Climatic consequences of the process of saturation of radiation absorption in gases

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ABSTRACT

This article provides a brief review of research on the impact of anthropogenic increase in atmospheric CO_2 concentration on Earth's climate. A simplified analysis of resonant radiation absorption in gases is conducted. Building upon the material from the cited articles, theoretical and empirical relationships between radiation absorption and the mass of the absorbing material are presented. The concept of saturation mass is introduced. Special attention is given to the phenomenon of thermal radiation absorption saturation in carbon dioxide. By comparing the saturation mass of CO_2 with the quantity of this gas in Earth's atmosphere, and analyzing the results of experiments and measurements, the need for continued and improved experimental work is suggested to ascertain whether additionally emitted carbon dioxide into the atmosphere is indeed a greenhouse gas.

Significance statement: • The impact of anthropogenic increase in atmospheric CO2 concentration on Earth's climate is analysed. • The concept of saturation mass is introduced. • By comparing the saturation mass of CO2 with the quantity of this gas in Earth's atmosphere, and analyzing the results of experiments and measurements, the need for continued and improved experimental work is suggested to ascertain whether additionally emitted carbon dioxide into the atmosphere is indeed a greenhouse gas.

1. Introduction

Due to the overlap of the absorption spectra of certain atmospheric gases and vapours with a portion of the thermal radiation spectrum from the Earth's surface, these gases absorb the mentioned radiation. This leads to an increase in their temperature and the re-emission of radiation in all directions, including towards the Earth. As a result, with an increase in the concentration of the radiation-absorbing gas, the temperature of the Earth's surface rises. Due to the observed continuous increase in the average temperature of the Earth and the simultaneous increase in the concentration of carbon dioxide in the atmosphere, it has been recognized that the increase in atmospheric carbon dioxide concentration associated with human activity may be the cause of climate warming.

This phenomenon was already noted by Arrhenius (1896). The United Nations, concerned about climate change, established the Intergovernmental Panel on Climate Change (IPCC) to provide objective scientific information about climate change. The IPCC prepares comprehensive Assessment Reports on climate change knowledge, its causes, potential impacts, and response options. IPCC reports are assessments of published literature and are prepared by hundreds of experts from various fields. In addition, since 1995, United Nations Climate Change Conferences (known as Conference of the Parties - COP) have been held to negotiate actions related to climate policy. Authors of various books and publications refer to the IPCC reports. They often agree on the credibility of the forecasts made by the IPCC (Anderson et al., 2016; Ramanathan 1988; Karl and Trenberth 2003; Hansen et al., 1981; Kellogg 1987). These forecasts provide compelling arguments that in response to the ongoing emissions of CO_2 and other greenhouse gases into the atmosphere, the global climate will continue to undergo significant warming. However, individual studies highlight various details often not included in IPCC reports. For example, studies Madden and Ramanathan (1980) demonstrate the possibility of climate warming delays compared to the projections based on mathematical models, caused, among other factors, by the thermal inertia of the oceans. Furthermore, the need for increased research to better understand climate processes is emphasized by the author of (Jain 1993). However, in most published works, the negative impact of anthropogenic increases in atmospheric CO_2 concentration on the Earth's climate is considered established, and much attention is given to the consequences of climate

change. In addition to glacial melting and rising sea levels, local issues are often highlighted. For instance, in Woolway et al. (2020), it is stated that climate change is one of the most serious threats to global lake ecosystems.

Sometimes, in addition to reducing CO_2 emissions, capturing this gas from the atmosphere is proposed (Breyer et al., 2019). Methods that aim to compensate for the greenhouse effect by increasing albedo are also proposed (Goldblatt et al., 2013; Akbari et al., 2009).

However, there are studies that recognize the need for more substantial supplements or changes to the computer models adopted by the IPCC. This includes emphasizing the role of clouds in the adopted models (Mitchell 1989; Abbood and Al-Taai 2018; Alados-Arboledas et al., 1995; Sakurai et al., 2005). In some studies, such as Trenberth and Fasull (2009), it is noted that the adopted models do not account for the fact that the greenhouse effect caused by increasing greenhouse gases and water vapor (as a feedback) is balanced by a decrease in cloud cover and, therefore, an increase in radiation emissions. The need for a more serious consideration of aerosols in the conducted research is also emphasized. For example, study (Landsberg 1970) states that aerosols produced by humans, due to their optical properties and potential influence on cloud and precipitation formation processes, pose a more significant problem than CO_2. Additionally, Wild (2016) shows that subtle changes in aerosols over large ocean areas, amplified by aerosol-cloud interactions, can significantly alter incoming solar radiation and, consequently, change sea surface temperatures. The author of Palmer (1999) pays significant attention to the accuracy of descriptions, stating, among other things, that errors in simulating local air-sea heat fluxes can exceed the direct effect of doubling CO_2. Reservations about the adopted models are also presented by the author of (Shine and de F Forster, 1999). The study shows that the research attributing observed climate changes to human activity only considered a part of all the mechanisms driving these changes, and therefore, the conclusions can be unquestionable only when all these mechanisms are taken into account.

The author of studies Stallinga (2018) and Stallinga (2020) Refs. to ice core drilling, which is often presented as evidence of the influence of CO_2 concentration on Earth's temperature. Based on the strong correlation observed between the concentration of CO_2 trapped in air bubbles in ice and temperature (measured through oxygen isotope ratios found in similar ice core drillings), the author demonstrates the relationship between CO_2 concentration and air temperature. However, as shown, this relationship cannot be explained by the greenhouse effect as the results differ practically by orders of magnitude. On the other hand, Henry's law concerning the release of CO_2 from the oceans explains these phenomena very well.

The use of current mathematical models in predicting climate change is strongly questioned by the author of Idso (1998). Based on the analysis of experiments conducted, it is stated that the complexity of the Earth's climate system makes accurate prediction of global climate changes very difficult for general circulation models and is likely the cause of the deviations observed. Therefore, it is believed that the currently used models do not yet provide a suitable basis for the development of rational policies related to potential climate changes.

The presented literature review shows that there is still much to be done in the field of climate change, especially in experimental studies that can decisively resolve many disputed issues.

2. Simplified description of resonant radiation absorption in gases

As noted in the introduction, the basis for considering climate change sources is the absorption of radiation in gases. To better understand this phenomenon, we will use a simplified model described in Kubicki et al. (2022) that explains this process. In this model, the propagation of radiation in an absorbing gas, neglecting scattering processes, can be described by the Schwarzschild equation. Assuming that the optical

thickness is proportional to the mass *m* of the absorbing substance per unit area perpendicular to the direction of radiation propagation, the equation takes the form:

$$\frac{dI_\lambda}{dm} = -\alpha \cdot I_\lambda + E \tag{1}$$

where I_λ – radiation intensity at a specific wavelength λ, $E = \alpha \cdot B_\lambda(T)$, α – absorption coefficient, $B_\lambda(T)$ – Kirchhoff-Planck function associated with the emission of radiation by a gas.

It should be noted that the Kirchhoff-Planck function introduces a strong dependence of radiation intensity on the temperature of the gas, and in the case of the Earth's atmosphere, it is influenced by numerous factors not directly related to radiation.

For monochromatic radiation, in the absence of additional sources of heat, the gas absorbs radiation within a narrow frequency range, while the emission of radiation from the gas occurs at all possible transitions. Therefore, only a very small, negligible portion of the power is allocated to the frequency band of the incident beam. As a result, the Schwarzschild equation transforms into the equation describing Lambert-Beer's law.

$$\frac{dI_\lambda}{dm} = -\alpha \cdot I_\lambda \tag{2}$$

Its solution for radiation of a specific wavelength λ is, of course, the exponential function:

$$I = I_0 e^{-\alpha \cdot m} \tag{3}$$

where I_0 – is the initial intensity of the radiation.

Absorption of radiation, defined as the ratio of absorbed radiation energy to the incident radiation energy within a specified time, takes the form:

$$A = \frac{I_0 - I}{I_0} = 1 - e^{-\alpha \cdot m} \tag{4}$$

For a sufficiently large value of *m*, which can be achieved by increasing the concentration of the absorbing substance, the intensity of radiation becomes negligibly small. Further increasing the concentration of the gas does not contribute anything because the radiation under consideration is practically not being examined. This condition is referred to as the saturation of the absorbing substance.

In the case of continuous radiation, the qualitative depiction of the occurring processes can be illustrated using Fig. 1 from reference (Kubicki et al., 2020b).

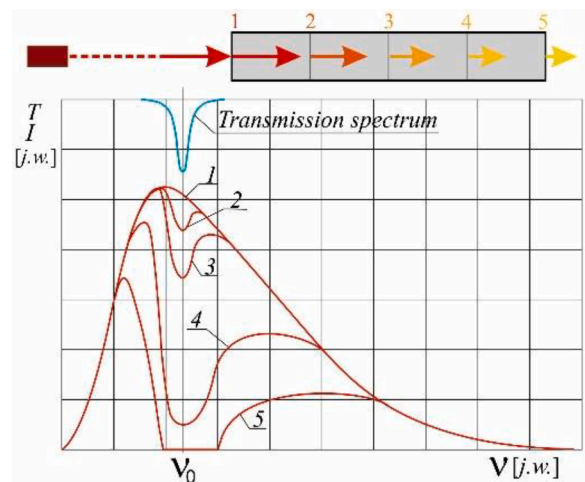After passing through successive layers of the medium, the intensity

Fig. 1. Distortion of the radiation spectrum passing through an absorbing medium (Kubicki et al., 2020b).

of radiation at the frequency ν_o decreases relatively quickly, while at other frequencies, the changes are slower or nonexistent. As a result, the radiation spectrum undergoes changes, taking on the form of 2, 3, 4, 5. Ultimately, in the spectrum of transmitted radiation, a specific funnel-shaped region is formed that coincides with the absorption spectrum. Radiation with such a spectrum cannot be absorbed because its spectrum does not overlap with the absorption spectrum. However, in the case where the absorption spectrum has a linear structure, as is the case with CO_2, an additional effect occurs involving the increasing role of "wings" in the oscillatory-rotational lines. As the central part saturates, the line broadens, leading to further increase in radiation absorption (Fig. 2) (Kubicki et al., 2020a).

Therefore, we are dealing with two opposing phenomena, each with different effects. Additionally, the frequency spacing between the lines is limited, and for sufficiently large absorption, the lines will start to overlap. Therefore, in the considerations presented in Kubicki et al. (2022), a simplified model was adopted in which the broadening effect of the oscillatory-rotational lines was neglected, and it was assumed that the radiation intensity is sufficiently small to disregard the increase in gas temperature associated with its absorption. In this case, the solution to Eq. (1) for this scenario takes the form:

$$I = \left(I_0 - \frac{E}{\alpha}\right)e^{-am} + \frac{E}{\alpha} \tag{5}$$

Substituting these values into the absorption equation and introducing a notation $\psi = 1 - \frac{E}{al_0}$, we have:

$$A = \frac{I_0 - I}{I_0} = \psi(1 - e^{-am}) \tag{6}$$

It should be noted that by neglecting the increase in gas temperature associated with radiation absorption, an overestimated value of radiation absorption in the gas is obtained.

From the conducted considerations, it follows that both in Eq. (4) and Eq. (6), the value of absorption is limited. In the first case, it cannot exceed 1, and in the second case, it cannot exceed the value of ψ less than unity. Therefore, for a sufficiently large mass m, saturation must occur, and further increase in mass will result in a negligible increase in absorption. This conclusion seems trivial since, regardless of the simplifications made, the absorbed radiation intensity cannot be greater than the incident radiation intensity, and thus the absorption can never exceed a value of 1.

The phenomenon of saturation was already noted by Ångström (1900), who, based on experiments and analysis, challenged Svante Arrhenius' hypothesis that continued use of fossil fuels would warm the planet (Arrhenius 1896). In 1972, Schack (1972), based on his considerations, demonstrated that for a concentration of 0.03% of carbon dioxide in the air, the absorption process in the troposphere is saturated.
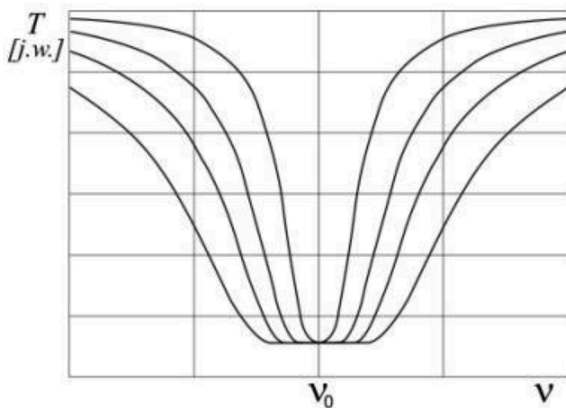
Taking into account the saturation process, Dieter Schildknecht also proved in his work (Schildknecht 2020) that, contrary to the IPCC reports, the impact of anthropogenic CO_2 increase on the Earth's climate is very small.

It is worth noting that there is relatively little empirical research in the available contemporary literature, although in the frequently cited work (Goody and Yung, 1989), the authors emphasize the need for experimental determination of coefficients in the presented equations for radiation absorption in gas.

Therefore, in this article, an attempt was made to emphasize the importance of empirical studies on the saturation process of radiation absorption in gas, and more space was devoted to them. In the described experimental works, efforts were made to answer the fundamental question: what is the saturation mass of the absorbing gas, i.e., the mass above which further increase in absorption can be considered "negligible"? It is necessary to provide a more precise definition of the term "negligible," and therefore, the definition was adopted in which the saturation mass, m_s, is defined as the mass of the dissolved absorbing substance in the gas per unit area perpendicular to the direction of radiation propagation, for which absorption reaches 95% of the maximum value that absorption asymptotically approaches with an increase in this mass.

In the case of a gas at a specific pressure and temperature placed in a cell with windows, this mass can be determined, among other methods, using the experiments presented below.

3. Examples of experimental determination of saturation mass

One example of monochromatic radiation absorption in a gas is the absorption of 3.39μm radiation emitted by a He-Ne laser in methane. Fig. 3 illustrates the overlap of a portion of methane's transmission spectrum with the spectrum of this radiation.

To determine the saturation mass of methane for this radiation, the experimental setup shown in Fig. 4 was used, which allows for the elimination of adverse effects of slow instabilities of the gas laser on the measurement results.

The split light beam, interrupted by a rotating disk with a single aperture, alternately passed through the test cuvette into which specific portions of methane were injected, and the reference cuvette filled with air. As a result, signals depicted in Fig. 5 were obtained on the oscilloscope.

Based on these signals, the absorption for the i th methane concentration was determined using the formula:

$$A_i = 1 - \frac{b_i}{\eta \cdot a_i} \tag{7}$$

where $\eta = \frac{b_0}{a_0}$, and ao, bo – values of the signals for zero methane

Fig. 2. Deformation of the absorption line due to saturation (Kubicki et al., 2020a).
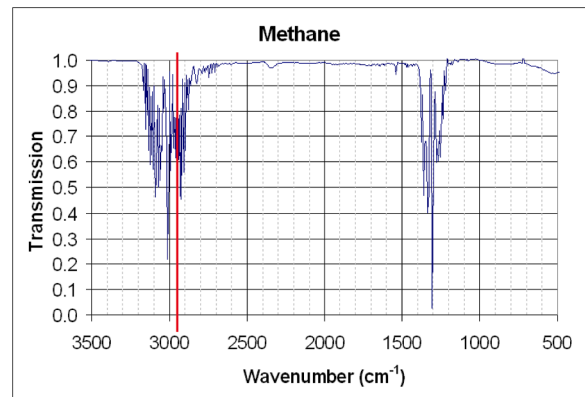

Fig. 3. Methane transmission spectrum (blue line) (HITRAN data base) overlaid with the spectrum of a 3.39μm He-Ne laser (red line).
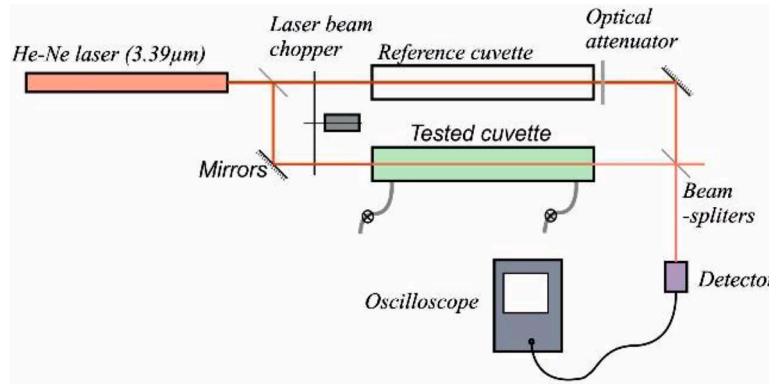
Fig. 4. Experimental setup diagram for determining the saturation mass of methane absorbing monochromatic radiation with a wavelength of 3.39μm.
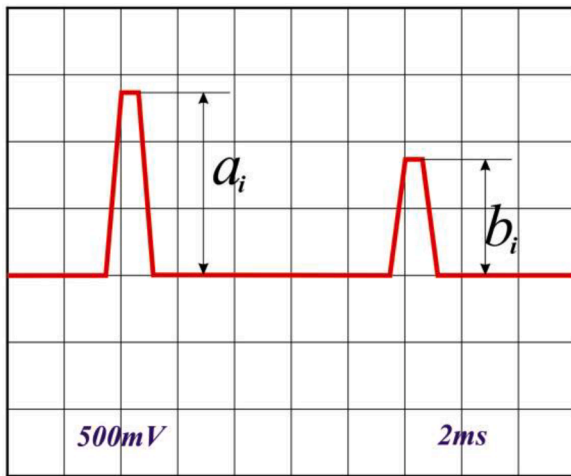

Fig. 5. Signals on the oscilloscope: a_i– for the reference cuvette; b_i – for the test cuvette.

concentration.

The conducted measurements allowed for the construction of the dependency graph of the absorption of the utilized radiation on the mass of injected methane, as presented in Fig. 6.

Based on this graph, the saturation mass of methane for radiation of 3.39μm was determined. This mass was approximately ~ 2,7·10–3kg/m².

As an example of continuous radiation absorption, the absorption of thermal radiation in carbon dioxide, as described in Kubicki et al. (2022), was chosen. The schematic diagram of the experiment described
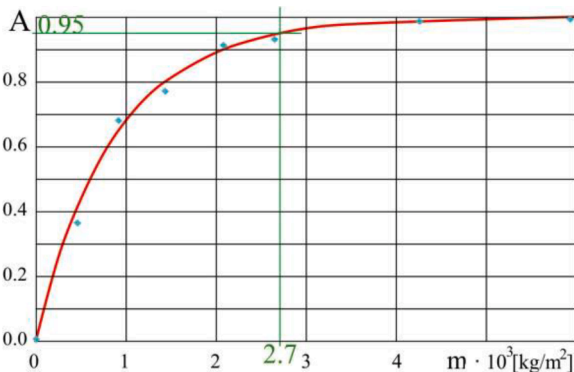
in the paper is presented in Fig. 7.

It utilized a thermal radiation source described in (Kubicki et al., 2020a), which consisted of a glass vessel filled with heated mineral oil to a specific temperature. A graphite layer was applied on one flat side of this vessel, emitting the thermal radiation used. The absorption cell used in Kubicki et al. (2022) was constructed as a horizontal PVC pipe with a length of 1m and a diameter of 150mm, sealed with polyethylene film windows. By varying the carbon dioxide concentration inside the absorption cell, the absorption of thermal radiation was measured at oil temperatures of 78.6°C and 109.5°C. The temperatures were chosen randomly but in a manner that allowed reliable measurement of radiation intensity and demonstrated the influence of temperature on the saturation mass value. Based on the conducted measurements, absorption characteristics of thermal radiation as a function of injected carbon dioxide mass were obtained for these temperatures (Fig. 8).

The determined saturation mass ms based on the plotted graph is 0.57kg/m² for a temperature of 78.6°C, and 0.66kg/m² for a temperature of 109.5°C. It should be noted that in the Earth's atmosphere, for the currently assumed concentration of CO_2 - 400ppm, the amount of carbon dioxide per 1 m² of horizontal surface is mz > 6kg/m². Extending the horizontal axis of the graph from Fig. 7 to this value, we obtain the image shown in Fig. 9, which suggests that there is currently a multiple exceedance of the saturation mass for carbon dioxide in the Earth's atmosphere.

However, it should be noted that unlike the used cuvette, the vertical structure of the atmosphere undergoes changes in both pressure and temperature. Nevertheless, the question arises as to whether the additionally emitted carbon dioxide into the atmosphere will absorb thermal radiation.
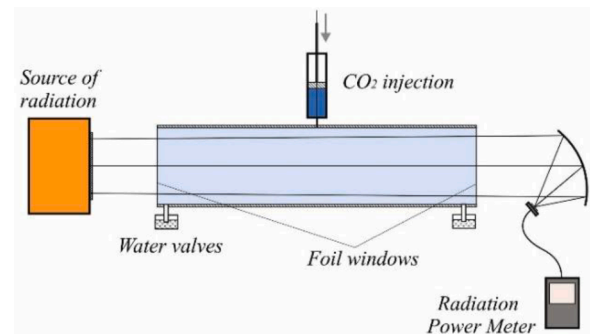

Fig. 6. Graph showing the dependence of absorption of transmitted radiation on the mass of injected methane.

Fig. 7. Diagram of the laboratory setup for measuring the absorption of thermal radiation in carbon dioxide (Kubicki et al., 2022).
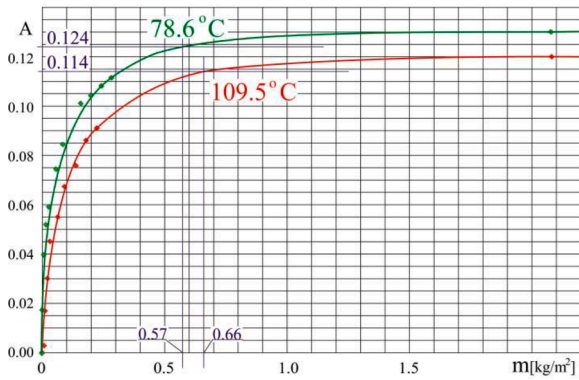
Fig. 8. The graph depicting the relationship between the absorption of transmitted thermal radiation and the injected mass of CO_2 (Kubicki et al., 2022).

4. Phenomena that confirm the hypothesis of the saturation process of thermal radiation absorption in the earth's atmosphere

4.1. Absorption of thermal radiation from the moon by carbon dioxide

It is known that the Sunlit surface of the Moon has a temperature of approximately 110°C and therefore must emit thermal radiation (aided by scattered infrared radiation from the Sun). When this radiation reaches the Earth's surface, it has to pass through the entire atmosphere, interacting, among other things, with the carbon dioxide dissolved in it. As a result, there should be a described dip in the spectrum of this radiation, reducing its overlap with the absorption spectrum of CO_2, and consequently, the absorption of this radiation by additional carbon dioxide should decrease. To test this hypothesis, an experiment described in (Kubicki et al., 2020b) and (Kubicki et al., 2020b) was conducted to

determine this mentioned absorption. The experiment consisted of two parts. In the first part, conducted in the laboratory, a lunar simulator was used in the form of a body heated to a temperature of 110°C. The thermal radiation emitted by this simulator was alternately passed through a cuvette containing CO_2 and a reference cuvette filled with air according to the scheme shown in Fig. 10. This allowed for the measurement of radiation absorption in the CO_2 cuvette.

The conducted measurements showed that the absorption of radiation in carbon dioxide in the cuvette was approximately 14%. In the second part of the experiment, shown in Fig. 11, radiation from the Moon was used.

This time, the radiation used, before passing through the inserted cuvettes, first passed through the Earth's atmosphere. It turned out that the absorption of this radiation in carbon dioxide in the cuvette (the same cuvette as in the first part of the experiment) was practically negligible. It can be clearly concluded that additional carbon dioxide does not absorb thermal radiation that has been emitted from the heated surface of the Moon and has passed through the Earth's atmosphere. This raises the question of whether, in the case of thermal radiation from the Earth's surface, passing through the atmosphere in the opposite direction, a saturation process will also occur and whether this radiation will be absorbed by carbon dioxide in the cuvette.

4.2. Comparison of periodic changes in the global concentration of CO_2 in the atmosphere with periodic changes in global temperature

In the study (Humlum et al., 2013), the authors demonstrated that peaks of cyclic changes in air and water temperature globally precede peaks of cyclic changes in atmospheric CO_2 concentration (Fig. 12). This finding supports the hypothesis that, as a result of saturation processes, emitted CO_2 does not directly cause an increase in global temperature. Instead, it suggests that an increase in temperature likely leads to the release of carbon dioxide from the oceans.
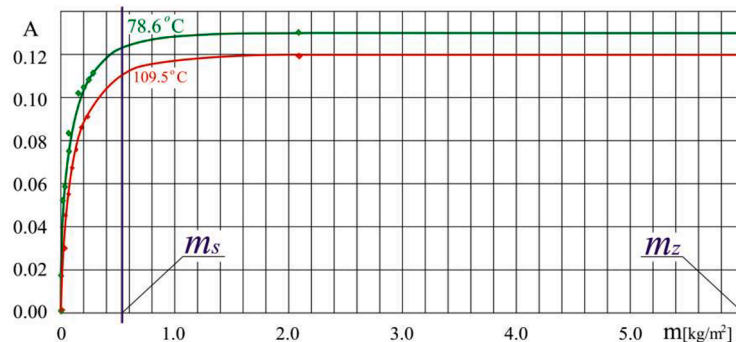

Fig. 9. Extended graph showing the relationship between absorption of transmitted thermal radiation and carbon dioxide mass (Kubicki et al., 2022).
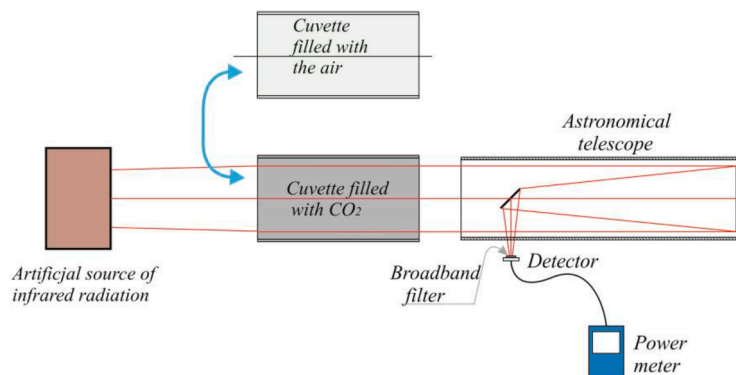

Fig. 10. Experimental setup for measurement of radiation absorption in the CO2 cuvette (Kubicki et al., 2020b).
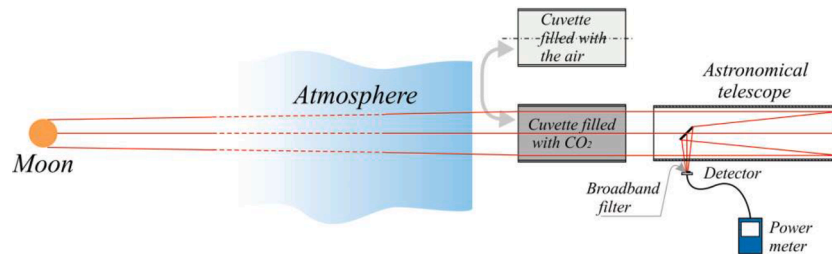
Fig. 11. Experimental setup for measurement of transmission of infrared radiation from the moon by carbon dioxide (Kubicki et al., 2020b).
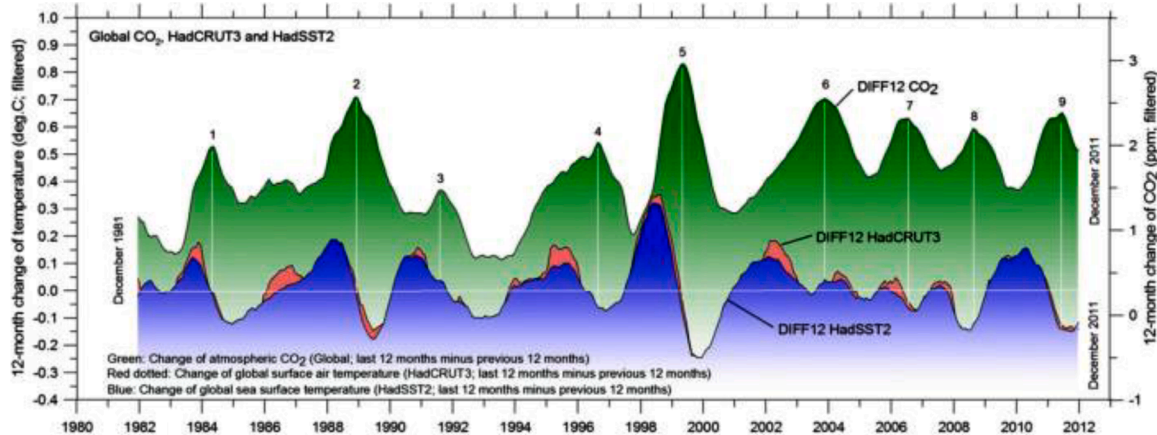

Fig. 12. Periodic change in global atmospheric CO2 concentration (green), global sea surface temperature (blue), and global surface air temperature (red) (Humlum et al., 2013).

5. Conclusions

The presented material shows that despite the fact that the majority of publications attempt to depict a catastrophic future for our planet due to the anthropogenic increase in CO_2 and its impact on Earth's climate, the shown facts raise serious doubts about this influence. Without delving into the accuracy of the utilized models, we should closely examine the possibilities of gathering reliable input data for these models. These data are directly related to the distribution of temperature on Earth's surface and in the atmosphere, the distribution of water vapor concentration in the atmosphere, the distribution of wind speed and direction, and the distribution of aerosols and particles in the atmosphere (clouds, aerosols above fluctuating oceans). It is obvious that simultaneous measurements of these variables across the entire globe are not feasible, and averaging them in situations where strong nonlinear dependencies exist can lead to significant errors. Moreover, the atmosphere exhibits high dynamics, which further complicates such measurements. Therefore, it is not surprising that the results in various significant works such as Schildknecht (2020) and Harde (2013), differ greatly from those presented by the IPCC, which is widely regarded as the sole reliable authority. This unequivocally suggests that the officially presented impact of anthropogenic CO_2 increase on Earth's climate is merely a hypothesis rather than a substantiated fact. Resolving these dilemmas requires further experimental work to verify the results of theoretical studies at every possible stage. To answer the question of whether the additionally emitted CO_2 in the atmosphere is indeed a greenhouse gas, it would be necessary, among other things, to conduct additional research for a radiation source with a temperature similar to Earth's surface temperature and measure the absorption of thermal radiation in a mixture of CO_2 and air at different temperatures and pressures, as is the case in Earth's atmosphere at various altitudes. It would also be beneficial to conduct field studies using an appropriate balloon, as suggested in (Kubicki et al., 2020b). By measuring the absorption of Earth's thermal radiation in atmospheric CO_2 under atmospheric pressure in a cuvette placed in the basket of a balloon in the upper layers of the troposphere, we could obtain results that would decisively settle many controversial issues. For example, if it turned out, just like in the case of thermal radiation from the Moon, that there is no noticeable absorption of Earth's thermal radiation in CO_2, it would mean that the spectrum of radiation emitted into space, as presented in the illustrative Fig. 1, exhibits a "funnel" created as a result of absorption in gases and water vapor in the atmosphere. It should be noted that CO_2 absorption lines at different altitudes are narrower than CO_2 absorption lines under atmospheric pressure, and thus, it could be authoritatively stated that we are dealing with atmospheric saturation, and the additional CO_2 emitted into the atmosphere, regardless of its altitude, will not be a greenhouse gas.

However, the intention of the authors of this article is not to encourage anyone to degrade the natural environment. Coal and petroleum are valuable chemical resources, and due to their finite reserves, they should be utilized sparingly to ensure they last for future generations. Furthermore, intensive coal mining directly contributes to environmental degradation (land drainage, landscape alteration, tectonic movements). It should also be considered that frequently used outdated heating systems burning coal and outdated internal combustion engines fueled by petroleum products emit many toxic substances (which have nothing to do with CO_2). Therefore, it seems that efforts towards renewable energy sources should be intensified, but unsubstantiated arguments, especially those that hinder economic development, should not be used for this purpose.

In science, especially in the natural sciences, we should strive to present a true picture of reality, primarily through empirical knowledge.

CRediT authorship contribution statement

Jan Kubicki: Conceptualization, Data curation, Formal analysis,

Investigation, Methodology, Validation, Visualization, Writing – original draft. **Krzysztof Kopczyński:** Supervision, Validation, Writing – review & editing. **Jarosław Młyńczak:** Formal analysis, Validation, Visualization, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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