

# Global warming by thermal absorption of CO<sub>2</sub>

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## Abstract

The present study describes the absorption of thermal radiation from the earth's surface by CO<sub>2</sub> in the atmosphere. For this purpose the absorption spectrum for the vibrational-rotational structure of the CO<sub>2</sub> molecule is described. Only the vibration bands around 15, 5, and 4.3-micron wavelengths are important for the absorption spectrum. The vibration band around 15-micron wavelength has by far the largest absorption and is located almost at the top of the emission curve at an average earth temperature of 294 K. The individual rotational absorption lines of both the *P*- and *Q*- branches are 50 GHz apart. Due to pressure broadening, they overlap. The absorption by CO<sub>2</sub> is saturated. The absorbed radiation is converted into local heat via collisional relaxation, which then reaches both the earth and space through radiation. In the case of only CO<sub>2</sub>, increasing the concentration of CO<sub>2</sub> will have a negligible effect on the global warming. In the presence of water vapor as well, absorption will occur over almost the entire spectrum. In this case too, increasing CO<sub>2</sub> will practically not affect the global temperature.

## Introduction

A climate catastrophe caused by CO<sub>2</sub> is nothing more than a hypothesis. The ideas of this hypothesis are based only on unproven assumptions. The models derived from this hypothesis predict a catastrophic climate with higher temperatures that have so far proven to be inaccurate over the past decades.

Climate is a very complex problem that unfortunately does not lend itself to experimental research. A better approach for research has been found in extensive computer calculations of all relevant physical processes<sup>1</sup>. The results have shown that there is little influence of CO<sub>2</sub> on the global temperature. However, these opaque

computer calculations and the accompanying database are difficult to understand.

In the present study, a simple model was chosen that addresses only the core of the problem. The question is whether an increase in greenhouse gases leads to more absorption of the emitted thermal radiation and whether this affects radiation transport into space, resulting in a global temperature increase. This is done by analyzing the absorption of CO<sub>2</sub>. For the current concentration of CO<sub>2</sub>, the absorption for thermal radiation is found to be saturated. In the absence of other greenhouse gases, the absorption is limited to only a few vibration bands of CO<sub>2</sub>. Increasing CO<sub>2</sub> will not give any remarkable change. In the presence of water vapor and CO<sub>2</sub>, absorption will occur over virtually the entire spectrum. It will be many times greater and it will give much less cooling. More CO<sub>2</sub> will not affect this process.

### **Vibrational- rotational structure of CO<sub>2</sub>**

The CO<sub>2</sub> molecule has strong infrared absorption bands that belong to the symmetric stretching vibration  $\nu_1$ , the degenerated bending vibration  $\nu_2$  and the asymmetric stretching vibration  $\nu_3$ . The vibrational states are usually indicated with the quantum numbers  $n_1, n_2$  and  $n_3$  of the respective vibrational modes. A vibrational transition during absorption may be accompanied with a rotational transition. For the electronic ground state the molecules with symmetric vibrations ( $\nu_1, \nu_2$ ) have only even rotational  $J$ -numbers in the rotational spectrum, while the asymmetric vibration  $\nu_3$  has only odd  $J$ -numbers. According to the selection rule the transition of the absorbing molecule changes the rotational quantum number of the molecule with  $\Delta j = \pm 1$ , or 0. The  $P$ -branch transitions correspond to  $\Delta j = -1$ , the  $Q$ -branch to  $\Delta j = 0$  and the  $R$  – branch to  $\Delta j = +1$ .

The bending mode is degenerated because there are two perpendicular vibrations. Quantum theory tells us that the degree of degeneracy if  $n_2$  quanta are excited is equal to the number of different ways in which  $n_2$  can be written as the sum of two integers which is equal to  $n_2 + 1$ . This degeneracy is indicated with the integer  $l$ . The vibrational state of CO<sub>2</sub> is described by the notation  $(n_1 n_2^l n_3)$ . For instance, for  $n_2=2$  we have  $l = 0, l = 2$  or  $l = -2$ . The value  $l = 0$  is considered as a linear motion of the C-atom, whereas  $l = 2$  and  $l = -2$  are circular motions in opposite directions. During the circular motion the

internuclear distances are unchanged and therefore they have no coupling with the  $\nu_1$  and  $\nu_3$  vibrations.

The rotational energy  $E_r$  of the molecule, according to quantum theory described by the integer  $j$ , is  $(2j+1)$ -fold degenerated. Its energy is

$$E_r = B h c j (j + 1) \quad (1)$$

The constant  $B = 0.42 \text{ cm}^{-1}$  depends slightly on the vibrational state. It is determined from the spectroscopy of the laser lines<sup>2</sup>. Since depending on the vibrational state either odd or even  $j$ -numbers are missing, the rotational distribution is expressed by

$$n_{vj} = N_v \left( \frac{2hcB}{kT} \right) (2j + 1) e^{\left[ -\frac{hcB}{kT} j(j+1) \right]} \quad (2)$$

where  $n_{vj}$  is the number of molecules with rotational number  $j$  and vibrational state  $v$ .  $N_v$  is the total number of molecules in that vibrational state per unit volume. The value of  $j$  for which the maximum density is reached is found as

$$j_{max} \approx \sqrt{\frac{kT}{2Bhc}} - \frac{1}{2} \quad (3)$$

For example,  $j_{max} = 16$  for  $T = 300 \text{ K}$  and  $14$  for  $T = 243 \text{ K}$ .

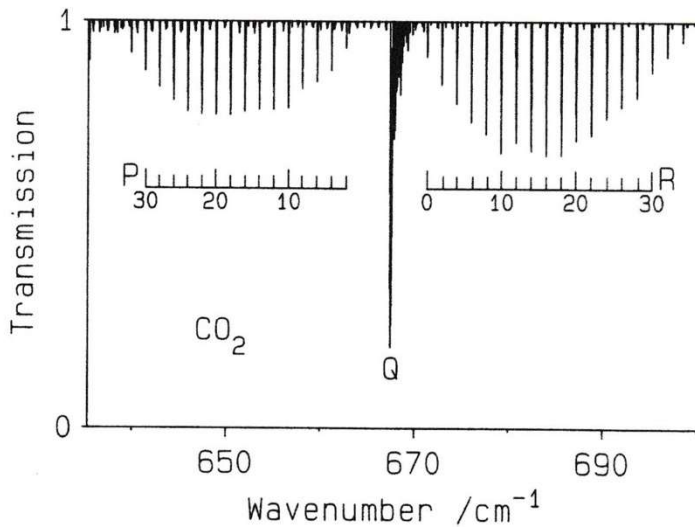


Fig. 1

For  $P$ -branch transitions the rotational energy is decreased by:

$$\Delta E_r = hcB[j(j + 1) - (j - 1)j] = 2hcBj$$

The frequency difference between two adjacent absorbing rotational lines in the  $P$ -spectrum is then:

$$\Delta \nu_r = 4cB = 5 \times 10^{10} \text{ Hz}$$

Similarly, we find the same difference in the  $R$ -branch. For the  $Q$ -branch all transitions coincide with the same absorbing wavelength. It should be noted that these frequency differences are independent on the rotational quantum number  $j$ . See fig.1.

At atmospheric pressure the line shape of the absorption is broadened by collisions where the kinetic energy of the colliding molecule takes care of the excess energy of the absorbing photon. The difference between the absorbed thermal photon and the molecular transition photon is exchanged with the kinetic energy of the colliding particles. The theory of this process turns out to be exceedingly complex, and is in many regards incomplete<sup>3,4</sup>. In the limit that collisions have zero duration time it can be shown that the line shape is Lorentzian<sup>5</sup>. The normalized Lorentzian line shape function is then given by:

$$S(\nu, \nu_{12}) = \frac{\Delta\nu_p}{2\pi} \left[ (\nu - \nu_{12})^2 + \left( \frac{\Delta\nu_p}{2} \right)^2 \right]^{-1} \quad (4)$$

where  $\Delta\nu_p$  is the pressure dependent linewidth that corresponds to the frequency separation between the points where  $S(\nu, \nu_{12})$  has half its maximum value and  $\nu_{12}$  is the center frequency of the transition. It is independent on the transition and can be measured as function of gas composition, pressure and temperature<sup>6,7</sup>. Derived from CO<sub>2</sub> laser measurements the linewidth can be represented by the following formula:

$$\Delta\nu_p = 5.53 p \sqrt{\frac{300}{T}} \text{ MHz} \quad (5)$$

where  $p$  is the air pressure in torr.

At a pressure of 760 torr and a temperature of 273 K we find  $\Delta\nu_p = 4.4 \times 10^9 \text{ Hz}$ , at 500 torr and 243 K this quantity is  $3 \times 10^9 \text{ Hz}$ .

Since the collisions last for a short but finite duration the line shape leads to small but observable deviations from the ideal Lorentzian line shape. Far away from its center the Lorentzian line decays rather slowly whereas there are both theoretical and observational reasons to believe that the tails of the line die off faster than predicted by this Lorentzian profile<sup>4</sup>. Thus, although there is a strong decrease of absorption far away from the line center the satellite observations show (Fig.2) homogeneous absorption between the rotational lines indicating that deviations from the Lorentz profile are not significant in the frequency range of our interest. Those are the frequencies from the center of a line to half the frequency distance with its neighboring rotational line where the absorption cross section is at its minimum. It is about 5 times the linewidth.

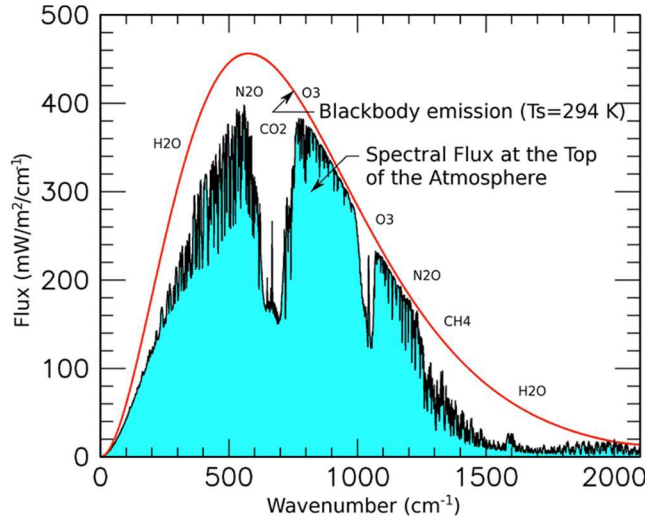


Fig.2

The absorption cross section for the simultaneous vibrational and rotational excitation from the initial state  $\nu_{vr}$  to the final state  $\nu_{vr}^*$  can be derived as<sup>2</sup>:

$$\sigma_{\nu_{vj} \rightarrow \nu_{vj}^*} = \frac{\lambda^2 (2j^* + 1)}{8\pi (2j + 1)} S(\nu, \nu_{12}) A_{\nu_{vj}^* \rightarrow \nu_{vj}} \quad (6)$$

where  $A_{\nu_{vj}^* \rightarrow \nu_{vj}}$  is the transition rate for spontaneous emission from the initial rotational-vibrational state  $\nu_{vj}^*$  to the final state  $\nu_{vj}$ . Its value depends slightly on the rotational state<sup>2</sup>.

### Low-lying infrared absorption bands of CO<sub>2</sub>

The population densities of the vibrational modes are conveniently described by the vibrational temperatures. They are 1800 K, 950 K and 3370 K for respectively the  $\nu_1$ ,  $\nu_2$  and  $\nu_3$  vibrations. Since these temperatures are much higher than the atmospheric temperatures only absorption from the ground and low-lying excited states are relevant to our purpose. The most intense ones involve excitations from the ground state to the lowest states of the bending and to the asymmetric vibrations (absorption involving the symmetric stretching vibration is not possible because of the absence of a dipole moment). Especially the absorbed radiation power by the ground state bending mode at about 15 microns is dominant. The absorbed radiation by the asymmetric vibration at about 4.3 micron is much less intense because the emitted radiation from the earth surface has its maximum at a wavelength of about 16-micron and drops fast for shorter wavelength. See fig.2. In the following table<sup>8</sup> the low-lying infrared absorption bands of CO<sub>2</sub> are collected.

( $cm^{-1}$ )	( <i>micron</i> )	Lower state	Upper state
667	14.99	000	01 <sup>1</sup> 0
2349	4.26	000	001
1932	5.17	000	03 <sup>1</sup> 0
2076	4.82	000	11 <sup>1</sup> 0
3609	2.77	000	02 <sup>0</sup> 1
3716	2.69	000	10 <sup>0</sup> 1
4860	2.06	000	04 <sup>0</sup> 1
4983	2.01	000	12 <sup>0</sup> 1
5109	1.96	000	20 <sup>0</sup> 1
618	16.18	01 <sup>1</sup> 0	02 <sup>0</sup> 0
720	13.89	01 <sup>1</sup> 0	10 <sup>0</sup> 0
1886	5.30	01 <sup>1</sup> 0	04 <sup>0</sup> 0
2094	4.78	01 <sup>1</sup> 0	12 <sup>2</sup> 0
2137	4.68	01 <sup>1</sup> 0	20 <sup>0</sup> 0
961	10.4	10 <sup>0</sup> 0	00 <sup>0</sup> 1
1063	9.4	02 <sup>0</sup> 0	00 <sup>0</sup> 1

### The absorption of the individual bands

The (000 – 01<sup>1</sup>0) band is by far the strongest absorber because its wavelength of 15 microns is near the top of the thermal emission curve. The absorption spectrum contains *P*-, *Q*- and *R*-branches<sup>9</sup>. See also fig.1.

The air density is about  $2.78 \times 10^{19}$  molecules per  $cm^3$ . The present content of CO<sub>2</sub> is 400 ppm or 0.04% so that  $N_{CO_2} = 1.1 \times 10^{16} cm^{-3}$ . At atmospheric temperatures practically all molecules are in the vibrational ground state. Although there is a decrease of density with height we consider it for our purpose as constant. For weak absorptions the effective absorption length will then be somewhat longer.

The line shape value  $S_c$  in the center of the line for an air pressure of 760 torr and a temperature of 273 K is according to eq. (4):  $S_c = \frac{2}{\pi \Delta \nu_p} = 1.4 \times 10^{-10} sec.$

The spontaneous emission rate  $A$  can be deduced from the data found in reference 9. These data are obtained with a completely enclosed spectrometer filled with CO<sub>2</sub> and using a grating of 1200 lines and a KBr prism. The slit included about  $0.45 cm^{-1}$ . From these data we calculate for the present

transition the value  $A = 0.08 \text{ sec}^{-1}$ . For  $A = 0.08 \text{ sec}^{-1}$  we obtain by using eq. (6):  $\sigma_{vj} = 10^{-18} \text{ cm}^2$ .

Considering the zero or  $Q$ -branch the absorbed thermal flux density by the  $\text{CO}_2$  molecules can be expressed according to the Lambert-Beer law as:

$$I_{th} = I_0 \exp[-\sigma_{vj} N_{CO_2} h] \quad (7)$$

where  $I_0$  is the emitted flux from the earth surface and  $h$  the height from the surface in cm. Substituting the values for  $\sigma_{vj} = 10^{-18} \text{ cm}^2$  and  $N_{CO_2} = 1.1 \times 10^{16} \text{ cm}^{-3}$  we find the absorption at the line center as  $I_{th} = I_0 \exp(-1.1 \times 10^{-2} h)$ .

Thus, an absorption length  $l_{ab} \sim 1\text{m}$ .

For the  $P$ - and  $R$ -branch the situation is different because the absorption deals with the density of the involved rotational lines. Since for atmospheric temperatures practically all molecules are in the ground vibrational state the number of molecules with the rotational quantum number  $j_{max} = 16$  is then obtained from eq. (2):  $n_{j=16} = 0.078 N_{CO_2} = 8.6 \times 10^{14} \text{ cm}^{-3}$ .

If we now substitute into eq. (7) the cross section  $\sigma_{vj} = 10^{-18} \text{ cm}^2$  and the density  $n_{vj} = 8.6 \times 10^{14} \text{ cm}^{-3}$  we obtain at the line center an absorption distance of 12m. The lowest absorption in the middle of two neighboring rotational lines at the frequency distance of  $2.5 \times 10^{10} \text{ Hz}$  from the center we calculate by using eqs. (4) and (6) ;  $\sigma_{vj}(2.5 \times 10^{10}) = 0.77 \times 10^{-20} \text{ cm}^2$ . The molecular density in the middle is twice the above value or  $17.2 \times 10^{14} \text{ cm}^{-3}$ . The corresponding absorption distance becomes 750 m.

The fraction of the molecules for  $j = 16$  is as indicated 0.078. This number decreases according to eq. (2) with increasing and decreasing  $j$ -values. For  $j = 40$  this fraction is 0.012. This means that emitted infrared radiation that falls within the absorption region of the line  $j = 40$  has at its center an absorption distance of 72m and in the middle of the lines an absorption distance of 4.5km. The fraction for  $j = 2$  is 0.02 so that also these molecules saturate the thermal emission flux at a distance of about 50m and for the frequency in between the lines at 2.6km.

Considering the molecules from  $j = 2$  to  $j = 40$  we find that about 98% of all  $\text{CO}_2$  molecules in the considered band completely saturates the emitted thermal radiation within a distance of about 4.5km. The same result will be obtained by considering the  $R$ - branch. Thus for the present concentration the thermal radiation is saturated and more  $\text{CO}_2$  does not increase absorption.

The (000 - 001) band with a wavelength of about 4.3 microns is also considered as a strong absorber, although the thermal radiation intensity at this wavelength is according to fig.2 a very small fraction compared to that at 15-

micron. For this band we are dealing with *P*- and *R*- transitions. The linewidths of the rotational transitions are again  $\Delta\nu_p = 4.4 \times 10^9 \text{ Hz}$  and the line shape at the center is also  $S_c = \frac{2}{\pi \Delta\nu_p} = 1.4 \times 10^{-10} \text{ sec}$ . From reference 9 we derive for this band  $A = 1.6 \text{ sec}^{-1}$ . Using this value, the cross section for a vibrational-rotational transition is obtained from eq. (6):  $\sigma_{vj} = 1.65 \times 10^{-18} \text{ cm}^2$ . The molecular density for vibrational-rotational transition with  $j=16$  is the same as in the previous band:  $n_{vj} = 8.6 \times 10^{14} \text{ (cm}^{-3}\text{)}$ . Using eq. (7) and the above values for  $n_{vj}$  and  $\sigma_{vj}$  we obtain the absorption distance from  $\sigma_{vj} n_{vj} h = 1$  so that  $h_{ab} \sim 7.5 \text{ m}$ . Similar to the previous band we consider for the *P*- and *R*- branches also the frequency space between the line centers. We find that all thermal radiation is absorbed at the height of 2.7 km.

The  $(000 - 03^10)$  and  $(000 - 11^10)$  bands with wavelengths of about 5 microns can be considered similar to the previous bands. The linewidths and line shapes are equal to those of the previous bands. This transition rate for spontaneous emission can be obtained from reference 9. We use the value  $A = 1.5$ . We find  $\sigma_{vj} = 2.1 \times 10^{-18} \text{ cm}^2$ . For  $j = 16$  we have again  $n_{vj} = 8.6 \times 10^{14} \text{ cm}^{-3}$ . Similar to the previous calculations, complete absorption over all frequencies of this band is reached at a height of 2.2 km.

The absorption of the  $(000 - 02^01)$ ,  $(000 - 10^01)$ ,  $(000 - 04^01)$ ,  $(000 - 12^01)$  and  $(000 - 20^01)$  bands occurs at wavelengths of about 2 microns. However, at 2-micron wavelength there is practically no emission at the considered temperature of 300 K.

The  $(01^10 - 02^00)$  and  $(01^10 - 10^00)$  bands are Fermi coupled at wavelength of about 15 microns. The wavelengths are close to those of the  $(000 - 01^10)$  band and the transitions of these three bands are always observed together in the absorption spectrum. However, the considered bands belong to the population density  $N_1(\nu_2)$  of the first excited state of the bending mode. The density for  $T = 273 \text{ K}$  can to a good approximation be given by  $2N_{CO_2} e^{-\frac{T_2}{T}}$  where  $T_2 = \frac{h\nu_2}{k}$ . We find  $N_1(\nu_2) = 0.06N_{CO_2} = 6.6 \times 10^{14} \text{ cm}^{-3}$ . The linewidths and the line shapes are again the same as used above. For  $j = 15$  we have then the density  $5.2 \times 10^{13} \text{ cm}^{-3}$ . This transition has according to reference 9 the value  $A = 0.8 \text{ sec}^{-1}$ . From eq. (6) we then find the absorption cross section equal to  $10^{-17} \text{ cm}^2$ . We obtain for the line center of  $j = 15$  an absorption height of 20 m. Similar to the previous calculations the absorption saturates over all frequencies at a distance of 7.5 km.



The  $(01^{10} - 04^{00})$ ,  $(01^{10} - 12^{20})$  and  $(01^{10} - 20^{00})$  with wavelength of about 5 microns are very weak absorbers because of their multi-transitions and low density of the excited bending states which depend on the temperature of the environment. Low absorption means that the radiation within their bandwidths will reach high altitudes where the temperature is far below zero and consequently the population of the excited bending state decreases. These transitions, even with higher  $\text{CO}_2$  concentration, are non-absorbers.

The  $(10^0 - 00^0)$  and the  $(02^0 - 00^0)$  bands at about 10-micron wavelength are Fermi coupled. Both bands have *P*- and *R*- branches. The linewidths and the line shapes are again the same as those used above. The transition rate  $A$  is obtained<sup>2</sup> from laser spectroscopy and is for both bands about  $0.17 \text{ sec}^{-1}$ . Using eq. (6) we obtain for the absorption cross section  $9.5 \times 10^{-19} \text{ cm}^2$ . Because the densities of the considered vibrational excited states are relatively low. The density of molecules in the  $(10^0)$  vibrational state can to a good approximation be given by  $N_1(\nu_1) = N_{\text{CO}_2} e^{-T_1/T}$  where  $T_1 = \frac{h\nu_1}{k}$ . Substituting  $T_1 = 1900 \text{ K}$  and  $T = 273 \text{ K}$  we find  $N_1(\nu_1) = 9.5 \times 10^{-4} N_{\text{CO}_2} = 1.1 \times 10^{13} \text{ (cm}^{-3}\text{)}$ . The number of these molecules with the rotational quantum  $J = 16$  is then  $8.5 \times 10^{11} \text{ (cm}^{-3}\text{)}$ . We find the absorption distance for the strongest rotation line at 12.5 km. A comparable result is obtained for the absorption of molecules in the  $(02^0)$  vibrational state. Practically, both bands do not contribute to thermal absorption.

### **Transfer of absorbed infrared radiation**

The two major greenhouse gases that control the thermal radiation flux from the earth surface are  $\text{CO}_2$  and water vapor. The thermal radiation excites vibrational-rotational transitions of these gases. Then, the excited molecules relax by molecular collisions, thereby heating the atmosphere.

Let us first consider the case that the only greenhouse gas is  $\text{CO}_2$ . By far most of the thermal radiation has no interaction with the air molecules and radiates directly into space. Only radiation with frequencies that fit the absorption spectrum of  $\text{CO}_2$  will be absorbed. It is not very likely that these excited molecules will lose their energy by radiation because the emission time is much larger than the relaxation time by molecular collisions. At atmospheric condition the relaxation processes<sup>11,12</sup> are in the order of  $10^{-6} \text{ sec}$ , whereas the emission time may be of the order of  $10^{-3} \text{ sec}$  or larger. The transfer of the energy of the excited stretching vibrations with energy quanta  $h\nu_1$  and  $h\nu_3$  occurs at first internal via collisions with air molecules to the bending modes having the much smaller energy quanta  $h\nu_2$ . The excess quantum energy in this

collision process is exchanged with the translational motion<sup>11</sup>. Then, on its turn the bending modes having the smallest energy quanta will transfer during collisions their energy directly to translational motion of the air molecules. This means that the initially absorbed thermal emission results to local heating of the atmosphere. A part of the thermal radiation from this heating of the atmosphere will reach the earth's surface and increase its temperature in order to balance the incoming radiation from the sun. This is the greenhouse effect of CO<sub>2</sub> connected to global warming. As we have seen above, the absorption of thermal emission is practical saturated by the present concentration values of CO<sub>2</sub>. Thus, in case of increasing the atmospheric CO<sub>2</sub> concentration we will observe a negligible effect on global warming.

In the presence of both CO<sub>2</sub> and water vapor, thermal absorption is strongly enhanced. The absorption by water vapor is very strong. This is due to the strong permanent dipole moment of the asymmetric-top molecule, which also allows for pure rotational transitions in the long wavelength region. Absorption therefore occurs over virtually the entire spectrum. The excited water vapor molecules relax also through molecular collisions, thereby similarly heating the atmosphere. A part of the resulting thermal radiation from this heated atmosphere will then again excite water vapor and CO<sub>2</sub> at a higher level. This process of excitation and heating will continue to occur over a part of the troposphere towards space as long as there is sufficient water vapor. Each time, a part of the produced radiation will go towards the earth's surface and another part towards space. A small part of the radiation produced in this process also excites CO<sub>2</sub> molecules that also heat the atmosphere. With the current concentration of CO<sub>2</sub> there will always be saturation as we have discussed. More CO<sub>2</sub> will also in the presence of water vapor not enhance the global warming.

## Conclusions

The absorption of thermal emission from the earth by CO<sub>2</sub> occurs at wavelengths around 15, 5 and 4.3-microns. By far most absorption is around the 15-micron wavelength of the (000 – 01<sup>1</sup>0) vibrational band. Within all absorption bands, the central frequencies of the rotation lines are 50 GHz apart. However, satellite observations show that the absorption of individual rotation lines overlaps, so that in the frequency domain, the absorption width is determined by the width of the vibration band.

The global heating is the result of radiation absorption by molecular gases, which then is converted into local heat of the air via collision relaxation. The

heated air cools down by radiation, also in the direction of the earth's surface. Only gases that can absorb thermal radiation are relevant for the global heating (oxygen and nitrogen do not do so).

The main greenhouse gases are water vapor and CO<sub>2</sub>. For the current and also for much lower concentrations of CO<sub>2</sub> saturation of absorption has been reached. More CO<sub>2</sub> will practically not absorb more thermal radiation.

The absorption by water vapor occurs over almost the entire thermal spectrum and is weak in areas where CO<sub>2</sub> is active. The global warming by water vapor is therefore much larger than that by CO<sub>2</sub>. However, this warming is not enhanced if the current concentration of CO<sub>2</sub> increases.

In conclusion: The greenhouse effect of CO<sub>2</sub> and water vapor connected to global warming will hardly increase at higher CO<sub>2</sub> concentrations.

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