority of anthropogenic annual CO_2 emissions come from energy production (1), still more than 80% based on fossil fuels, and land use (2), approximately 3%. The contribution of volcanoes (7) fluctuates widely and has likely been underestimated. Annual anthropogenic and volcanic CO_2 emissions are significantly smaller than the exchange processes between the atmosphere and the biosphere (3), (4) or the oceans (5), (6), but the differences between absorption and emission must be taken into account. Little is known about the time constants in this carbon cycle. Doubts about the accuracy of such estimates are justified, with the exception of the data for (1) and (9).

Table 2. Comparison of CO_2 exchange amounts.
(One ton of carbon corresponds to 3.67 tons of CO_2.)

Emission and exchange of CO_2	in t C/a
(1) Anthropogenic emissions	9,4 x 10^9
(2) Land use	1,6 x 10^9
(3) Atmosphere → Biospeäre	-142 x 10^9
(4) Biosphere → Atmosphere	137 x 10^9
(5) Atmosphere → Oceans	- 80 x 10^9
(6) Oceans → Atmosphere	78 x 10^9
(7) Emissions from volcanoes	> 0,1 x 10^9
CO_2-Inventories of the spheres	**in t C**
(8) Hydrosphere	37 000 x 10^9
(9) Atmosphere *	< 120 x 10^9

* at 480 Vppm CO_2 content, (Ullmann *et al*., this paper)

Oceans and inland waters, with their vast inventory of CO_2 and their more than 70% share of the Earth's surface, play an important role in the exchange with the atmosphere. Mixing processes of the air with the moving water surface of the oceans play a role in balancing the CO_2 concentration in the atmosphere and the ocean. The transport of CO_2 to deeper water layers or from them to the surface proceeds through global ocean currents, but also through the precipitation or dissolution of solid carbonates, all of which are time-consuming processes (Resplandy *et al.* 2018).

3.2 Radiative equilibrium and climate

Radiative equilibrium means that the magnitudes of energy flows from space to Earth and back are balanced. This is determined by comparing the average daily energy flows of solar radiation, geothermal energy, and human energy production (Table 3). Human energy flow is by four orders of magnitude lower than sunlight exposure. Human “energy production” is comparable to the small energy flow from the Earth's interior through radioactivity and reaction heat (Ullmann *et al.* 2025).

Table 3. Comparison of natural and anthropogenic energy flows.

Energy flows	Joule	Source
Daily sunlight exposure	$1,06 \times 10^{22}$	(IPCC2021, Trenberth 2009)
Anthropogen. energy production, daily	$1,6 \times 10^{18}$	(Statista 2024)
Daily heat flux from the Earth's interior	$3,8 \times 10^{18}$	(Pollack 1993)
Energy stored in the atmosphere	$1,26 \times 10^{24}$	(Ullmann 2025)

Radiation equilibrium is therefore essentially established by solar radiation and the reflection of the incoming solar radiation from the Earth's surface and the atmosphere. The balance between solar radiation and Earth's reflection is understood here as a so-called flow equilibrium, comparable to a lake that is temporarily filled by stronger inflow, but also temporarily empties more rapidly. An equilibrium within days or years would not be expected. There are too many periodic events (solar activity, pole shifts, ocean currents, etc.) or random events (volcanism, particle radiation, *etc*.) as well as delayed adjustments of the equilibrium (CO_2 dissolution in the ocean, CO_2 uptake/ release by flora and fauna, *etc*.) for this to happen. Climate changes over longer periods in limited regions, including those with significant stress factors for the ecosphere, are therefore to be expected. Due to the large number of influences, the climate is described as a chaotic process that cannot be predicted or extrapolated.

4. The Climate History of the Earth

Current discussions about the Earth's climate are limited to roughly the past 200 years and 100 years of the future. Attempts to create a climate model based on this period, valid until the end of our century, have failed. Looking back at the history of our planet's climate could provide new explanations. The analysis of ice cores from Antarctica (Petit *et al.* 1999) and Greenland (Dansgaard *et al.* 1992) yielded results going back 800 000 and 500 000 years, respectively. In this process, temperatures as well as CO_2 and CH_4 levels were recorded through gas analyses over the course of multiple ice ages and warm periods. A periodic alternation between warm and ice ages in cycles of approximately 100 000 years has been observed. One hypothesis attributes this to periodic changes in the gravitational relationships within the planetary system, with changes in the eccentricity of Earth's orbit and shifts in the tilt of Earth's axis, the so-called Milanković cycles (Petrovic *et al*. 2009). In addition, the Earth's climate is influenced by a number of geophysical and cosmic cycles (Bülow 2020).

Ice ages last significantly longer than warm periods. The rise from the temperature minimum of the ice age to the new warm period occurs within a few thousand years, whereas the transition to a new ice age happens much more slowly, over tens of thousands of years. Similar to the temperature-time patterns, CO_2 and CH_4 concentrations behaved in the same way. Peak values during warm periods reach up to 300 ppm, and minimum values around 190 ppm CO_2 during cold periods were analyzed (Lindzen *et al.* 2025). A more detailed analysis of the last 11 000 years, starting after the last ice age, reveals that around 8 000 years ago, the maximum temperature of the warm period had already been surpassed, and the slow decline toward a new ice age had begun. Peaks downward and upward of up to 2 °C for several decades occurred multiple times. The Little Ice Age of the Middle Ages in Europe is an example of this.

Some phenomena of this extraterrestrial regulatory mechanism of Earth's climate could be of interest for clarifying the mechanisms of climate change. The authors of (Petit *et al*. 1999) claim to have found that the CO_2 peaks lagged behind the temperature peaks by several thousand years. They did not provide an explanation for this. This would mean that the temperature of the ocean is the driving force, which, when increased, releases more CO_2 from the seawater. With this insight, the framework of current climate research would be turned upside down.

What is the mechanism for the slower transition from warm periods to low temperatures? It could be the significantly slower change in ocean water temperature due to its high heat capacity compared to that of the atmosphere, as well as the high energy transformations occurring in the water cycle. Added to this would be the incoming and reflected solar radiation with a changed *Albedo*. Changes in CO_2 concentrations in the range of 100 ppm could rather be seen as a secondary effect and not as a driving force.

Conclusions

The observed relationship between CO_2 levels and global temperature rise in the Earth's atmosphere since the beginning of the Industrial Revolution cannot be confirmed as a causal relationship by physical and chemical results. Water vapor has the highest concentration among the IR-absorbing greenhouse gases. However, the climate at present focuses on man-made CO_2.

The heat of gas molecules is composed - depending on the structure of the molecules - of translational, rotational, and vibrational energy. The heat in the “Greenhouse Earth” is stored by all gas components according to their molar heat capacities and concentrations in the atmosphere, thus being located primarily in its main components, nitrogen and oxygen. Viewing the CO_2 molecule as an energy storage medium cannot be justified by the molar heat values. Only water vapor can make a significant contribution at appropriate concentrations and taking into account the transformation enthalpies of water.

Greenhouse gases are considered as transmitters and transducers between translational, rotational, and vibrational energy on the one hand, and photon energy on the other, without which the process of energy exchange between the Earth's atmosphere and Space cannot proceed in both directions.

The CO_2 exchange between the atmosphere and the plant world, as well as the oceans, is estimated to be several times greater than annual anthropogenic emissions. The oceans, with their vast inventory of CO_2, play an important role in this exchange. The ocean has so far been predominantly assumed to be a CO_2 sink; however, it should also be a CO_2 source with rising temperatures.

Human “energy production” is four orders of magnitude smaller than solar radiation and comparable to the small energy flow from the Earth's interior. The Earth's “greenhouse” atmosphere is an open system; the existing flow equilibrium establishes the climate changes.

The history of the Earth's climate should be included in the climate discussion. A planetary regulatory system with ice ages and warm periods occurring in cycles of a hun dred thousand years overrides short-term climate forecasts, so the next ice age is certain to come.

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Acknowledgments: Helmut Ullmann and Martin Bülow would like to thank their wives Erika Ullmann and Heidrun M. Bülow, respectively, for their patience and constant encouragement while working on the manuscript in difficult times.

Research ethics: Not applicable.

Informed consent: Not applicable

Author contributions: Both authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Helmut Ullmann and Martin Bülow: Developing ideas, writing and interpreting data as well as reviewing the manuscript.

Competing interests: The authors state no conflict of interest.

Research funding: None declared.

Use of Large Language Models, AI and Machine Learning Tools: None declared

Data availability: Not applicable

The Authors

Helmut Ullmann, born in 1936, Professor Dr. sc. nat., Dipl.-Ing. in radiochemistry. Until 1991, at the Central Institute for Nuclear Research in Rossendorf, Germany, high-temperature chemistry of nuclear reactors. 1993 – 2009: project leader at the Technical University of Dresden, focusing on ion transport in oxides. Thermodynamics of gas-solid reactions. Materials for high-temperature fuel cells.

Martin Bülow, born in 1939, Professor Dr. rer. nat. habil.; Dipl.-Chem. in physical chemistry; 1965 - 1992, at the Central Institute for Physical Chemistry, Berlin, of the Academy of Sciences of the GDR; 1985 - 1990 Deputy Director there; 1992 - 2005, Schuftan Fellow at the BOC Group Technical Center, Murray Hill, USA; development of adsorbents for separation and purification of gas mixtures, and clarification of underlying microprocesses.