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ATMOSPHERIC AND EARTH PHYSICS

EFFECTS OF THE ATMOSPHERIC AEROSOL ON THE OPTICAL PROPERTIES OF CLOUD

GABRIELA IORGA[*], SABINA STEFAN**

** Department of Physics, Faculty of Chemistry, University of Bucharest,*
Regina Elisabeta Avenue, No. 4-12, 70346 - Bucharest, Romania
*** Department of Atmospheric Physics and Earth Sciences, Faculty of Physics, University of Bucharest, P.O. Box MG-11, 077125, Bucharest-Magurele, Romania*

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Abstract.The work is focussed on the role that pre-cloud aerosol (number concentration and spectral dispersion) plays in cloud reflectivity. Rural and marine aerosols have been described by lognormal aerosol size distributions, and the cloud condensation nuclei were obtained based on Abdul-Razzak's activation parameterisation. Cloud albedo including dispersion in the scattering asymmetry factor and specified aerosol properties has been estimated. The sensitivity of cloud albedo to changes in cloud droplet number concentration in connection to the changes in liquid water content and the cloud droplet effective radius has been investigated. For the same liquid water path, clouds over rural areas have more and smaller cloud droplets and thus a higher cloud albedo than marine clouds.

Key words: aerosol size distribution, indirect aerosol effect, effective radius, liquid water content, cloud albedo.

Atmospheric aerosol and optical properties of clouds

1. INTRODUCTION

The aerosol number or mass concentration is a key parameters in the studies of atmospheric radiative effects. The assessments of radiative effects of aerosols, especially of the indirect effect on the radiative balance of the Earth are highly uncertain (IPCC, 1996). Uncertainty arises in part, because of the differences in the assumed aerosol concentration and evaluation of the cloud droplets number concentration (CDNC). On the other hand, the uncertainty in the estimates of indirect radiative forcing results from the complexity of the relationship between the aerosol mass input to the atmosphere and both the radiative properties of clouds and cloud dynamic.

In the IPCC (2001) is shown, comparatively with the estimates from the Second Assessment Report (SAR), the level of scientific understanding regarding to the global, annual mean radiative forcing (W/m^2) due to a number of agents for the period from pre-industrial (1750) to present (late 1990s, about 2000). It is striking that consideration of all of the estimates available since 1996 lead to the same best estimate (–0.4 W/m^2) and uncertainty (–0.2 to –0.8 W/m^2) range. Here are again emphasized the large uncertainties appearing from the fact that the indirect forcing due to tropospheric aerosols is poorly understood and the determining of the radiative forcing is a hard challenge.

[*] Corresponding author.
E-mail addresses: giorga@gw-chimie.math.unibuc.ro (G. Iorga), sabina_stefan@yahoo.com (S. Stefan)

The aim of this study is to provide results of the theoretical experiments in order to improve the estimates of indirect effect of aerosol on the cloud albedo and consequently on the radiative forcing.

The cloud properties could be changed primarily because of changing aerosol concentration in the atmosphere. Only a part of aerosol interacts effectively with water and will, in turn, determine the number concentration of cloud droplets. The relationship between the total aerosol number concentration and the cloud droplet number concentration is a non-linear one, and may vary with location and season and many model studies have included it. Martin et al. [12] proposed a one-to-one correspondence between number concentration of cloud condensation nuclei (CCN) and cloud droplet number concentration (CDNC) in the sub-cloud layer in clean maritime air-masses. We assumed this suggestion in our study, in section 2, to calculate the effective radius (r_{eff}), cloud optical thickness (τ) and liquid water path (LWP). The CDNC and r_{eff} were calculated for various types of aerosol using the parameterisations proposed by Abdul-Razzak [1,2] and Liu and Daum [11], respectively.

Several studies, prove that, during the cloud-aerosol interaction, cloud geometrical thickness can change, so the predicted CDNC may change combining the effects of changes in droplet concentrations and changes in geometrical thickness of clouds. The results reported by Han et al [6] from analysed satellite data show for most continental clouds with $\tau > 15$ clouds albedo increase with decreasing r_{eff} and for the $\tau < 15$ over oceans, cloud albedo decreases with decreasing r_{eff} .

In the present work using the microphysical properties of clouds carried out in section 2 we emphasized the sensitivity of cloud albedo to changes in CDNC in connection to the changes in liquid water content (LWC) and the cloud droplet effective radius (r_{eff}), in section 3. We have also presented in this section the results related to changes in shortwave radiative forcing as function of cloud albedo, optical thickness and liquid water path. In section 4 we summarize the results obtained.

2. MICROPHYSICS AND OPTICAL PROPERTIES OF THE CLOUD

In the climate models, the optical properties of clouds and cloudiness are the important parameters. Aerosol optical thickness, cloud optical thickness and the effective radius of cloud droplets are among these important retrieval parameters but all of them are related to microphysical processes of aerosol and clouds.

2.1 The activation of aerosol particles, theoretical considerations and data used

The aerosol types used in this study were rural and urban as continental aerosol and marine aerosol. The size distributions of these aerosol types were derived from measurements [8,9,15]. Also, the chemical composition of the aerosol is based on the measurements reported in literature, which show that the continental aerosol contains 15-30% sulfate, while marine aerosol contains somewhat more (30-60%), and clearly most of the sulfate for both types of aerosol can be found in the accumulation mode.

The percent of 15-30% sulfate assures that fine particles aerosols at common humidity are hygroscopic ones and they behave as optically ideal droplets.

Aerosol characteristics and dynamics determine the number of nucleated droplets. In order to investigate the importance of physico-chemical properties of aerosol particles on the radiative properties of stratiform clouds we have considered an aerosol size distribution given by the sum of three lognormal size distributions, as described by the equation 1:

$$\frac{dn_a}{d\log a} = \sum_{i=1}^{3} \frac{n_{a,i}}{\sqrt{2\pi}\log\sigma_i} \exp\left(- \frac{\left[\log(a/a_{m,i})\right]^2}{2(\log\sigma_i)^2} \right) \tag{1}$$

where $n_{a,i}$, $a_{m,i}$ and σ_i are the total number concentration, geometric mean dry radius, and geometric standard deviation of aerosol mode *i*, respectively. The modes are: *i* = 1 nucleation mode, *i* = 2 accumulation mode and *i* = 3 coarse mode.

The chemical composition of the rural aerosols is assumed to be a mixture of ammonium sulfate (75%) and mineral dust (25%) as appeared in Bott [4]. The corresponding coefficients of the aerosol number size distributions are shown in Table 1.

Table 1

Modal parameters of number size distributions for rural aerosols

	$n_{r,i}$ (cm^{-3})	r_i (μm)	$\log\sigma_i$
R-W	1000	0.008	0.20
	800	0.034	0.32
	0.72	0.460	0.34
R-J	6650	0.007	0.225
	147	0.027	0.557
	1990	0.042	0.266

Table 2

Modal parameters of number size distributions in the case of urban aerosol

	$n_{r,i}$ (cm^{-3})	r_i (μm)	$\log\sigma_i$
U-W	$2120 \cdot 10^3$	0.006	0.240
	$37 \cdot 10^3$	0.031	0.297
	4.9	0.540	0.328
U-J	$9.93 \cdot 10^4$	0.006	0.245
	$1.11 \cdot 10^3$	0.007	0.666
	$3.64 \cdot 10^4$	0.025	0.337

Table 3

Modal parameters of number size distributions for a marine aerosol

	$n_{r,i}$ (cm^{-3})	r_i (μm)	$\log\sigma_i$
M-HF	100	0.027	0.25
	120	0.105	0.11
	6	0.210	0.45
M-W	340	0.005	0.20
	60	0.035	0.30
	3.1	0.310	0.43
M-J	133	0.004	0.66
	66.6	0.133	0.21
	3.1	0.290	0.40

The Table 2 contains the modal parameters of the urban aerosols as given by Whitby (U-W) and Jaenicke (U-J).

The marine aerosols are assumed to consist of pure ammonium sulfate for the aerosol size distributions of Hoppel and Frick (M-HF) and Jaenicke (M-J). The marine aerosols of Whitby (W) consist of 61% of $(NH_4)_2SO_4$ and 39% NaCl. The coefficients appearing in Eq.(1) are given in Table 3.

The parameterization used to compute the fraction of activated aerosol particles as function of supersaturation S_m , for the aerosol mode "*i*" is that resulted from the analysis of Abdul-Razzak [1,2], pertaining to the cloud droplets formation from an activated aerosol in a parcel of air rising adiabatically.

$$f_i(Sm) = \frac{1}{2} \cdot erfc\left\{ \frac{\ln\left[f_1(\ln\sigma)\cdot\left(\frac{\xi}{\eta}\right)^{3/2} + f_2(\ln\sigma)\cdot\left(\frac{Sm^2}{\eta+3\xi}\right)^{3/4} \right]}{3\sqrt{2}\cdot\ln\sigma} \right\} \qquad (2)$$

where the two functions of standard deviation are considered as:
$f_1(\ln\sigma) = 0.5\cdot\exp\left\{2.5\cdot(\ln\sigma)^2\right\}$, and $f_2(\ln\sigma) = 1 + 0.25\cdot\ln\sigma$, respectively.

2.2. Effective radius

In liquid water clouds the parameters that describe the radiative properties of clouds, the optical thickness of cloud, the single scattering albedo and asymmetry factor, can be parameterized in terms of the effective radius, r_{eff} . This parameter was defined by Hansen and Travis [10] as the ratio between the third moment to the second moment of a droplet size distribution.

Many studies [11,12] show the effective radius can be parameterized as a "1/3" power law of the ratio between the cloud liquid water content and the cloud droplet number concentration, with some pre-factors which are dependent of spectral dispersion of droplet size distribution.

$$r_{eff} = PF \cdot \left(\frac{LWC}{N_{CDNC}}\right)^{\frac{1}{3}} \qquad (3)$$

The effective radius of cloud particles has been computed using the pre-factors as [11]:

$$PF = 62.04 \cdot \frac{\left(1+2d^2\right)^{2/3}}{\left(1+d^2\right)^{1/3}} \qquad (4)$$

where the d signifies the spectral dispersion ($d = \frac{\sigma}{\bar{r}}$) of droplet size distribution. An increase in d may act to negate the effect of increased N on r_{eff} and on cloud reflectivity [11]. We have assumed that the spectral dispersion of the aerosol number size distribution characterizes the CDNC size distributions and we have considered only the values of the spectral dispersion corresponding to the accumulation mode of the various aerosol types.

Figures 1 and 2 show the effective radius dependence of CDNC for different values of liquid water content, for continental (rural and urban) stratiform clouds. The dependence of values of effective radius on CDNC, in case of two values for liquid water content of 0.1 g/m^3 and 0.3 g/m^3, are shown in Fig. 3, for maritime case.

Results show the strong dependences between the effective radius, liquid water content, and spectral dispersion. For the maritime case, results show, for the same liquid water content of 0.1 g/m^3, effective radius range is between 5.4 and 15 µm for a spectral dispersion of 0.1 and 5.9 and 20 µm for a spectral dispersion of 0.3. An increase of liquid water content leads to higher values for effective radius from about 9 to about 14 µm. For larger values than 150 cm^{-3} of CDNC, the values of r_{eff} are the same, closed on 4 µm. The dependences are similar in the case of continental clouds; one can observe the smaller values of r_{eff} for higher concentrations of CDNC. The most likely values for effective radius for continental stratiform clouds lie in the range 2 ÷ 4µm. The values are similar with those measured by Han et al. [6] and simulated by Boucher and Lohmann [3] with ECHAM and LMD GCM models.

For the maritime case, results show, for the same liquid water content the effective radius range is between 6 and 20µm, depending on the CDNC values. An increase of liquid water content leads to higher values for effective radius from about 9 to about 19µm. The values are also similar with those measured by Han et al. [6].

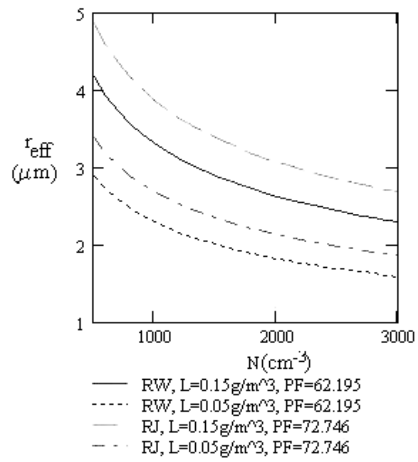


Fig. 1 - Cloud droplet effective radius as a function of CDNC for rural air masses.

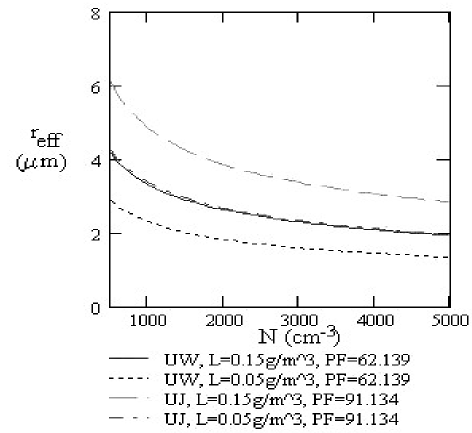


Fig. 2 - Cloud droplet effective radius as a function of CDNC for urban air masses.

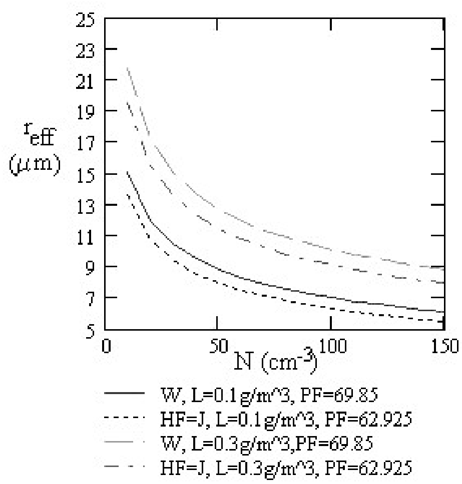


Fig.3 - Cloud droplet effective radius as a function of CDNC for marine air masses.

2.4. The Liquid Water Path and the cloud optical thickness

Another important parameter for study of the albedo of clouds and consequently of the radiative forcing is liquid water path LWP (gm^{-2}), the product between the liquid water content (LWC) and the cloud geometrical thickness. The LWC (g/m^3), cloud droplet number concentration (cm^{-3}) and the effective radius (µm) are related by the following equation:

$$LWC = \frac{4}{3}\pi r_{eff}^3 \rho N \tag{5}$$

where ρ is the water mass concentration.

We have considered in further computations of cloud parameters the values of liquid water path below 150 g/m^2, tacking into account thus only non-precipitating stratiform clouds. In addition we have assumed all CCN become cloud droplets. LWP increases with increase of cubic effective radius.

We have used the correlation between effective radius and liquid water path and optical thickness [13]:

$$\tau = \frac{3}{2} \cdot \frac{LWP}{\rho \cdot r_{eff}} \tag{6}$$

Regarding the dependence of τ of r_{eff} for a fixed value of LWP (not shown here), we have found an increase of optical thickness with the decrease of effective radius, so that for r_{eff} = 12 µm and a value of LWP of 150 g/m^2, corresponds value of about τ = 18 for a cloud with geometrical thickness of 500 m.

3. RESULTS RELATED TO CLOUD ALBEDO AND RADIATIVE FORCING OF CLOUDS

3.1. Cloud albedo and the sensitivity study

The cloud albedo, A, was computed taking into account the two-stream approximation of a nonabsorbing, horizontally homogeneous cloud [10]:

$$A = \frac{\sqrt{3}(1-g)\tau}{2+\sqrt{3}(1-g)\tau} \tag{7}$$

where □ is the optical depth of the cloud, and g denotes the asymmetry factor.

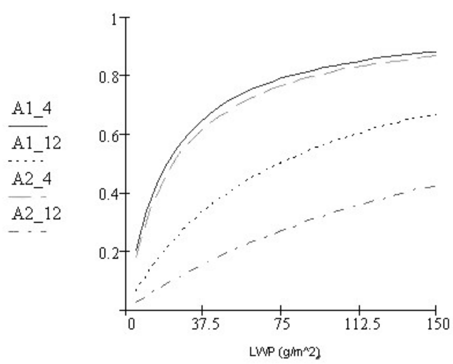


Fig. 4a - Dependence of cloud top albedo of LWP for values of r_{eff} of 4 and 12 µm, for visible (A1) and near-infrared ranges (A2).

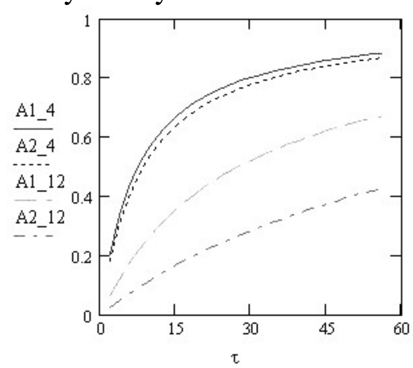


Fig. 4b - Dependence of cloud top albedo of optical thickness for values of r_{eff} of 4 and 12 µm, for visible (A1) and near-infrared ranges (A2).

The dependence of albedo on liquid water path and optical thickness is shown in Figs. 4a, b for maritime stratiform clouds. The higher albedo values for the visible range than those for the near infrared and the increase of albedo with the decrease of r_{eff} for both spectral ranges were obtained. This significant contrast between the albedo values obtained for the considered spectral ranges is much more reduced in the case of continental clouds. As an example, for LWP of 75 g/m^2, there is a difference in cloud reflectivity of about 0.2 in the maritime case and one of only about 0.02 in the continental cloud case.

Pertaining to the variation of albedo function of optical thickness, Fig. 4b shows that τ = 15 can be considered as a separator value for two areas with different rate of increase of the albedo, because the curve slopes are different; we have found a anti-correlation between A and r_{eff} for the entire considered range of τ.

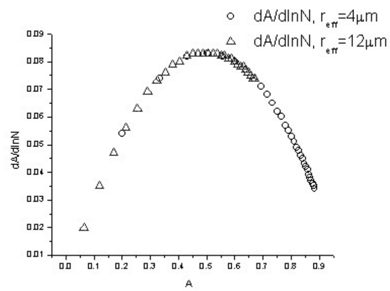


Fig. 5a - Estimates of sensitivity of cloud albedo in the visible range for maritime stratiform clouds of effective radius of 4 and 12 μm.

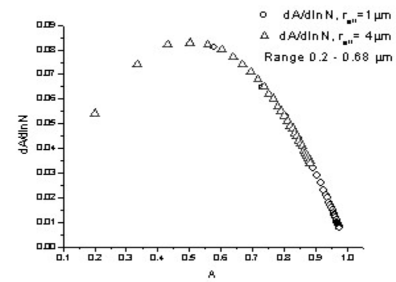


Fig. 5b - Estimates of sensitivity of cloud albedo in the visible range for continental stratiform clouds of effective radius of 4 and 1 μm.

Sensitivity of cloud albedo, dA/dlnN, is shown in Fig. 5a for two values of effective radius (12μm and 4μm) and for 1μm and 4μm in Fig. 5b, in visible spectral range. Sensitivity of cloud albedo for near-infrared range and for the same effective radii is shown in Fig. 6a, and Fig. 6b, respectively.

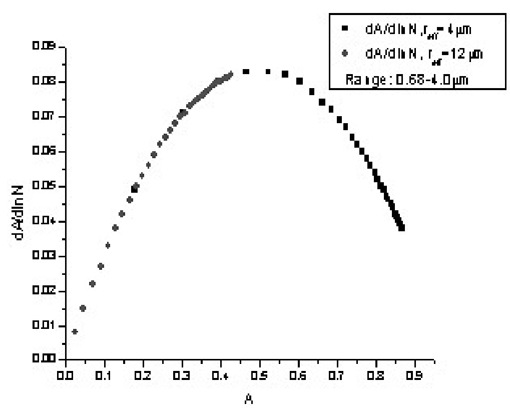


Fig 6a Estimates of sensitivity of cloud albedo in the near-infrared range for maritime stratiform clouds of effective radius of 4 and 12 μm.

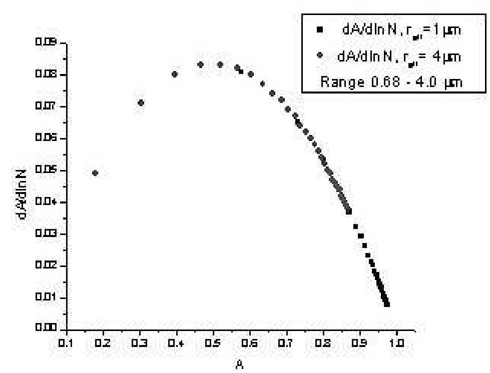


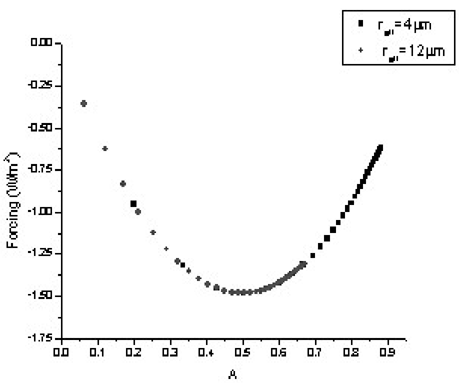
Fig 6b Estimates of sensitivity of cloud albedo in the near-infrared range for continental stratiform clouds of effective radius of 4 and 1 μm.

The sensitivity of cloud albedo to changes in cloud droplet number concentration has maximum values in the range 0.3 ÷ 0.7, as Charlson et al. [5] predicted. For visible range, one can observe a small flat range for A between 0.43 and 0.53 (as Twomey [14] predicted) only in the case of maritime clouds. Furthermore, for near infrared range, for both types of clouds, as for visible in the case of continental clouds, the sensitivity reaches its maximum for a value of cloud top albedo of 0.5. An increase in cloud droplet number concentration of 1% leads to an increase of ~ 8.4 10^{-4}in cloud albedo and in the case of maritime clouds in the visible range an increase of 1% in CDNC determine a increase of ~ 8.3 10^{-4}.

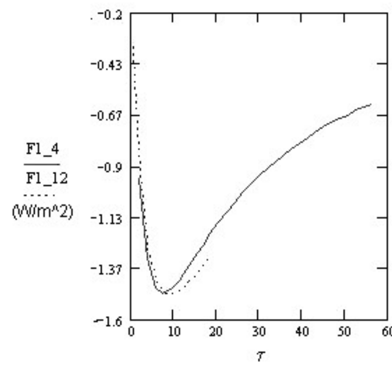
3.2. Changes in shortwave radiative forcing

Among possible radiative forcing that can cause long-term climate change, the aerosol indirect effect is still the most uncertain (IPCC, 2001). In idea to improve the knowledge in this subject, we have studied the changes of radiative forcing versus albedo, LWP and τ, for the 12 µm and 4 µm values of effective radius of cloud droplets (Figs. 9a, b, c). The macrophysical parameters, transmittance and cloud coverage are considered as constant values of 0.76 and 0.3, respectively.

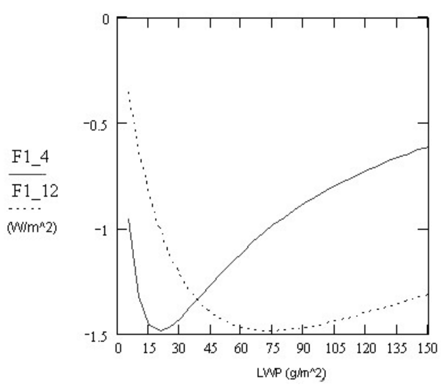
One can be observed (Figs. 7a,b,c) that the radiative forcing reaches its maximum value of ~-1.48 W/m^2 for an albedo value of 0.5, $\tau < 15$ and LWP ~ 20 g/m^2 for effective radius of 4 µm. When the effective radius ranges between 4 and 12 µm, the forcing values seem to be the same but they are attained for large values of LWP. A similar result was obtained from the dependences of forcing of LWP for values of cloud fractional coverage ranging from 0.2 to 1 and for different values of atmospheric transmittance (not shown here). As we expected, the forcing increased as the cloud fractional coverage increased or, when the atmospheric transmittance increases.



(a)



(b)



(c)

Fig. 7 - Dependences of indirect forcing as function of cloud albedo (Fig. 7a), of the optical thickness (Fig. 7b), and liquid water path (Fig 7c), for two values of the effective radius (4 and 12 µm) in the visible range of the electromagnetic spectrum and for maritime stratiform clouds.

In the same macrophysical conditions but for effective radius of 4 and 1 µm, the values of the radiative forcing of clouds are about ~-0.45 W/m^2. Thus, for the 0.5 value of albedo, the maximum value of radiative forcing is –1.48 W/m^2, about three times grater than in case with smaller effective radius.

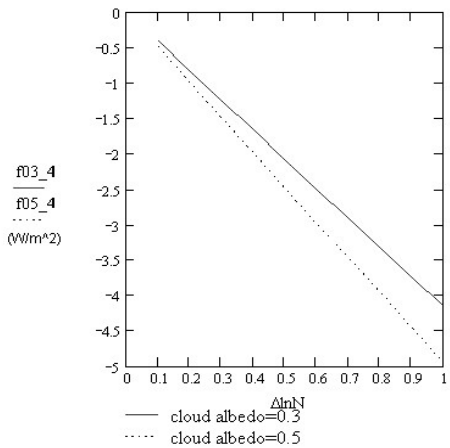


Fig. 8 - Dependence of radiative forcing of the relative increase in the cloud droplet number concentration for two values of cloud top albedo.

The dependence of radiative forcing of the relative increase in the cloud droplet number concentration (Fig. 8) for the 0.3 and 0.5 values of the cloud top albedo, shows that the values of radiative forcing are greater in case of albedo of 0.5 than 0.3, and increase with the increasing of concentration.

Can also be seen that an increase with 30% could determine an increase of forcing with about –0.25 W/m^2 in case of maritime clouds and an increase with about –0.20 W/m^2 in case of continental clouds.

This result suggests that would be interesting to carry out some controlled experiments with a global climate model to find out how this value of 0.5 for cloud albedo can influence the climatic response.

4. CONCLUSIONS

We have carried out a detailed study related to influence of microphysics on albedo clouds and consequently on radiative forcing to show that cloud feedback processes are not limited to macrophysical cloud properties, such as cloud amount and cloud altitude. The study was carried out for maritime and continental stratiform clouds.

First of all, the results show that the cloud radiative properties are sensitive to cloud microphysical aspects such as droplet concentrations. They emphasize the following:

For the same liquid water content, effective radius increase with the increasing of the spectral dispersion of CDNC. For larger values than 150 cm^{-3} of CDNC, one can observe the smaller values of r_{eff} for higher concentrations of CDNC.

Variations of cloud albedo with optical thickness (τ) or LWP are correlated in two ranges of τ divided at a value of about 15, confirming the results of Han [6].

Higher values for cloud albedo in the case of the continental clouds and in the optical spectral range of 0.2-0.68 μm were obtained.

The maximum value of sensitivity of cloud albedo is obtained for an exact 0.5 value of the albedo for both types of clouds, except the case of maritime stratiform clouds in the visible range where was found maximum value for 0.43 ÷ 0.53 range of A.

The radiative forcing reaches its maximum value of ~-1.48 W/m^2 for an albedo value of 0.5, $\tau < 15$ and LWP ~ 20 g/m^2 for effective radius of 4 µm. When the effective radius ranges between 4 and 12 µm, the forcing values seem to be the same but it is attained for large values of LWP. In the same macrophysical conditions but for effective radius of 4 and 1 µm, the values of the radiative forcing of clouds are about ~-0.45 W/m^2. Thus, for the 0.5 value of albedo, the maximum value of radiative forcing is –1.48 W/m^2, about three times grater than in case with smaller effective radius.

REFERENCES

1. H. Abdul-Razzak, S. J. Ghan, C. Rivera-Carpio, 1998: A parameterization of aerosol activation, 1. Single aerosol type, J. Geophys. Res., 103, 6123-6132.
2. H. Abdul-Razzak, S. J. Ghan, 2000: A parameterization of aerosol activation, 2. Multiple aerosol type, J. Geophys. Res., 105, 6837-6844.
3. O. Boucher, U. Lohmann, 1995: The sulfate-CCN-cloud albedo effect: A sensitivity study with two general circulation models, Tellus 47B, 281-300.
4. A. Bott, 2000: A numerical model of the cloud-topped planetary boundary layer: influence of the physico-chemical properties of aerosol particles on the effective radius of stratiform clouds, Atmos. Res., 53, 15-27.
5. R. J. Charison., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley, JR., J. E. Hansen, D. J. Hofmann, 1992. Climate forcing by anthropogenic aerosols, Science, 25, 426–430.
6. Q. Han, W. B. Rossow, J. Chou, R. M. Welch, 1998: Global survey of the relationships of cloud albedo and liquid water path with droplet size using ISCCP, J. Climate, 11.
7. J. E. Hansen, L. D. Travis, 1974. Light scattering in planetary atmospheres, Space Sci. Rev., 16, 527–610.
8. W. A. Hoppel, G. M. Frick, 1990: Submicron aerosol size distributions measured over the tropical and South Pacific, Atmos. Environ., 3, 645-659.
9. R. Jaenicke, 1988: Aerosol physics and Chemistry, in Landolt-Boernstein (Ed.), Zahlenwerte und Funktionen aus Naturwissenschaften und Technik, Vol. 4b, Springer, 391-457.
10. A. A. Lacis, J. E. Hansen, 1974: A parameterization for the absorption of solar radiation in the Earth's atmosphere, J.Atmos. Sci., 31.
11. Y. Liu, P. H. Daum, 2000: Which size distribution function to use for studies related to effective radius, Extended abstract for presentation at the 13th International Conference on cloud and precipitation, Reno, NV, 14-18 August.
12. G. M. Martin, D. W. Johnson, A. Spice, 1994: The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds, J. Atmos. Sci., 51, 13.
13. S. Stefan, G. Iorga, 2004, Effects of aerosol on the optical properties of clouds, Extented abstract for poster presentation at the 14th International Conference on Clouds and Precipitation, Bologna, Italy, 18-23 July
14. S. Twomey, 1991. Aerosols, clouds and radiation, *Atmos. Environ.*, 25A, (11), 2435-2442.
15. K. T. Whitby , 1978: The physical characteristics of sulfur aerosol, Atmos. Environ., 12, 135-159.
**INTERGOVERNMENTAL PANEL ON CLIMATE CHANGE (IPCC), 2001. Climate Change, The Scientific Basis: Aerosols, their direct and indirect effects, Intergovernmental Panel on Climate Change, Cambridge University Press, New York.
**INTERGOVERNMENTAL PANEL ON CLIMATE CHANGE (IPCC), 1996. Climate Change 1995, The Science of Climate Change, J. T. Houghton, L.G. Meira Filho, B. A. Callander, N. Harris, A. Kattenberg, and K. Maskell, (Eds.), Cambridge Univ.Press.