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Analytical Studies of the Cloud Droplet Spectral Dispersion Influence on the First Indirect Aerosol Effect

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ABSTRACT

Atmospheric aerosols (acting as cloud condensation nuclei) can enhance the cloud droplet number concentration and reduce the cloud droplet size, and in turn affect the cloud optical depth, as well as the cloud albedo, and thereby exert a radiative influence on climate (the first indirect aerosol effect). In this paper, based on various relationships between cloud droplet spectral dispersion (ε) and cloud droplet number concentration ($N_{\rm c}$), we analytically derive the corresponding expressions of the cloud radiative forcing induced by changes in the cloud droplet number concentration. Further quantitative evaluation indicates that the cloud radiative forcing induced by aerosols for the different $\varepsilon-N_{\rm c}$ relationships varies from -29.1% to 25.2%, compared to the case without considering spectral dispersion ($\varepsilon=0$). Our results suggest that an accurate description of $\varepsilon-N_{\rm c}$ relationships helps to reduce the uncertainty of the first indirect aerosol effect and advances our scientific understanding of aerosol-cloud-radiation interactions.

Key words: spectral dispersion, cloud radiative forcing, the first indirect aerosol effect

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1. Introduction

Atmospheric aerosols have a direct influence on the local and global radiation balance by their scattering and absorption of solar radiation (Schwartz, 1996). However, by acting as cloud condensation nuclei or ice nuclei, they also affect the cloud optical properties by enhancing the cloud droplet number concentration and reducing the cloud droplet size, as reviewed by Ramanathan et al. (2001). The first aerosol indirect effect (Twomey, 1977) is defined by aerosol effects on cloud droplet sizes for a constant liquid water path, which has attracted much more attention because of its complexities and uncertainties in aerosol–cloud interactions (Li et al., 2009; Wang et al., 2010). Recent research has indicated that, for the first indirect aerosol effect, the cloud radiative forcing can exist over a very broad range varying from -0.3 W m^{-2} to -1.8 W m^{-2} (Forster et al., 2007).

Recent studies have shown that cloud droplet spectral dispersion, which represents the relative dispersion of cloud droplet size distribution, can influence the aerosol indirect effects including the radiative forcing of clouds and precipitation change induced by increasing aerosols. According to the relationships between cloud droplet number concentration ($N_{\rm c}$) and the cloud droplet spectral dispersion (ε), several analytical and numerical modeling results have indicated that aerosols can exert additional effects by enhancing the cloud droplet spectral dispersion in polluted backgrounds and reduce the indirect radiative forcing by anthropogenic aerosols. The research analytically estimated that the cloud radiative forcing for the first indirect aerosol effect can be significantly reduced by in-

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cluding the spectral dispersion (Liu and Daum, 2002). Using GCMs, subsequent studies have confirmed the estimated corresponding magnitude of the reduced cloud radiative forcing to be about 15% according to Peng and Lohmann (2003), and between 12% and 35% according to Rotstayn and Liu (2003). Additionally, it has also been found that the cloud droplet spectral dispersion can significantly influence the cloud-rain autoconversion process, whereby cloud droplets grow into embryonic raindrops (Liu and Daum, 2004; Xie and Liu, 2009, 2010). More recently, numerical experiments have indicated the increasing spectral dispersion significantly affects the cloud microphysical properties including warm and ice phase clouds, and it also changes the surface accumulated precipitation in mesoscale convective systems (Xie and Liu, 2011).

Many observed results have indicated that there is a direct dependence of the cloud droplet spectral dispersion (ε) on the cloud droplet number concentration (N_c) (Martin et al., 1994; Grabowski, 1998; Liu and Daum, 2002; Peng and Lohmann, 2003; Rotstayn and Liu, 2003; Daum et al., 2007; Ma et al., 2010; Xie et al., 2013[a]). However, it should be pointed out that the $\varepsilon-N_\mathrm{c}$ relationships remain highly uncertain. Martin et al. (1994) derived a $\varepsilon-N_\mathrm{c}$ positive relationship (i.e. an increase of ε with N_c) according to the warm stratocumulus clouds with continental air masses and maritime air masses. A few observational studies also indicated a similar $\varepsilon-N_\mathrm{c}$ positive relationship to evaluate the first aerosol indirect effect (Liu and Daum, 2002; Peng and Lohmann, 2003; Rotstayn and Liu, 2003). Nevertheless, it is important to point out that there can also exist a negative relationship (a decrease of ε with N_c) (Grabowski, 1998; Daum et al., 2007; Ma et al., 2010; Xie et al., 2013[a]). Actually, some other results have indicated that there are different relationships between ε and N_c (Zhao et al., 2006; Brenguier et al., 2011). It is suggested that the change of spectral dispersion is very complex, which depends on several factors, e.g. aerosol physical and chemical properties, environmental parameters including atmospheric temperature and humidity, and dynamical effects (Khain et al., 2000; Lu and Seinfeld, 2006; Liu et al., 2006; Peng et al., 2007). In this paper, according to various $\varepsilon-N_\mathrm{c}$ relationships including positive and negative relationships (Martin et al., 1994; Grabowski, 1998; Rotstayn and Liu, 2003; Daum et al., 2007; Ma et al., 2010; Xie et al., 2013[a]), we analytically study the cloud radiative forcing of the first indirect aerosol effect to derive their corresponding variation range.

We review the various $\varepsilon-N_\mathrm{c}$ relationships and cloud optical properties including spectral dispersion, in section 2. Section 3 evaluates the cloud radiative forcing for the first indirect aerosol effect according to the different $\varepsilon-N_\mathrm{c}$ relationships. Finally, a discussion is presented and conclusions are drawn in section 4.

2. The cloud optical properties including spectral dispersion

2.1 *Relationships between spectral dispersion and cloud droplet number concentration*

The cloud droplet spectral dispersion ε can be described as the ratio of standard deviation σ and mean radius $\bar{r}_\mathrm{c}$ ($\varepsilon=\sigma/\bar{r}_\mathrm{c}$). This parameter stands for the relative dispersion of cloud droplet size distribution: a small value represents a relatively uniform distribution of the cloud droplet size, while a large value stands for a mixture of large and small cloud droplets. The parameter ε will directly affect the cloud optical properties by changing the cloud droplet effective radius.

In the following, we will display several $\varepsilon-N_\mathrm{c}$ relationships according to the different observed results. Note that the units of cloud droplet number concentration N_c are cm^{-3} in all the following formulas. According to the observations from Martin et al. (1994), the $\varepsilon-N_\mathrm{c}$ relationship has been approximated by Morrison and Grabowski (2007) (hereafter, Martin relationship)

$$\varepsilon=0.0005714N_\mathrm{c}+0.271\ . \tag{1}$$

These analytical results are from warm stratocumulus clouds with continental air masses and maritime air masses, where $\varepsilon=0.43$ for continental air masses, and $\varepsilon=0.33$ for maritime air masses.

The Rotstayn-Liu relationship is given by the formula (Rotstayn and Liu, 2003)

$$\varepsilon=1-0.7\mathrm{e}^{-\alpha N_\mathrm{c}}\ , \tag{2}$$

where the data were obtained from measurements in clean and polluted marine stratiform and shallow cumulus clouds at a variety of locations. Here $\alpha=0.001$, 0.003 and 0.008 for the three parameterizations. However, Rotstayn and Liu (2003) also pointed out that the parameterization of $\alpha=0.003$ is more reasonable, so we adopt this value in our study.

The Grabowski relationship can be written as follows(Grabowski, 1998):

$$\varepsilon=0.146-5.964\times10^{-2}\ln\left(\frac{N_\mathrm{c}}{2000}\right)\ , \tag{3}$$

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[a] Xie, X. N., X. D. Liu, and Z. G. Yue, 2013: Cloud microphysical properties and parameterization of cloud droplet effective radius from aircraft measurements: A case study. To be submitted.

which is calculated from these two relations about $\varepsilon = 0.366$ with $N_c = 50$ cm^{-3} for maritime clouds, and $\varepsilon = 0.146$ with $N_c = 2000$ cm^{-3} for continental clouds.

The Daum relationship adopts the following formula (Daum et al., 2007):

$$\varepsilon = 0.82 - 0.00134N_c\ . \tag{4}$$

Here, the corresponding data were measured from low altitude marine stratus/stratocumulus clouds over the Eastern Pacific Ocean on seven days during July 2005.

According to the observed data given by Ma et al. (2010), the $\varepsilon - N_c$ relationship can be approximated as (hereafter, Ma relationship):

$$\varepsilon = 0.694 - 0.0004231N_c\ . \tag{5}$$

The corresponding data were from aircraft measurements of stratiform clouds over North China during the period of April–May 2006.

The Xie relationship is given by the following formula (Xie et al., 2013):

$$\varepsilon = 0.579 - 7.42 \times 10^{-4}N_c + 4.2 \times 10^{-7}N_c^2\ . \tag{6}$$

The corresponding data of this formula were observed from a case study based on aircraft measurements of precipitating stratiform clouds (Yan'an, Northwest China) on 17 September 2003.

However, it should be pointed out that there exists great uncertainty in these $\varepsilon - N_c$ relationships (Fig. 1). This figure shows that the Martin relationship [Eq. (1)] and the Rotstayn-Liu relationship [Eq. (2)] have a positive relationship, while the Grabowski relationship [Eq. (3)], the Daum relationship [Eq. (4)], Ma relationship [Eq. (5)], and Xie relationship [Eq. (6)], indicate that the $\varepsilon - N_c$ relationships are negative. These different $\varepsilon - N_c$ relationships will result in distinct cloud optical properties, and then change the cloud radiative forcing induced by aerosols for the first indirect aerosol effect.

2.2 *Review of cloud optical properties including spectral dispersion*

The effective radius of cloud droplets R_e (Liu and Daum, 2002), the cloud optical depth τ (Peng and Lohmann, 2003), and the cloud albedo α (Lacis and Hansen, 1974; Mendor and Weaver, 1980; Bohren, 1987) can be described by the following forms:

$$R_e = \left(\frac{3}{4\pi\rho_w}\right)^{\frac{1}{3}} \frac{(1+2\varepsilon^2)^{\frac{2}{3}}}{(1+\varepsilon^2)^{\frac{1}{3}}} \left(\frac{L_c}{N_c}\right)^{\frac{1}{3}}\ , \tag{7}$$

$$\tau = \frac{3H_c}{2\rho_w}\frac{L_c}{R_e}\ , \tag{8}$$

$$\alpha = \frac{b(1-g)\tau}{a + b(1-g)\tau}\ , \tag{9}$$

where ρ_w is water density, N_c represents the total cloud droplet number concentration (cm^{-3}) and L_c

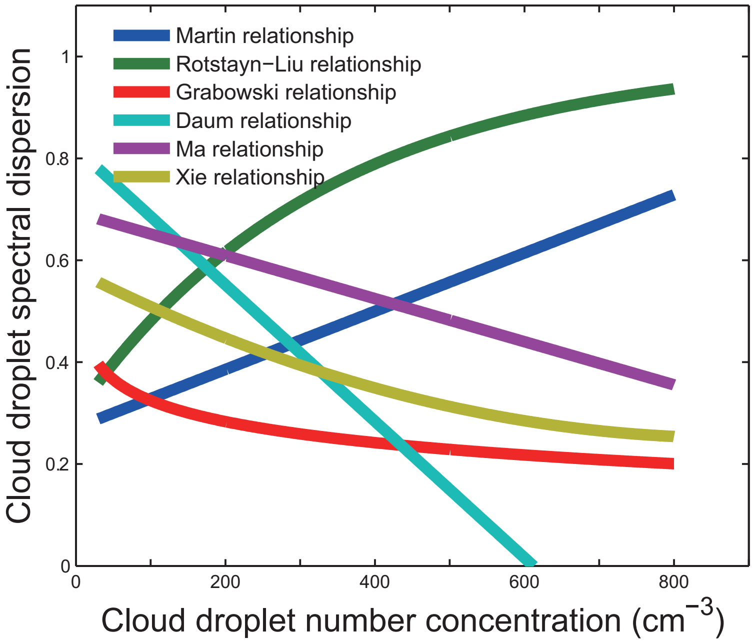

Fig. 1. Relationships between cloud droplet spectral dispersion and droplet number concentration.

represents the total cloud water content (g cm^{-3}). In Eqs. (8) and (9), H_c is the cloud thickness, and a and b are constants: $a=2$ and $b=\sqrt{3}$ (Lacis and Hansen, 1974); $a=1$ and $b=1$ (Mendor and Weaver, 1980); $a=2$ and $b=1$ (Bohren, 1987). It is noted that in Eq. (7) the cloud droplet spectral dispersion ε is the function of the cloud droplet number concentration N_c described by Eqs. (1), (2), (3), (4), (5) and (6).

If L_c and H_c constants, we calculate the derivative of the cloud properties R_e, τ, and α, then display the following expressions:

$$\frac{dR_\mathrm{e}}{dN_\mathrm{c}}=-\left[\frac{1}{3N_\mathrm{c}}-\frac{2\varepsilon(3+2\varepsilon^2)}{3(1+\varepsilon^2)(1+2\varepsilon^2)}\frac{d\varepsilon}{dN_\mathrm{c}}\right]R_\mathrm{e}\,, \quad (10)$$

$$\frac{d\tau}{dR_\mathrm{e}}=-\frac{\tau}{R_\mathrm{e}}\,, \quad (11)$$

$$\frac{d\alpha}{d\tau}=\frac{\alpha(1-\alpha)}{\tau}\,. \quad (12)$$

According to Eqs. (10), (11), and (12), we can then obtain the expression of $\frac{d\alpha}{dN_\mathrm{c}}$:

$$\begin{aligned}\frac{d\alpha}{dN_\mathrm{c}}&=\frac{d\alpha}{d\tau}\frac{d\tau}{dR_\mathrm{e}}\frac{dR_\mathrm{e}}{dN_\mathrm{c}}\\&=\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\left[1-\frac{2\varepsilon N_\mathrm{c}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\frac{d\varepsilon}{dN_\mathrm{c}}\right]. \quad (13)\end{aligned}$$

The global mean shortwave cloud radiative forcing resulting from a perturbation in the cloud albedo α, which is induced by a change of the cloud droplet number concentration N_c, can be given as the following expression (Charlson et al., 1992):

$$dF_\varepsilon=-0.2FA_\mathrm{c}d\alpha\,, \quad (14)$$

where F is the solar constant, and A_c is the cloud fraction. Using the results of Eq. (13), the formula of $\frac{dF_\varepsilon}{dN_\mathrm{c}}$ is given by:

$$\begin{aligned}\frac{dF_\varepsilon}{dN_\mathrm{c}}&=\frac{dF_\varepsilon}{d\alpha}\frac{d\alpha}{dN_\mathrm{c}}\\&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1-\frac{2\varepsilon N_\mathrm{c}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\frac{d\varepsilon}{dN_\mathrm{c}}\right]. \quad (15)\end{aligned}$$

This expression, which could be utilized to evaluate the cloud radiative forcing induced by aerosols for the first indirect aerosol effect, is basically equivalent to the result given by Daum and Liu (2001).

3. The first indirect aerosol effect based on the different $\varepsilon-N_\mathrm{c}$ relationships

Based on the different $\varepsilon-N_\mathrm{c}$ relationships, we derive the analytical expressions of the cloud radiative forcing ΔF_ε caused by changes in ΔN_c and then evaluate the first indirect aerosol effect and give their variation range. For the Martin relationship [Eq. (1)], the derivation of ε can be given by:

$$\frac{d\varepsilon}{dN_\mathrm{c}}=0.0005174\,, \quad (16)$$

and the form of $\frac{dF_\varepsilon}{dN_\mathrm{c}}$ takes the form

$$\begin{aligned}\frac{dF_\varepsilon}{dN_\mathrm{c}}&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1-\frac{0.0011428\varepsilon N_\mathrm{c}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]. \quad (17)\end{aligned}$$

Then, we can derive the change in $F_\varepsilon(\Delta F_\varepsilon)$

$$\begin{aligned}\Delta F_\varepsilon&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1-\frac{0.0011428\varepsilon N_\mathrm{c}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\Delta N_\mathrm{c}\,. \quad (18)\end{aligned}$$

The derivation of ε in the Rotstayn-Liu relationship [Eq. (2)] can be expressed as:

$$\frac{d\varepsilon}{dN_\mathrm{c}}=0.0021\mathrm{e}^{-0.003N_\mathrm{c}}\,. \quad (19)$$

We can derive $\frac{dF_\varepsilon}{dN_\mathrm{c}}$ and ΔF_ε:

$$\begin{aligned}\frac{dF_\varepsilon}{dN_\mathrm{c}}&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1-\frac{0.0042\varepsilon N_\mathrm{c}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\mathrm{e}^{-0.003N_\mathrm{c}}\right], \quad (20)\\\Delta F_\varepsilon&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1-\frac{0.0042\varepsilon N_\mathrm{c}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\mathrm{e}^{-0.003N_\mathrm{c}}\right]\Delta N_\mathrm{c}\,. \quad (21)\end{aligned}$$

For the Grabowski relationship [Eq. (3)], the derivation of ε can be given by:

$$\frac{d\varepsilon}{dN_\mathrm{c}}=-\frac{0.05964}{N_\mathrm{c}}\,. \quad (22)$$

The mathematical expressions of $\frac{dF_\varepsilon}{dN_\mathrm{c}}$ and ΔF_ε can be derived as:

$$\begin{aligned}\frac{dF_\varepsilon}{dN_\mathrm{c}}&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1+\frac{0.11928\varepsilon(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right], \quad (23)\\\Delta F_\varepsilon&=-0.2FA_\mathrm{c}\frac{\alpha(1-\alpha)}{3N_\mathrm{c}}\times\\&\quad\left[1+\frac{0.11928\varepsilon(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\Delta N_\mathrm{c}\,. \quad (24)\end{aligned}$$

The derivation of ε for the Daum relationship [Eq. (4)] is given by

$$\frac{d\varepsilon}{dN_{\mathrm{c}}}=-0.00134\ , \tag{25}$$

and we also have $\frac{dF_\varepsilon}{dN_{\mathrm{c}}}$ and ΔF_ε:

$$\frac{dF_\varepsilon}{dN_{\mathrm{c}}}=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\times\left[1+\frac{0.00268\varepsilon N_{\mathrm{c}}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\ , \tag{26}$$

$$\Delta F_\varepsilon=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\times\left[1+\frac{0.00268\varepsilon N_{\mathrm{c}}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\Delta N_{\mathrm{c}}\ . \tag{27}$$

For the Ma relationship [Eq. (5)], we can have the derivation of ε:

$$\frac{d\varepsilon}{dN_{\mathrm{c}}}=-0.0004231\ , \tag{28}$$

and the expressions of $\frac{dF_\varepsilon}{dN_{\mathrm{c}}}$ and ΔF_ε can be obtained:

$$\frac{dF_\varepsilon}{dN_{\mathrm{c}}}=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\times\left[1+\frac{0.0008462\varepsilon N_{\mathrm{c}}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\ , \tag{29}$$

$$\Delta F_\varepsilon=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\times\left[1+\frac{0.0008462\varepsilon N_{\mathrm{c}}(3+2\varepsilon^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\Delta N_{\mathrm{c}}\ . \tag{30}$$

The derivation of ε in the Xie relationship [Eq. (6)] can be expressed as:

$$\frac{d\varepsilon}{dN_{\mathrm{c}}}=-7.42\times10^{-4}+8.4\times10^{-7}N_{\mathrm{c}}\ , \tag{31}$$

and the expressions of $\frac{dF_\varepsilon}{dN_{\mathrm{c}}}$ and ΔF_ε can be obtained:

$$\frac{dF_\varepsilon}{dN_{\mathrm{c}}}=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\left[1+\frac{2\varepsilon(3+2\varepsilon^2)(7.42\times10^{-4}N_{\mathrm{c}}-8.4\times10^{-7}N_{\mathrm{c}}^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\ , \tag{32}$$

$$\Delta F_\varepsilon=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\left[1+\frac{2\varepsilon(3+2\varepsilon^2)(7.42\times10^{-4}N_{\mathrm{c}}-8.4\times10^{-7}N_{\mathrm{c}}^2)}{(1+\varepsilon^2)(1+2\varepsilon^2)}\right]\Delta N_{\mathrm{c}}\ . \tag{33}$$

When $\varepsilon=0$, we can also obtain the following form of $\frac{dF_0}{dN_{\mathrm{c}}}$ and ΔF_0:

$$\frac{dF_0}{dN_{\mathrm{c}}}=\frac{dF_0}{d\alpha}\frac{d\alpha}{dN_{\mathrm{c}}}=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\ , \tag{34}$$

$$\Delta F_0=-0.2FA_{\mathrm{c}}\frac{\alpha(1-\alpha)}{3N_{\mathrm{c}}}\Delta N_{\mathrm{c}}\ . \tag{35}$$

In all the analytical expressions of ΔF_ε [Eqs. (18), (21), (24), (27), (30) and (33)], as well as ΔF_0 [Eq. (35)], the properties A_{c}, α, F are set to the same values ($A_{\mathrm{c}}=0.3$, $\alpha=0.5$, and $F=1370$ W m^{-2}) (Charlson et al., 1992), and we also choose $N_{\mathrm{c}}=100$ cm^{-3} and $\Delta N_{\mathrm{c}}=15$ cm^{-3}. Hence, the indirect aerosol radiative forcing ΔF_ε for the different $\varepsilon-N_{\mathrm{c}}$ relationships, the first indirect aerosol radiative forcing without considering the spectral dispersion ΔF_0, and the corresponding variations $(\Delta F_\varepsilon-\Delta F_0)/\Delta F_0$ are evaluated in Table 1. It shows that the value of the first indirect aerosol radiative forcing without considering the spectral dispersion is -1.03 W m^{-2}. The cloud radiative forcing induced by aerosols for these different $\varepsilon-N_{\mathrm{c}}$ relationships varies from -0.73 W m^{-2} (Roststayn-Liu relationship) to -1.29 W m^{-2} (Daum relationship). Their corresponding variations are -29.1% and 25.2%, compared to the case setting the cloud droplet spectral dispersion ε to zero.

4. Discussion and conclusions

In this paper, we have analytically derived the mathematical expressions of the cloud radiative forc-

Table 1. The first indirect aerosol radiative forcing (ΔF_ε) for the different $\varepsilon-N_{\mathrm{c}}$ relationships, the first indirect aerosol radiative forcing without considering the spectral dispersion (ΔF_0), and the corresponding variations [$(\Delta F_\varepsilon-\Delta F_0)/\Delta F_0$)].

$\varepsilon-N_{\mathrm{c}}$ relationships	ΔF_0 (W m^{-2})	ΔF_ε (W m^{-2})	$(\Delta F_\varepsilon-\Delta F_0)/\Delta F_0)$
Martin relationship	−1.03	−0.94	−8.7%
Rotstayn-Liu relationship	−1.03	−0.73	−29.1%
Grabowski Relationship	−1.03	−1.13	9.7%
Daum relationship	−1.03	−1.29	25.2%
Ma relationship	−1.03	−1.11	7.8%
Xie relationship	−1.03	−1.16	12.6%

ing induced by a change of N_c, according to the six observed $\varepsilon - N_c$ relationships including the Martin, Roststayn-Liu, Grabowski, Daum, Ma, and Xie relationships. The results showed that the cloud radiative forcing induced by aerosols for these different $\varepsilon - N_c$ relationships varies from -29.1% (Roststayn-Liu relationship) to 25.2% (Daum relationship), compared to the case setting cloud droplet spectral dispersion ε to zero. It is noted that the factors that determine $\varepsilon - N_c$ relationships are still poorly understood, and that exact quantification of the spectral dispersion influence on the first aerosol indirect effect is also in its infancy (Zhao et al., 2006; Brenguier et al., 2011). Our results suggest that further understanding of $\varepsilon - N_c$ relationships may reduce the uncertainty of the first indirect aerosol effect and enhance comprehension of aerosol-cloud-radiation interactions.

Nevertheless, it should be pointed out that we did not consider all aerosol-induced variations in cloud properties in the current analysis, e.g. the cloud albedo, cloud fraction, the total cloud water content and cloud depth. Actually, these cloud properties can be changed with an increase in aerosol concentrations because more activation of aerosols forms more cloud droplets (Fan et al., 2010; Xie et al., 2011). Hence, additional simulation studies with modern radiative transfer models are necessary to further evaluate the first indirect aerosol effect considering cloud droplet spectral dispersion.

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