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The Influence of Atmospheric Trace Gases and Aerosols on Climate

Guido Visconti

Summary

Perturbations in atmospheric trace gases other than CO_2 can affect the surface temperature of the Earth. Some of these gases interact chemically with other compounds and thus they can affect the climate both directly by perturbing the radiation field, or indirectly by inducing perturbations in other radiatively active gases. In the first part of the paper the direct radiative effects are reviewed showing how their cumulative effect is comparable to the warming expected due to CO_2 changes. For the indirect radiative effects results from an interactive radiative-convective photochemical model are presented showing that multiple perturbations can particularly affect the ozone depletion expected from use of Chlorofluorocarbons. In the final part of the paper some results concerning the effects of both tropospheric and stratospheric aerosols on climate are reported. It is shown how background aerosols are responsible for a global cooling comparable to the expected warming introduced by a doubling in the CO_2 concentration.

Zusammenfassung

Auch Veränderungen des Gehalts anderer atmosphärischer Spurengase als CO_2 können die Oberflächentemperatur der Erde beeinflussen. Entweder durch direkte Veränderung des Strahlenhaushaltes oder indirekt über die Beeinflussung anderer strahlungsaktiver Bestandteile durch chemische Reaktionen mit ihnen. Zuerst werden die direkten Strahlungseinflüsse der Spurengase beschrieben und gezeigt, dass deren Gesamteinfluss auf das Klima für mögliche Veränderungen vergleichbar ist mit demjenigen, der bei einer

Veränderung des CO_2 Gehaltes erwartet wird. Die indirekten Strahlungseffekte werden anhand von Modellrechnungen erläutert. Sie zeigen, dass das Zusammenwirken verschiedener Spurengase den durch die Chlorofluororokarbone verursachten Ozonabbau beeinflussen kann. Abschliessend wird der Einfluss der Aerosole in der Troposphäre und Stratosphäre auf das Klima beschrieben. Es wird gezeigt, dass die Hintergrund-Aerosole eine ebenso grosse globale Abkühlung bewirken können, wie der CO_2 Anstieg eine entsprechende Erwärmung.

Résumé

Des perturbations des gaz présents sous forme de traces dans l'atmosphère, autres que le CO_2 , peuvent affecter la température de la surface de la Terre. Certains de ces gaz interagissent chimiquement avec d'autres composés, influençant ainsi le climat à la fois directement, en perturbant le champ radiatif, ou indirectement, par réaction avec d'autres gaz actifs radiativement. Dans la première partie de cet article, on passe en revue les processus radiatifs directs, montrant comment leur effet cumulatif est comparable à un réchauffement présumé dû à une variation du CO_2 . Avant aux effets radiatifs indirects, on présente les résultats obtenus par un modèle photochimique interactif et radiatif-convectif, qui montre que des perturbations multiples peuvent affecter en particulier la baisse du taux d'ozone attendue à la suite de l'usage des composées organiques chlorées et fluorées. Finalement on présente quelques résultats récents concernant les effets des aérosols sur le climat troposphériques et stratosphériques. On montre comment le fond d'aérosols est responsable d'un refroidissement global comparable au réchauffement auquel on

peut s'attendre en doublant la concentration de CO₂.

Introduction

Trace gases present in the Earth's atmosphere influence its thermal structure. Most of these gases have strong absorption band in the infrared and interact with the IR radiation emitted both by the surface and the atmosphere itself and hence contribute to the greenhouse effect. Among these gases water vapor, carbon dioxide and ozone together account for most of the greenhouse radiative effect. Concern has intensified recently over the possible man made perturbations in CO₂ and O₃. In particular carbon dioxide is likely to produce a global warming as its mixing ratio increase in the atmosphere due to anthropogenic activity.

The atmosphere however contains other trace gases of natural or anthropogenic origin. In particular in the last few years attention has been paid to possible perturbations in the concentration of N₂O, CH₄ and chlorofluorocarbons (CFC), notably CFCl₃, CF₂Cl₂ and CCl₄. All these gases have absorption bands in the infrared which may contribute to the greenhouse effect. However unlike CO₂ some of them have also strong chemical interactions with other atmospheric compounds which can also be radiatively active.

A typical example may be CFC's which could affect the ozone concentration especially in the 30-40 km region. In turn the perturbed ozone will have an effect on both the stratospheric thermal structure and surface temperature. At the same time CFC's themselves directly influence the global radiative balance.

From this example we see that trace gases can have a climatic effect both directly by absorbing infrared radiation, or indirectly via chemical interactions with other radiative active gases.

In this paper we will review briefly some of the direct radiative effects, and after a simple introduction we will present some calculation of a fully chemical interacting model to show the indirect radiative effects. A brief mention will also be made of the effect of atmospheric aerosols.

The direct radiative effects

This review of the radiative effects will be based mainly on two documents, the WMO Report No. 14 and the papers by Chamberlain et al. (1982), Ramanathan (1982), Hansen et al. (1982) and Luther (1982).

The Earth's atmosphere is transparent in the 7-13 μm spectral region and several trace gases have absorption bands in the same region. Table 1 gives a list of the gases we will discuss in this paper together with some rele-

Table 1. Trace gases considered in this paper. For each gas the location of main bands are reported together with the absorptance, A(cm⁻¹), the band intensity S(atm⁻¹cm⁻²), the absorber amount (atm cm) present in the atmosphere (Adapted from WMO, Report n. 14).

molecule	Band (μ)	spectral range (cm ⁻¹)	Absorptance, A	Band Strength, S	amount
CO ₂	15	550- 800	190	220	250
	10	850-1100		0.04	
	4.3	2100-2400		2440	
O ₃	9.6	950-1200	70	340	0.3
	14	600- 800			
CH ₄	7.6	950-1650	55	134	1.2
N ₂ O	16.9	520- 660	37	24	0.2
	7.8	1200-1350		218	
	4.6	2120-2270		1247	
CFCl ₃ (CF11)	9.2	800-1200	0.2	1800	1.1 × 10 ⁻⁴
	11.8				
CF ₂ Cl ₂ (CF12)	9.1	850-1250	0.5	2200	2.3 × 10 ⁻⁴
	8.7				
	10.9				

vant spectroscopic data. For a complete list the interested reader may refer to the WMO report. The parameter values shown in this table refer to a particular formulation of the absorption process in which the net flux is calculated according to the relation (Cess and Ramanathan, 1972; Ramanathan, 1976)

$$(1) \quad Q_{\lambda} = -B_{\lambda}(T_s)A(O, \infty) - \int B_{\lambda}(T) dA \quad (5)$$

where Q_{λ} is the net flux at the wavelength centered on the band of interest, $B(T_s)$ is the Planck function at the surface temperature T_s , and $A(z, z')$ is the absorptance between altitudes z and z' . The total band absorptance can be expressed as

$$(2) \quad A(u) = 2A_0 \ln \left\{ 1 + \frac{u}{(4 + u(1 + 1/\beta))} \right\}^{1/2}$$

where A_0 (cm^{-1}) is the bandwidth parameter, u is the dimensionless optical depth, β is the lineshape parameter. The parameters u and β can be expressed as

$$(3) \quad u = Sw/A_0,$$

$$(4) \quad \beta = \beta_0 (p/p_0).$$

In this case S is the band strength in $\text{cm}^{-1} (\text{cm atm})^{-1}$, w is the absorber amount in atm , β is the lineshape parameter calculated for standard atmospheric conditions and p and p_0 are local and reference pressure respectively.

From the above definitions any gas can be characterized by A_0 , S and β_0 . Table 1 also includes values of A for typical unperturbed amount of gases present in the Earth's atmosphere.

The outgoing flux calculated with Eq. (1) and in particular any variation ΔF , due to changes in the mixing ratio of the corresponding gas can be easily related to the change in surface temperature T_s . Following Hansen et al. (1982) we can write for the outgoing total infrared flux:

$$F = \sigma T_e^4$$

where T_e is the equivalent temperature, $T_e = 255 \text{ K}$. If the flux changes by an amount ΔF , we have

$$(6) \quad \Delta T_e / \Delta F = 1/4\sigma T_e^3 = 0.2666 \text{ K}(\text{W/m}^2)^{-1}$$

Changes in the equivalent temperature are simply related to the surface temperature when a fixed lapse rate is assumed. We have

$$(7) \quad \Delta T_s / \Delta F = \Delta T_e / \Delta F$$

Table 2 adapted from Hansen et al. (1982) shows that Eq. (6) holds for almost all trace gases except carbon dioxide. The latter gas requires a much higher change in surface temperature. The reason is that when carbon dioxide increases it also cools the stratospheric temperature, thus decreasing the transmissivity for IR radiation. To restore equilibrium the surface temperature must have a further increase to compensate for the lower transmissivity.

This simple example shows that a more sophisticated approach than Eq. (5) has to be used in order to calculate the climatic effects due to trace gases. In general global radiative equilibrium requires:

$$(8) \quad F(T_s) = S(1 - \alpha_p(T_s))$$

Table 2. Changes in surface temperature resulting from perturbations of trace gases. The Table also reports the computed temperature change between 1970–1980 (Lacis et al. 1981) and between 1960–2010 expected from a more realistic scenario. ΔT_s in this case was calculated from Eq. (5) of Lacis et al. (1981).

Molecule	mixing ratio, X (ppb)	ΔX	$F(\text{Wm}^{-2})$	$\Delta T_s(\text{K})$	$\Delta T_s / \Delta F$	ΔT_s (1970–1980)	X (1960)	X (2010)	$\Delta T_s(\text{K})$
CO_2	3×10^5	3×10^5	2.57	1.3	0.5	0.14	3.1×10^5	9.8×10^4	1.15
CH_4	1600	1600	0.37	0.12	0.31	0.032	1300	960	0.19
N_2O	280	280	0.91	0.29	0.31	0.016	250	70	0.18
CFC1_1	0	2	0.43	0.13	0.29	0.020	0	1.5	0.22
CF_2Cl_1	0	2	0.53	0.15	0.29	0.034	0	1.5	0.27

where α_p is the planetary albedo, S the solar flux and F the outgoing infrared flux. This equation shows explicitly the link between the infrared flux, the planetary albedo and the global average temperature T_s . In particular the albedo α_p could depend on T_s through the so called ice-albedo feedback. Any change ΔT_s will imply a change in both the outgoing infrared flux and the planetary albedo, so we have:

(9)

$$F(T_s + \Delta T_s) = \frac{S}{4} (1 - \alpha_p(T_s + \Delta T_s)) + \Delta Q$$

where

(10)

$$F(T_s + \Delta T_s) = F(T_s) + dF/dT_s \Delta T_s$$

$$\alpha_p(T_s + \Delta T_s) = \alpha_p(T_s) + d\alpha_p/dT_s \Delta T_s$$

and ΔQ is the net change in total flux considering also the flux absorbed in the troposphere and surface. The relation between ΔT_s and ΔQ is now similar to Eq. (7) through a quantity δ called climate sensitivity parameter

(11)

$$\Delta T_s = \delta \Delta Q$$

$$(12) \quad \delta = \left[\frac{dF}{dT_s} + \frac{S}{4} \frac{d\alpha_p}{dT_s} \right]^{-1}$$

For the simple case considered earlier $\delta = 1/4 \sigma T_e$.

Several studies have been reported on the direct effect of atmospheric trace gases e.g. Donner and Ramanathan (1982), Ramanathan (1975), Lacis et al. (1981), Wang et al. (1976). All these studies suggest that the direct combined effects of trace gases can be comparable to the single effect of carbon dioxide. Lacis et al. (1981) in particular show how in the decade 1970-1980 the combined effect of CH_4 , N_2O , CF11 and CF12 amounts to a warming of 0.1 K compared to 0.14 K for CO_2 . The same authors show that doubling the present concentration of N_2O and CH_4 and an increase up to 2 ppb in CF11 and CF12 will introduce a global warming of 1.5 K as compared to 2.9 K for a doubling of CO_2 . It is to be noted however that the effect of CFC could be underestimated with respect

Table 3. List of trace gases whose perturbations have detectable effect on climate. For each gas the main sources and the sinks are reported, its atmospheric lifetime and the possible increase in tropospheric mixing ratio. This is an update of a similar table shown by Wang et al. (1976).

Species	Source	Sinks	Lifetime	increase (%/year)
N_2O	Anaerobic decay	$\text{N}_2\text{O} + h\nu$ $\text{N}_2\text{O} + \text{O}(\text{'D})$	-100 years ^(a)	0.2 ^(a)
CH_4	Anaerobic decay	$\text{CH}_4 + \text{OH}$ $\text{CH}_4 + \text{O}(\text{'D})$ $\text{CH}_4 + h\nu$	8 years ^(b)	1.4 ^(b)
NH_3	Anaerobic decay	$\text{NH}_3 + \text{OH}$ $\text{NH}_3 + h\nu$ washout	10 days ^(c)	
CO	Fossil fuel oxidation of hydrocarbons	$\text{CO} + \text{OH}$		3 ^(b)
CCl_2F_2	Anthropogenic	$\text{CCl}_2\text{F}_2 + h\nu$	70-80 years ^(a)	6 ^(a)
CCl_3F	Anthropogenic	$\text{CCl}_3\text{F} + h\nu$	70-80 years ^(a)	5.7 ^(a)
SO_2	Oxidation of H_2S combustion	washout	20 days ^(c)	
CCl_4	Natural or Anthropogenic	$\text{CCl}_4 + h\nu$	-50 years ^(a)	2.4 ^(a)

a) Golombek and Prinn (1983)

b) Rasmussen and Kahil (1981)

c) Wang et al. (1976)

to CO_2 because of the different growth rates of their concentration in the atmosphere. This means that when the CO_2 concentration will double, the CFC will reach a much higher concentration than 2 ppb.

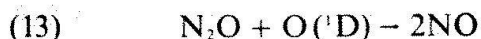
Also shown in Table 2 is the expected warming due to a more realistic scenario for the expected increase in concentration between 1961 and 2010. It can be seen that the effect introduced by trace gases amounts to something more than 43% of the total expected warming.

Indirect climatic effects of trace gases

We will limit the discussion of the indirect climatic effects of trace gases to those gases listed in Table 3. In particular we consider here only the effects of increasing N_2O , CH_4 , CF_{11} , CF_{12} coupled to the increase in carbon dioxide.

a) Nitrous oxide

Nitrous oxide is a source of stratospheric NO_x mainly through the reaction

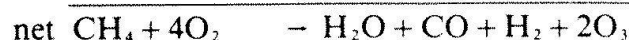
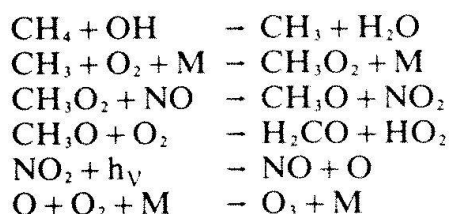


This NO_x source is small (0.3 Tgyr^{-1} , Crutzen and Gidel, 1983) with respect to ground source (20 Tg yr^{-1}). However due to the long residence time of NO_x in the stratosphere, NO_x produced by reaction (13) could actually determine the NO_x abundance in the stratosphere and hence influence ozone abundance. If the N_2O concentration increases a crude approximation would give corresponding NO_x increase in the stratosphere. In the troposphere on the other hand due to decreasing concentration of $\text{O}(^1\text{D})$, the production of NO_x from N_2O oxidation has to be compared to other sources e.g. lightning and the industrial production. On the other hand NO_x contributes both to production and loss of odd oxygen. A net loss will occur above the tropopause so that N_2O increase should correspond to a net ozone decrease in the stratosphere, and an increase in the troposphere. This would actually produce a downward shift in the ozone profile and so contribute to a further warming of the surface.

b) Methane

Methane interacts chemically in a number of ways with other atmospheric compounds and its indirect effect on climate are more complex. In the troposphere, oxidation of methane can produce ozone through the so called smog mechanism (Crutzen, 1974).

(14)



Critical to this mechanism is the presence of NO_2 which is responsible for the production of odd oxygen. In the stratosphere methane forms HCl through the reaction

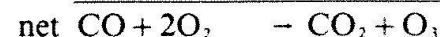
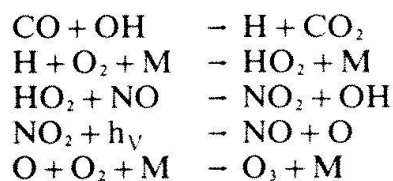


HCl is the only sink for chlorine atoms so that increasing methane could dampen the ozone depletion due to CFC's. Another important effect of methane in the stratosphere is production of H_2O and consequently the production of odd hydrogen. This is however balanced by the first reaction of chain (14). The possible ozone increase due to reaction (15) could provide an additional small contribution to the greenhouse effect.

c) Carbon Monoxide

Carbon monoxide has no direct radiative effects on climate. However its strong interaction, mainly with hydroxyl radicals, makes its perturbation very important. CO may increase due to industrial activity and as for CH_4 its oxidation starts by reacting with OH

(16)



Ozone production however is possible only if the ratio of NO to O_3 is greater than 2×10^{-4}

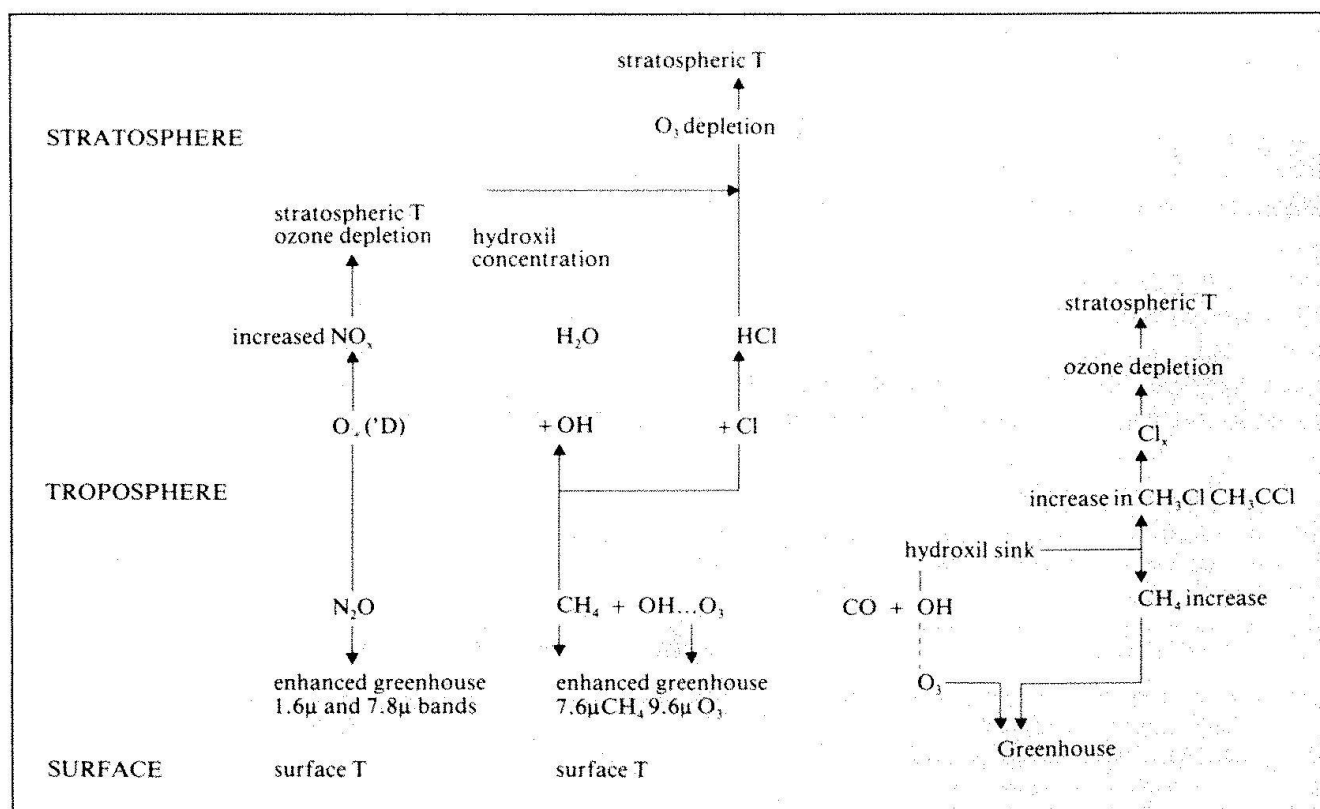


Fig. 1. Summary of the direct effects and indirects of Nitrous Oxide, Methane and Carbon Monoxide on the radiation balance in the atmosphere.

(Crutzen and Gidel, 1983) otherwise oxidation of carbon monoxide will lead to ozone destruction. The most important effect of a CO increase however would be the depletion of OH. This in turn would affect the concentration of CH₄, CH₃Cl and CH₃CCl₃ which are destroyed mainly by reacting with OH. As a result an increase in carbon monoxide concentration could lead to an ozone increase in the troposphere accompanied by larger mixing ratios of chlorine bearing gases. This will result in the availability of more chlorine atoms in the stratosphere for the ozone destruction process. Increase in methane concentration will also result in an enhanced greenhouse effect. Fig. 1 summarizes some of the effects we have discussed.

Studies of the effect of such complex interactions can only be carried out using coupled radiative photochemical models (Wuebbles et al., 1983; Callis et al., 1983; Nicoli and Visconti, 1983). We report here some of the results from the model of Nicoli and Visconti (1983). These results illustrate some typical consequences of coupled trace gas

concentrations and are summarized in Table 4. (Notice that all the cases referred to are for the steady state situation.) Some interesting conclusions can be drawn from these results. The simple temperature feedback introduced by the ozone depletion has an important self healing effect. When ozone decreases in the 40 to 50 km region then the UV heating in the same region is reduced. As a consequence temperature in the upper stratosphere decreases more than 10 K. There is a resulting slowing down of the reaction $\text{O} + \text{O}_3$ (which is temperature dependent) and is responsible for most of the ozone destruction. An additional effect of the same kind is introduced by the CO₂ doubling. This doubling brings the depletion figure to 9% in comparison to the 10.9% that would be obtained with no temperature feedback at all. Simple perturbations in methane and nitrous oxide alone also result in ozone and temperature changes. As expected increase in methane concentration corresponds to an ozone increase in the stratosphere (due to enhanced HCl production) and in the troposphere (due to the smog mechanism). The

corresponding temperature changes is a warming of both the troposphere and stratosphere.

The effect of increasing nitrous oxide is to decrease slightly stratospheric ozone (via enhanced NO_x production) which in turn produces a stratospheric cooling. The additional greenhouse effect produces a surface warming of a few tenths of degree.

When perturbations are coupled together the most notable results is the non linearity in both the temperature and ozone changes.

Climatic Impact of Aerosols

We conclude this paper with a discussion of some recent result on the effects of atmospheric aerosols on temperature. Aerosols are present in both the troposphere and stratosphere. Tropospheric aerosols account for most of the atmospheric optical thickness in the visible, but they have a short residence time. Consequently their distribution changes widely depending on local or regional factors. However it is possible to say

Table 4. Summary of the results from coupled perturbations of trace gases on stratospheric and surface temperature, and changes on columnar and local ozone density. These results were obtained with a fully interactive one-dimensional radiative-convective, photochemical model (Nicoli & Visconti, 1983).

Case	Perturbation	Change in temperature (k) stratospheric	surface	Change in ozone (%) columnar	local
1	Stratospheric Chlorine increase from 2.3 to 10 ppb no temperature feedback			- 10.9	- 4.5 at 40 km
2	Same as above with temperature feedback	- 12 at 45 km	0.1	- 9.3	
3	CO_2 concentration doubled from 320 to 640 ppm		+ 1.8	+ 2.2	+ 28 at 42 km
4	CO_2 doubled and 10 ppb of Cl_x	- 18 at 45 km	1.67	- 9	- 42 at 40 km
5	Methane concentration doubled	+ 0.5 at 45 km	+ 0.27	+ 1.7	+ 5 at 42 km + 10 at 10 km
6	Nitrous oxide concentration doubled	- 0.7 at 40 km	+ 0.35	- 2.7	- 11 at 40 km
7	doubled N_2O , doubled CH_4 and 10 ppb of chlorine with no feedback	-	-	- 6.7	- 8 at 40 km
8	doubled N_2O , CH_4 , CO_2 and 10 ppb of chlorine	- 16.5 at 45 km	+ 2.30	- 4.7	- 32 at 40 km

Table 5. Albedo and temperature changes, introduced by the background natural occurring aerosols (Coakley et al. (1983)). Both changes are annual averages.

latitude (deg)	unperturbed albedo	visible optical thickness	perturbed albedo	change in surface temperature (k)
5	0.08	0.16	0.0037	- 2.2
15	0.09	0.16	0.0030	- 2.2
25	0.10	0.16	0.0026	- 2.4
35	0.10	0.12	0.0021	- 2.5
45	0.12	0.12	0.0020	- 2.9
55	0.14	0.12	0.0018	- 3.7
65	0.24	0.07	0.0006	- 4.1
75	0.46	0.07	- 0.0001	- 5.2
85	0.61	0.07	- 0.0004	- 5.5
Average			0.0025	- 2.9

that on a global basis the amounts of tropospheric aerosols is expected to increase. An example is the possible perturbation in the SO_2 content of the troposphere.

Stratospheric aerosols have much longer residence times and considerably smaller optical thickness. The stratospheric aerosol load can be perturbed by catastrophic volcanic eruptions which sometime increase the optical thickness in the visible by up to 0.1, i.e. to values comparable to those produced by tropospheric aerosols. These large injections are transient phenomena, and hence are very useful tools to study the response of the climatic system to perturbations in the radiative field. The main effect of atmospheric aerosols is to change the albedo of the Earth-atmosphere system. This effect however depends on both the nature of the aerosols and the underlying surface. If the reflectivity of the surface is the only parameter, then the effect of the aerosols is to smooth the albedo difference, i.e., the albedo will increase over ocean surface (low albedo) and decrease over land (high albedo). The effect as a function of latitude is more complex because an account has to be taken of the change in average cloud and the effect of increasing zenith angle of the sun. Both factors have a latitudinal dependence. One of the more recent and comprehensive studies on the effect of tropospheric aerosols has been reported by Coakley et al. (1983). In this study detailed radiative calculations show the expected change in albedo introduced by the background, natural occurring aerosols. An energy balance model is then used to assess the effects of the same aerosol on the surface temperature. Table 4, adapted from the work just mentioned, summarizes some of their results. These depend somewhat on the absorbing properties of aerosols. However they indicate an average global cooling of the order of 2-3 K which is comparable to the warming expected by doubling the trace gases concentrations. On a global basis if we refer to Eq. (5) and (9) we would expect, based on a change $\Delta\alpha$ in the albedo, a change $\Delta\alpha_p$ in temperature $\Delta T = \delta S \Delta\alpha_p / \alpha_p$ (with α global average albedo and S the solar constant). Using $\alpha_p = 0.3$ we have $\Delta T = 3.3$ K. The conclusion suggests that background tropospheric aerosols play a very important role in the

thermal balance of the Earth and that perturbations in their optical thickness could results in a global cooling.

This conclusion seems to be supported by some evidence that volcanic eruptions produced a transient global cooling. In this case however the albedo change is introduced by stratospheric aerosols. These are formed in situ from the sulfur bearing gases emitted during the eruption. Stratospheric aerosols by absorbing solar radiation could also produce local heating raising the stratospheric temperature by a few degrees. The most complete simulation of both the surface and local effect is that of Hansen et al. (1978). Based on an optical thickness of 0.1 they were able to reproduce an hemispheric surface cooling of 0.3 K and a stratospheric heating between 4 and 6 K as observed (Newell, 1970; Newell and Weare, 1970; Angell and Korshover, 1977). It is to be noted however that a similar analysis carried out for the 1982 El Chichon eruption did not verify with observations with the same accuracy for either the troposphere or the stratosphere.

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