

SATURATION OF THE ABSORPTION OF THERMAL RADIATION BY ATMOSPHERIC CARBON DIOXIDE

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Abstract. The article presents a concise review of the works concerning the impact of an increase of the concentration of carbon dioxide in the atmosphere on the increase of its absorption of thermal radiation. Attention was paid to differences in the results of calculations presented in the works by various authors. Experimental verification was carried out, which confirmed the possibility of saturation of the process of thermal radiation absorption by CO₂ in the atmosphere. Possibilities of improving climate models by using direct measurement results of experimental works were pointed out.

Keywords: greenhouse gases, radiation processes, Schwarzschild equation, climate sensitivity

NASYCENIE PROCESU ABSORPCJI PROMIENIOWANIA TERMICZNEGO W ATMOSFERYCZNYM DWUTLENKU WĘGLA

Streszczenie. W artykule przedstawiono zwięzły przegląd prac dotyczących wpływu wzrostu stężenia dwutlenku węgla w atmosferze na wzrost w niej absorpcji promieniowania termicznego. Zwrócono uwagę na różnice wyników obliczeń w pracach różnych autorów. Przeprowadzono weryfikację eksperymentalną, która potwierdziła możliwość nasycenia się procesu absorpcji promieniowania termicznego dla CO₂ w atmosferze. Wskazano możliwości doskonalenia modeli klimatycznych poprzez wykorzystywanie bezpośrednich wyników pomiarów w pracach eksperymentalnych.

Słowa kluczowe: gazy cieplarniane, procesy radiacyjne, równanie Schwarzschilda, wrażliwość klimatyczna

Introduction

Radiation processes occurring in the atmosphere, on the Earth's surface and in the surface layer of the ocean play a key role in the energy balance of the climate system [10]. Some of the solar radiation that illuminates the Earth is scattered and reflected by clouds and aerosols or absorbed by the atmosphere. The remaining radiation is absorbed or reflected by the Earth's surface. The energy of solar radiation is transformed into heat, latent energy (including various water states), potential energy and kinetic energy, and then it is emitted as long-wave radiation energy into space [13].

Finally, a state of equilibrium is established for which the average radiation flux from the Sun must be equal to the average radiation flux from the Earth.

The heated lithosphere is a gray body whose emission properties are similar to those of a black body. The Planck formula describing them clearly shows that the flux of the emitted radiation is an increasing function of the body temperature emitting this radiation. Thus, for a constant value of the radiation flux from the Sun, any reduction of radiation emission into space must cause an increase of the Earth's temperature (lithosphere). This is caused primarily by the processes of interaction of the lithosphere thermal radiation with active gases in the atmosphere. Thus, these gases, called greenhouse gases, have an uncontested effect on the Earth's climate and the related phenomena have been described, among others, in book [1] and in many other works on this subject [5, 6, 8, 9, 13].

Often, a special parameter is used to describe the effect of the greenhouse gas on the increase of the average temperature on the Earth's surface. This parameter, called climate sensitivity, determines the increase of the average temperature of the Earth when the amount of a greenhouse gas is doubled.

Concern for the Earth's climate inspires further work on this topic. In 1988, at the UN, upon request of the World Meteorological Organization (WMO) according to the United Nations Environment Program (UNEP), an intergovernmental scientific advisory body The Intergovernmental Panel on Climate Change (IPCC) was set up. Its purpose is to provide objective, scientific information on climate change.

However, despite the involvement in the climate change of many outstanding research centres equipped with the best apparatus, there is a lack of experimental work verifying the results of theoretical calculations. The necessity to undertake such work is justified by the fact that the described phenomena are relatively complex while some data taken for calculations may raise objections (due to the diversity of the analysed areas and the

dynamics of the processes). As a result the final calculations may be very different.

In this work, taking into account the presented literature, an attempt to remind the processes of thermal radiation interaction with the active gases was made. Then, the possibilities of verification and supplementing existing knowledge with the help of conducted experiments and proposals of further research were shown.

1. Phenomenon of thermal radiation absorption in the atmosphere

Propagation of long-wave radiation with the intensity I and wavelength λ , emitted by a black body and passing through absorbing gaseous medium, neglecting scattering, can be described by the Schwarzschild equation in the form [10]:

$$\frac{dI_\lambda}{d\tau} = -I_\lambda + B_\lambda(T) \quad (1)$$

where: $d\tau = k_\lambda \rho ds$, τ – optical thickness measured rectilinearly (with neglecting refraction in the atmosphere), k_λ – mass absorption coefficient, ρ – density of the absorbing medium, s – propagation path, $B_\lambda(T)$ – Kirchhoff-Planck function.

When the temperature of the gas medium is much lower than the temperature of the radiator, the function $B_\lambda(T)$ can be omitted and equation (1) will take the differential form of Lambert-Beer law. In the case of thermal radiation emission from the lithosphere (decreasing its temperature), the above situation occurs in the higher layers of the troposphere, where the temperature is much lower than on the Earth's surface. If the emitted radiation interacting with active gases in the atmosphere was monochromatic, its intensity during propagation in the atmosphere would decrease exponentially. Then at sufficiently high value of τ , increasing e.g. due to the increase of the concentration of the gases, the value of this intensity would become negligible. It could be said, then, that the saturation of the concentration of the absorbing substance occurred. Further increase of the concentration of the gases would not bring any effect, because there would be no radiation. However, in reality the situation is more complex. The radiation is not monochromatic because it is emitted by the lithosphere that is a grey body and the spectrum of this radiation is continuous similar to the spectrum of the black body described by the Planck formula

The carbon dioxide present in the atmosphere has a linear spectrum and with increase of optical thickness, these lines are deformed (Fig. 1). First, the saturation effect occurs at the central frequency ν_0 , and then at farther frequencies.

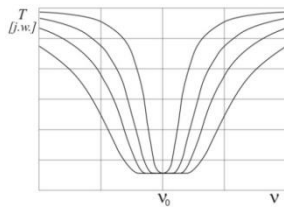


Fig. 1. Transmission line deformed by saturation

The final effect of such a process can be shown on the basis of the image of the transmission spectrum of layers of air of different thickness and with CO₂ concentration of 380 ppm (Fig. 2) [7].

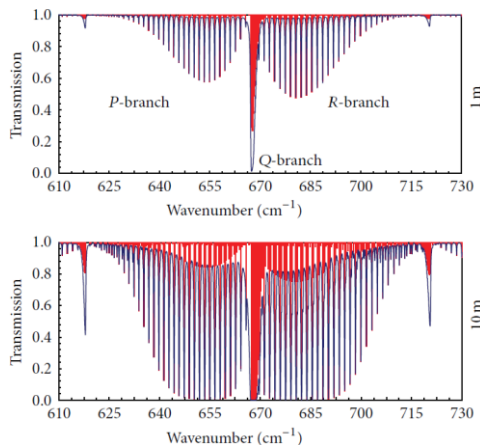


Fig. 2. Transmission spectra of dry air with CO₂ concentration of 380 ppm for two different thicknesses (air pressure: 1013 hPa, temperature: 288 K; red colour show transmission for central frequencies of the absorbing lines, while blue colour show total transmission for these lines) [7]

The phenomenon is quite complex and therefore in theoretical considerations various approximations are used to explain and estimate the effects of the happening processes. For example, in the work [5], it was shown that the absorption of the entire extended Lorentzian line (if there is a lot of absorbing substance) is equal to:

$$A = C + D \ln(m) + E \ln(p) \quad (2)$$

where: C , D i E – constants, m – amount of the absorbing substance, p – pressure.

This formula shows that when the concentration of the absorbing gas increases the absorption also increases. However, it should be noted that this is a logarithmic increase and thus the effect of the same amount of absorbing gas additionally introduced into the medium will depend on its initial concentration. Despite this, this work is cited by many authors, who try to prove that the so-called saturation effect for CO₂ in the atmosphere does not occur. They show the results of numerical calculations for their models, where each subsequent portion of the absorbing gas introduced into the atmosphere causes strong increase of the absorption of thermal radiation emitted by the lithosphere. The report [12], based on the presented reasoning, shows relatively high sensitivity of climate change to CO₂ emission (temperature increases by 3.2 K when CO₂ emission is doubled).

Meanwhile, in the work [7], it was shown that when CO₂ emission is doubled the temperature increases only by 0.6 K. In turn, work [3] shows that when CO₂ concentration is changed from 300 ppm to 600 ppm, the temperature on the Earth's surface increases by no more than 0.2 K.

In the report by Legalization Laboratory in Livermore [4], the authors point out the complexity of problems related to climate description and the very high uncertainty of the results obtained by computer calculations on the basis of assumed models. Attention is also drawn to the hasty adoption of visions of other planets, especially Venus, due to many different factors that are not taken into account in the models.

Thus, as already mentioned in the introduction, these differences inspire the experimental verification of the obtained results. The phenomenon of the absorption of infrared radiation (emitted by the lithosphere) by carbon dioxide contained in the higher cooler layers of the atmosphere, described very precisely in [7], cannot raise any doubts about its occurrence. However, doubts may relate to the amount of the saturation of the absorption when the concentration of CO₂ in the atmosphere increases. It should also be noted that other gases that absorb thermal radiation in the atmosphere have a significant impact on this phenomenon. Their absorption spectra overlap to some extent with the CO₂ absorption spectrum. Thus, the impact of the CO₂ concentration on the absorption of thermal radiation is reduced.

2. Application of radiation emitted by the Moon to study the atmosphere

Investigating the transmission spectrum of the atmosphere at small distances, especially horizontally, seems to be relatively simple. To make such measurement only the radiator (warmed-up body) and the monochromator with a photodetector set at the right distance from each other are required. However, if the greenhouse effect is considered, vertical tests using objects at a sufficiently high altitude should be carried out. Artificial satellites can be used for this experiment, but it is easier to use the natural satellite – the Moon. The temperature of its surface varies a lot, but for the part illuminated by the Sun, according to encyclopaedic information, it may slightly exceed 1100°C. Therefore, the spectrum of thermal radiation emitted by it can be described with a good approximation as a Planck distribution. This spectrum along with the CO₂ transmission spectrum are shown in Fig. 3.

It can be seen that part of the spectrum emitted by the black body overlaps with the carbon dioxide absorption spectrum (made using the HI-TRAN2004 software).

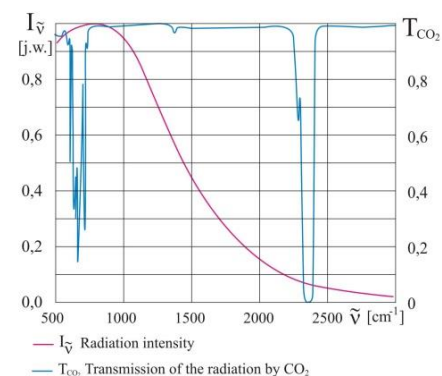


Fig. 3. Comparison of the spectrum of the black body radiation at 1100°C with the CO₂ transmission spectrum

Of course, it should be remembered that the surface of the moon is heated by solar radiation with a spectrum distribution corresponding to black body with a temperature of over 50,000 K. Part of this radiation is reflected by the surface of the Moon and, along with its thermal radiation, propagates towards the Earth. The spectrum of the combined radiation is, of course, continuous, but due to high temperature of the sun, its maximum should shift towards shorter wavelengths, better overlapping with the 4.3 μm (2326 cm⁻¹) absorption band of CO₂. Thus, infrared radiation coming from the moon should be more attenuated by carbon dioxide than radiation emitted by black body at 1100°C.

The concept of using the moon radiation to study the Earth's atmosphere is not a new issue. At the end of the nineteenth century the spectrum of radiation coming from the moon after passing through the Earth's atmosphere as well as absorption bands corresponding to various active gases were described [2]. At present, the Moon is also used in many experimental works to investigate the Earth's atmosphere [11, 14].

3. Experiment

The considerations presented in the previous chapters inspire to ask the question of what is the transmission of radiation with a continuous distribution, e.g. emitted by the black or grey body at a specific temperature, that vertically passes through the Earth's atmosphere with a CO₂ layer of appropriate thickness and pressure and temperature much lower than the temperature of the radiation source.

To answer this question, the experiment was performed where infrared radiation from the Moon reaching the Earth's surface after passing through the Earth's atmosphere was used to light the cuvette filled with the carbon dioxide. The appropriate experimental setup was assembled. The first experiments were carried out in a laboratory using an artificial source of radiation simulating the Moon. The experimental setup is presented in Fig. 4.

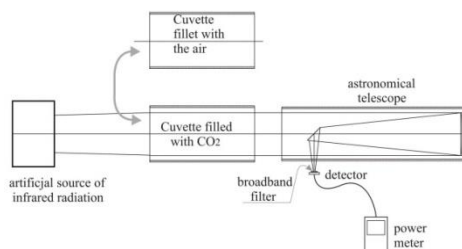


Fig. 4. Experimental setup for measuring the transmission of infrared radiation by a cuvette filled with CO₂

The setup uses two identical cuvettes in the form of plexiglass pipes with a diameter of 250 mm and a length of 500 mm closed with polyethylene foil windows. One cuvette was filled with carbon dioxide while the other was used as reference cuvette filled with air. The radiation emitted by the artificial source, after passing through the cuvette, was focused by the astronomical telescope Soligor MT-800/8"E on the S401C detector connected to PM200 power meter. The telescope was characterized by the diameter of the first mirror $D = 200$ mm, focal length $F = 800$ mm and the shading diameter of the second mirror $d = 60$ mm. The active surface of the detector was shielded by a broadband filter in the form of a flat-parallel germanium plate.

Polyethylene foil windows and a filter in the form of a flat-parallel germanium plate were chosen because of their appropriate transmission spectra. The comparison of these spectra with the carbon dioxide transmission spectrum and the emission spectrum of black body at the temperature of 1100°C (corresponding to the moon's temperature) are shown in Fig. 5.

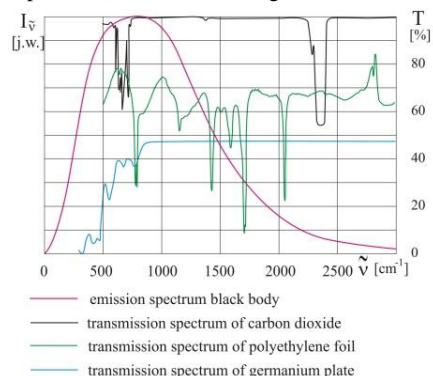


Fig. 5. Transmission spectra of polyethylene foil, germanium plate and CO₂ compared with the emission spectrum of black body at 1100 °C

It can be seen that from the infrared spectrum emitted by black body at a temperature of 1100°C, the germanium plate cuts out unimportant part of low-frequency radiation and slightly reduces the radiation at ~ 15 μm (666 cm⁻¹) wavelength absorbed by CO₂. In turn, the polyethylene foil cuts out bands that do not overlap with CO₂ absorption bands. Finally, it can be stated that the setup's sensitivity to measure the absorption of black body radiation by

CO₂ was increased. The photo of the developed experimental setup is shown in Fig. 6.

The experimental setup was assembled on a tripod enabling vertical full angle rotation and horizontal rotation from zero to ninety degrees.

The diagram of the artificial source of radiation simulating the Moon is shown in Fig. 7.

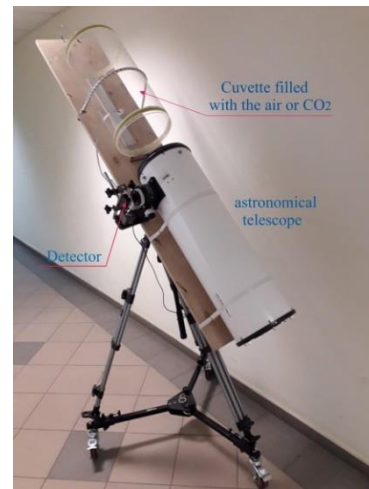


Fig. 6. Photo of the experimental setup

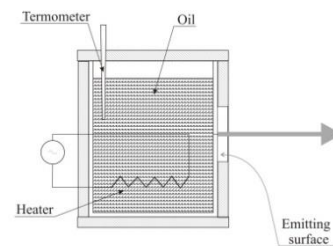


Fig. 7. Diagram of the artificial source of radiation simulating the Moon

The source consisted of a cylindrical glass vessel that was characterized by 100 mm diameter and 150 mm height. The vessel was filled with mineral oil and thermally insulated with 10 mm thick polyurethane foam with a hole of 40 mm diameter for emitting the radiation. The emissive surface of the vessel (behind the hole in the foam) was matted. With the help of an electric heater the oil was heated to 1100°C. The temperature was measured using the ETI 810-930 electronic thermometer. The radiation source was placed at a distance of 50 cm from the polyethylene window of the cuvette as shown in Fig. 4. Just before the right measurements the power meter was reset covering the radiation source with a plywood plate (at temperature of ~ 200°C).

The measurements of the power of the infrared radiation incident on the detector were made alternately for the cuvette filled with the carbon dioxide and filled with the air. In each case, 500 measurements every 0.01 seconds were made and the average power was determined. The measurement was repeated three times for the first cuvette and then three times for the second cuvette. The above measurements were repeated again six times. The results of the described experiment are shown in Tab. 1.

Table 1. The power of infrared radiation emitted by the artificial source after passing through the cuvettes

Subsequent measurements	Power for cuvette with CO ₂ , [μW]	Subsequent measurements	Power for cuvette with air, [μW]
1	267.2	2	310.4
	265.1		309.4
	263.7		308.5
3	264.6	4	309.2
	263.2		310.1
	262.9		308.9
5	264.2	6	307.9
	266.7		310.1
	263.9		308.7
Average value	264.6	Average value	309.2

The presented measurements show that the power of radiation incident on the detector in the case of the cuvette filled with air is equal to $I_p = 309.2 \mu\text{W}$ while for the cuvette filled with CO_2 is equal to $I_{\text{CO}_2} = 264.6 \mu\text{W}$.

Assuming, that the power of radiation incident on the first cuvette window is I_0 , the power of radiation incident on the detector for the carbon dioxide and the air can be expressed by the following formulas, respectively:

$$I_{\text{CO}_2} = I_0 \cdot T_{ok}^2 \cdot T_{\text{CO}_2} \cdot T_L \cdot T_F \quad (3)$$

$$I_p = I_0 \cdot T_{ok}^2 \cdot T_p \cdot T_L \cdot T_F \quad (4)$$

where: T_{ok} – transmission of the radiation for the polyethylene window, T_L – transmission of the radiation for the telescope, T_F – transmission of the radiation for the germanium plate, T_{CO_2} – transmission of the radiation for CO_2 in the cuvette, T_p – transmission of the radiation for the air in the cuvette.

Using equations (3) and (4) and assuming that $T_p = 1$, the following formula can be derived:

$$T_{\text{CO}_2} = \frac{I_{\text{CO}_2}}{I_p} \quad (5)$$

By inserting the measured values from Tab. 1 into formula (5), the value of transmission of radiation for a 50 cm thick carbon dioxide layer at atmospheric pressure and temperature of 20°C can be obtained and it is equal to $T_{\text{CO}_2} = 0.86$.

It should be remembered that this radiation was emitted by a grey body (matted glass surface) at a temperature of 1100°C and passed through two windows of polyethylene foil and the germanium plate, while it did not pass through the thick layer of the Earth's atmosphere, which is the main object of the investigation. The second part of the experiment was devoted to this issue. The infrared radiation from the moon after passing through the Earth's atmosphere was measured. The experiment was carried out at cloudless night, during the full moon, at $\sim 2^\circ\text{C}$, on the roof of the IOE WAT building. The measurement system used for the experiment is shown in Fig. 8.

When the optical axis of the experimental setup was set towards the clear sky (away from the face of the Moon), the power meter was zero. When the optical axis was set towards the face of the Moon, the power of the radiation incident on the detector was measured. During the measurement, the optical axis of the experimental setup was inclined from the vertical by an angle of $\sim 35^\circ$.

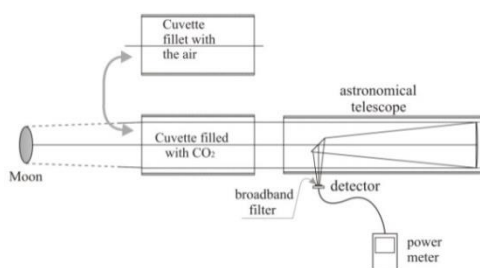


Fig. 8. Experimental setup for measuring the transmission of infrared radiation from the Moon by CO_2

The measurements were carried out in the same way as in case of the laboratory experiment exchanging the cuvette filled with CO_2 and with the air. The achieved results are shown in Tab. 2.

Table 2. The power of infrared radiation from the Moon after passing through the cuvettes

Subsequent measurements	Power for cuvette with CO_2 , [μW]	Subsequent measurements	Power for cuvette with air, [μW]
1	123.2	2	122.4
	121.7		123.1
	119.5		120.0
3	120.7	4	119.4
	122.1		121.3
	119.5		122.0
5	121.5	6	120.8
	119.1		118.3
	120.7		122.1
Average value	120.9	Average value	121.0

The obtained results show that the average value of radiation power from the Moon, after passing through the cuvette filled with carbon dioxide and with the air, is the same this time. This means that according to formula (5), the value of infrared radiation coming from the moon to the Earth's surface through carbon dioxide at atmospheric pressure, temperature of $\sim 2^\circ\text{C}$ and thickness of 50 cm is equal to $T_{\text{CO}_2} \cong 1$. Of course, it should be referred to the difference of temperature in laboratory ($T = 20^\circ\text{C}$) and on the terrace ($T = 2^\circ\text{C}$). Thus, according to the Schwarzschild equation (1) and analysis presented in [7], stronger attenuation of the infrared radiation by CO_2 should be expected at lower temperature than at a higher temperature. Thus, the temperature change could not cause an increase of transmission of CO_2 .

4. Summary of the results of experimental work

The designed and carried out experiment has both methodological and cognitive value. It shows a relatively simple way to measure the attenuation of radiation emitted by grey body with a modified spectrum when passing through a specific gas with a defined thickness, temperature and pressure. The results of the measurements can be interpreted as follows. Infrared radiation from the moon, as a result of absorption by various gases dissolved in the atmosphere, especially by CO_2 and water vapour, changes its spectrum after reaching the Earth's surface. The "holes" in this spectrum overlaps with the carbon dioxide absorption bands in the cuvette. Of course, it can be argued whether this phenomenon can be called saturation of CO_2 concentration, because after all due to the overlapping of absorption bands, other gases also participate in it. However, the most important conclusion is that, unlike during the measurements in the laboratory, the additional layer of CO_2 with thickness of 50 cm at atmospheric pressure, placed on the Earth's surface, does not noticeably reduce the infrared radiation flux from the Moon, despite the fact that Moon's temperature is much higher than the temperature of carbon dioxide in this layer.

5. Proposal for further experiments

As was already mentioned, the flux of thermal radiation emitted by a solid, when passing through a gaseous media can decrease due to absorption in this gas only when the temperature of this gas is lower than the temperature of the solid. Therefore, active gases dissolved in the air at low altitudes, where the temperature is comparable to the surface temperature of the Earth, will not attenuate this radiation and only at higher altitudes, where the temperature is much lower, such attenuation may occur. Therefore, without going into all the complexities of the described phenomena, one can carry out a simple ideological experiment consisting in the measurement of the transmission of thermal radiation from the Earth by carbon dioxide in the cuvette (similarly to the conducted "moon" experiment) at appropriate subsequent heights. This can be accomplished by placing the cuvettes with focusing optics and measuring devices in the balloon basket, and after dropping subsequent portions of ballast, carry out measurements at the corresponding heights. Of course, the temperature and pressure in the cuvettes should be the same as outside of them, while the CO_2 concentration in the cuvette filled with carbon dioxide should always be 100%. In this way, assuming that carbon dioxide is evenly distributed throughout the globe, one can consider a layer with thickness equal to the length of the cuvette and estimate how much carbon dioxide is contained in it and how such amount of CO_2 at the considered height will reduce transmission of the radiation from the Earth.

After taking into account the results of the measurements carried out at different heights, a reliable model could be prepared to determine the effect of the increase of the concentration of CO_2 in the atmosphere on the decrease of transmission of thermal radiation from the lithosphere.

It is worth adding that in the balloon basket, in addition to the aforementioned apparatus and basic instruments for measuring air

parameters, it would also be possible to place a set of appropriate gas collection containers in order to accurately check the distribution of the concentration of individual gases in the atmosphere, and thus enable to provide more reliable data for currently used models.

It seems that the use of a balloon is an optimal solution, because it can be relatively easily to reach to all layers of the atmosphere that are important in climate processes and, additionally, the balloon stopping at the appropriate altitudes, should not introduce major distortion of gas concentrations in the surrounding atmosphere.

6. Conclusions

One could think that the cognitive value of the experiment is small and of little practical importance. Moreover, the measurements of transmission of infrared radiation from the Moon were done, not from the earth, as it is in case of the greenhouse effect. It is known that the Earth's temperature is much lower than Moon's temperature and therefore the emission spectrum of the Earth is slightly shifted towards longer wavelengths. Additionally, based on results presented in [7], in contrast to radiation from the Moon absorbed by the entire atmosphere, noticeable absorption of radiation from the Earth will occur only in the colder layers of the atmosphere located at higher altitudes. However, it should be noted that in both cases there is absorption of radiation of continuous spectrum, emitted by the grey body and passing through thick layers of the atmosphere. Thus, in both cases, the "widening" of absorption lines associated with saturation effects described at the beginning of this work will happen and the phenomena described in [5] will have the impact on the absorption of radiation. Thanks to this, the presented work allows to look more critically at the frequently presented reality. As was already mentioned in the introduction, there is the conviction, that based on the formula (2) derived in paper [5], the increase of the concentration of absorbing gas in the air, regardless of its initial state, will cause strong increase of the absorption of the infrared radiation of continuous spectrum. Meanwhile, the experimental results obtained in this work, disprove this general theorem while it is the main basis for introducing restrictions on entities responsible for CO₂ emissions.

Nevertheless, disapproval of one of the more serious arguments regarding this statement, based on the presented results of the measurement, it is not possible to determine what is the real impact of the increase of CO₂ concentration in the atmosphere on the decrease of the transmission of thermal radiation from the lithosphere. Only on the basis of measurements carried out in the proposed experiment, it would be possible to get closer to the true picture of reality.

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