What is the effect of cloud inhomogeneities on actinic fluxes and chemical species concentrations?

Christel Bouet, Frédéric Szczap, Maud Leriche, Albert Benassi

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Christel Bouet, Frédéric Szczap, Maud Leriche, and Albert Benassi

Laboratoire de Météorologie Physique, Centre National de la Recherche Scientifique, Université Blaise Pascal, Aubière, France

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[1] The purpose of the present paper is to evaluate the bias introduced by the commonly used homogeneous plane-parallel cloud hypothesis in the computation of actinic fluxes, photolysis coefficients, and main tropospheric species concentrations. Accordingly, these quantities obtained for an inhomogeneous cloud field generated with a stochastic model are compared to their homogeneous plane-parallel equivalent. Results show that neglecting cloud inhomogeneities has a significant impact on actinic flux. For instance, the bias introduced by the homogeneous plane-parallel cloud hypothesis can reach more than 100% below the cloud leading to a comparable bias in photolysis coefficients and, in turn, creating an enhancement of the oxidizing capacity of the system. **Citation:** Bouet, C., F. Szczap, M. Leriche, and A. Benassi (2006), What is the effect of cloud inhomogeneities on actinic fluxes and chemical species concentrations?, *Geophys. Res. Lett.*, *33*, L01818, doi:10.1029/2005GL024727.

1. Introduction

[2] Small changes in the cloud-radiative forcing (which is the effects of clouds on the radiation balance of the Earth fields) can play a significant role as a climate feedback mechanism [*Ramanathan et al.*, 1989]. Observations show that clouds are far from being homogeneous revealing autosimilar and fractal statistical properties [*Lovejoy*, 1982; *Cahalan and Snider*, 1989; *Davis et al.*, 1999]. They also exhibit numerous structures in their shapes, aspect ratio and cloud coverage as well as a high horizontal and vertical variability in their microphysical and optical properties [*Stephens and Platt*, 1987; *Korolev et al.*, 2001]. Numerous studies were conducted to estimate the radiative impact of cloud inhomogeneities in order to improve cloud properties retrieval from various radiometers and to better estimate the role of clouds on Earth's climate [*Harshvardhan*, 1982; *Welch and Wielicki*, 1984, 1985, 1989; *Barker et al.*, 1998, 1999]. *Barker et al.* [1999] showed that it was crucial to consider horizontal fluctuations of cloud extinction in order not to underestimate surface absorption and not to overestimate reflected fluxes.

[3] Daytime atmospheric chemistry is driven by photolysis processes. Due to their impact on radiation, clouds can disrupt these processes. Photolysis rate coefficients are derived from actinic flux [*Madronich*, 1987] and numerous studies have already demonstrated the impact of clouds on actinic flux [*Madronich*, 1987; *Junkermann et al.*, 2002; *Kanaya et al.*, 2003; *Monks et al.*, 2004]. For example, *Madronich* [1987] showed that actinic fluxes inside the clouds can frequently exceed the clear-sky values (up to 5 times greater). However, all of these studies as well as those concerning the impact of clouds on photolysis coefficients [*Van Weele and Duynkerke*, 1993; *Crawford et al.*, 1999, 2003; *Früh et al.*, 2000; *Tie et al.*, 2003], considered clouds as a single homogeneous, plane-parallel layer. A recent study of *Stockwell and Goliff* [2004] showed that NO_2 photolysis coefficients derived from actinic fluxes simulated with the TUV model (Tropospheric Ultraviolet Visible model [*Madronich and Flocke*, 1998]) using the delta-Eddington method [*Joseph et al.*, 1976] could be up to 56% greater than photolysis coefficients derived from measured actinic fluxes. However, a few studies have already investigated the three-dimensional (3D) effects of inhomogeneous clouds on actinic fluxes and photolysis coefficients [*Los et al.*, 1997; *Trautmann et al.*, 1999; *Várnai and Davis*, 1999; *Brasseur et al.*, 2002; *Trentmann et al.*, 2003]. *Brasseur et al.* [2002] demonstrated that, in presence of a deep convective cloud, actinic fluxes can be increased by a factor of 2 to 5 compared to clear-sky values, leading to changes in ozone production rates (+15%) and enhancement in OH concentrations (120–200%) in the upper troposphere. They also showed that photolysis coefficients are inhomogeneously distributed throughout the cloud.

[4] Since numerical chemistry transport models usually use the homogeneous plane-parallel cloud assumption, it is critical to investigate the bias introduced in tropospheric chemistry compared to inhomogeneous clouds, which are closer to real clouds. The aims of this numerical modeling study are firstly to quantify the effects of cloud optical and geometrical inhomogeneities on actinic flux in order to estimate, in a second step, the impact of these cloud inhomogeneities on tropospheric photochemistry.

2. Methodology

[5] A two-dimensional (2D) inhomogeneous cumulus cloud field is generated with the stochastic tdMAP model (tree driven Mass Accumulation Process [*Benassi et al.*, 2004]), which is able to provide stratocumulus and cumulus cloud fields with properties close to those observed in real clouds. In our case, the simulated 2D cloud field represents a horizontal extent of 12.8×12.8 km^2 (256×256 pixels) with a 58 % cloud cover. Each 50×50 m^2 cloud pixel has the same vertical extent and is assumed to be vertically homogeneous leading to a 3D cloud field. The cloud layer is located between 1 and 1.5 km high. The SHDOM model (Spherical Harmonics Discrete Ordinate Method [*Evans*,

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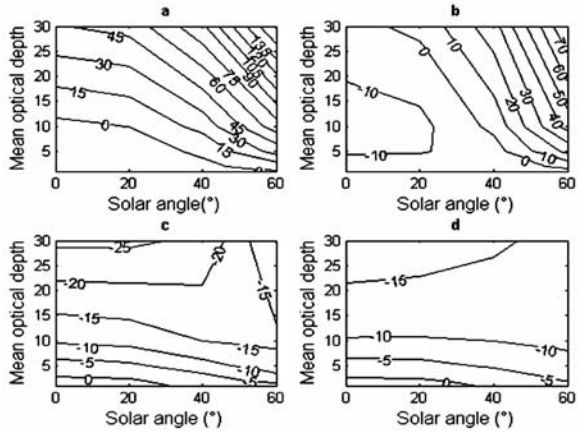

Figure 1. ΔF as a function of solar angle and mean optical depth below the cloud at z = 200 m (a), in the cloud at z = 1050 m (b) and 1450m (c), and above the cloud at z = 2 km (d).

1998]) is used to solve the radiative transfer equation (RTE) in the inhomogeneous 3D cloud field. Thereby, actinic fluxes at a wavelength of 400 nm corresponding to the wavelength of maximum dissociation of NO_2 are computed for several cloud mean optical depths (1, 2, 5, 10, and 30) and for different solar zenith angles (0, 20, 40, and 60°). The RTE is also solved using SHDOM for the homogeneous plane-parallel equivalent clouds in order to estimate the bias introduced by the plane-parallel homogeneous cloud approximation. For all cases, droplets are assumed to follow a lognormal distribution with an effective radius of 10 μm and a standard deviation of 0.35 μm. Optical parameters are calculated using Mie theory. Then, the actinic fluxes computed by SHDOM for the inhomogeneous case are averaged over the whole cloud field. These averaged fluxes and the ones computed for the plane-parallel case are parameterized as a function of the solar zenith angle and the wavelength in order to estimate photolysis coefficients with the TUV model [*Madronich and Flocke*, 1998] for the main tropospheric chemical species. Photolysis coefficients are computed only for the cloud of optical depth 10 at location (51°N; 0°E). Finally, the M2C2 box model (Model of Multiphase Cloud Chemistry [*Leriche et al.*, 2003]) is used to simulate the multiphase cloud chemistry for two different scenarios (remote and urban) from *Ervens et al.* [2003]. These scenarios include emissions and dry deposition with variable photolysis for three days in order to assess the bias introduced in tropospheric photochemistry by the homogeneous plane-parallel assumption.

3. Results and Discussion

3.1. Effect of 2D Inhomogeneities on Actinic Fluxes

[6] The bias between actinic fluxes calculated with SHDOM in the inhomogeneous ($F_{SHDOM3D}$) and homogeneous plane-parallel ($F_{SHDOMPP}$) cases is computed as:

$$\Delta F = \frac{F_{SHDOM3D} - F_{SHDOMPP}}{F_{SHDOMPP}} \times 100 \quad (1)$$

This bias is shown on Figure 1 as a function of mean cloud optical depth and solar zenith angle below the cloud at z = 200 m, in the cloud at z = 1050 m and 1450 m, and above the cloud at z = 2 km. Cloud inhomogeneities significantly enhance the actinic flux below the cloud and in its lower part whereas they slightly decrease it above. For instance, for an optical depth of 10, which is a typical value for stratocumulus clouds, and a solar zenith angle of 60°, below the cloud, ΔF is about 70% whereas above, it is around −10%. Figure 1 also points out that $|\Delta F|$ increases with both solar zenith angle and cloud optical depth. Indeed, the greater cloud optical depth or solar zenith angle is, the more important multiple scattering from the cloud sides is, which leads to an enhancement in actinic fluxes. Finally, this means that the closer the sun is to the zenith, the more the inhomogeneous cloud becomes closer to the homogeneous one.

[7] Considering the cloud effect toward clear sky onto actinic fluxes as shown for instance by *Madronich* [1987], our results imply that the assumption of the homogeneous plane-parallel cloud overestimates this negative effect.

3.2. Effect of 2D Inhomogeneities on Photolysis Coefficients

[8] Prior to the computing of photolysis coefficients with the TUV model, actinic fluxes at 400 nm need to be extended to the visible spectrum. The parameterization used is based upon the statement that, as the mean free path, defined by the mean distance between two collisions undergone by the photon, is constant in our simulations, the effects of cloud inhomogeneities are assumed to be constant with wavelength.

[9] The bias between photolysis coefficients for inhomogeneous (J_{3D}) and homogeneous (J_{PP}) cases is defined in the same way as for the actinic fluxes:

$$\Delta J = \frac{J_{3D} - J_{PP}}{J_{PP}} \times 100 \quad (2)$$

ΔJ is the same for a given hour for all the photolysis reactions because the bias in photolysis coefficients is directly related to the ones on actinic fluxes by definition. Figure 2 presents this bias below the cloud, at z = 200 m, where the bias computed on actinic fluxes is the highest, as a function of time and solar zenith angle. As ΔF, ΔJ is minimum at noon for a solar zenith angle of 27° with a value around 5%. Thus, the impact of the bias introduced by the homogeneous plane-parallel assumption on the photochemistry varies over time. At noon, when photochemistry is the most active, the impact will be the lowest. However, the most important emissions of pollutants occur in the morning around 8 a.m. where the bias is around 50% and where some important photolysis reactions take place, such as the photolysis of nitrous acid which is the largest source of OH radicals in the morning [*Lammel and Cape*, 1996].

3.3. Impact on Tropospheric Trace Gases Concentrations

[10] As the first day of simulation is the time for the chemical system to reach the equilibrium, results are presented averaged over days 2 and 3.

[11] In the same way as for the actinic flux, the bias between concentrations for inhomogeneous (C_{3D}) and ho-

mogeneous (C_{PP}) cases below the cloud, at z = 200 m, is defined as:

$$\Delta C = \frac{C_{3D} - C_{PP}}{C_{PP}} \times 100 \quad (3)$$

Table 1 shows this bias averaged over days 2 and 3 for main gas phase tropospheric species in the morning, at noon and in the afternoon for the two chemical scenarios, remote and urban. The bias introduced by the use of the homogeneous plane-parallel approximation is different from one species to another: either positive or negative, it is high for radicals (between 5.4 and 54%) whereas it is negligible for methane (less than 0.5%). Neglecting cloud inhomogeneities will have a significant impact on tropospheric chemistry because radicals drive tropospheric chemistry; for instance, ozone concentrations are underestimated using the homogeneous plane-parallel assumption. The stronger photolysis for the 3D case (cf. Figure 2) and the positive bias on OH radicals lead to a larger oxidizing capacity of the atmosphere. Moreover, the minimum values of the positive bias on radicals concentrations is at noon when the bias on actinic fluxes is the lowest. For methane, the bias is close to zero because of its low reactivity driven by its slow oxidation by OH radicals. For NO_2, the bias is negative due to a more efficient photolysis in the 3D case while for NO, the bias is negative in the morning and at noon and positive in the afternoon due to the combined effects of the continuous NO emission, the NO_2 photolysis and the NO to NO_2 conversion by RO_2 radicals. For organic compounds, the behavior is more complicated with either positive or negative impact depending on the time of the day and on chemical species considered underlying the non-linearity of the tropospheric chemistry system.

Table 1. ΔC (in %), Averaged Over Days 2 and 3 of the Simulation, for Main Tropospheric Species in Gaseous Phase at 8 a.m., Noon and 4 p.m. for the Two Cases, Remote and Urban

	Remote			Urban		
Species	8 a.m.	Noon	4 p.m.	8 a.m.	Noon	4 p.m.
O_3	11	7.5	5.8	13	9.2	7.6
OH	42	8.3	50	46	9.7	54
HO_2	28	5.4	23	32	7.5	26
H_2O_2	13	11	7.3	19	17	13
NO	−13	−8.2	12	−7.4	−9.3	14
NO_2	−31	−6.0	−15	−27	−6.1	−15
HNO_2	−16	−5.7	9.8	−6.1	−5.8	14
HNO_3	6.4	−3.3	−1.4	6.9	−2.2	1.1
CH_4	−0.2	−0.3	−0.3	−0.2	−0.2	−0.3
HCHO	0.4	1.3	0.3	−10	−1.4	−2.2
CH_3OOH	16	16	8.9	8.8	21	13
CH_3O_2	51	16	32	48	19	35

4. Conclusions

[12] The aim of this study was to quantify the bias introduced by the commonly used homogeneous plane-parallel cloud hypothesis on actinic flux, photolysis coefficients, and tropospheric chemistry. It was shown that cloud inhomogeneities have a significant impact on actinic flux: for example, for a solar zenith angle of 60° and an optical cloud depth of 10, differences greater than 100% are found. It was also shown that the bias introduced neglecting cloud inhomogeneities depends on both solar zenith angle and cloud optical depth. The same behavior was found for photolysis coefficients. However, due to the non-linearity of tropospheric chemistry system, the bias on chemical species concentrations showed a more complex behavior with either positive or negative bias depending on the chemical species considered and on the time of the day. Finally, for the remote and urban chemical scenarios used, the effect of the cloud inhomogeneities on tropospheric chemistry in comparison with homogeneous clouds is to enhance the oxidizing capacity of the system by higher concentrations of radicals and stronger photolysis.

[13] The next step of this study will be to parameterize the effects due to cloud inhomogeneities in radiative transfer models in order to compare the photolysis coefficients obtained with measurements.

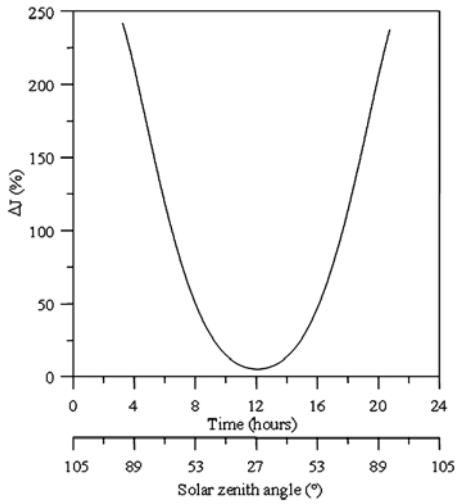

Figure 2. ΔJ as a function of solar zenith angle and time below the cloud, at z = 200 m.

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A. Benassi, C. Bouet, M. Leriche, and F. Szczap, Laboratoire de Météorologie Physique, Centre National de la Recherche Scientifique, Université Blaise Pascal, 24 avenue des Landais, F-63177 Aubière Cedex, France. (c.bouet@opgc.univ-bpclermont.fr)