

Analysis of aerosol-cloud interaction from multi-sensor satellite observations

Lorenzo Costantino¹ and François-Marie Bréon¹

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[1] Aerosol interaction with clouds is the main uncertainty for the quantification of the anthropogenic forcing on climate. The first step of the so-called “aerosol indirect effect” is the change of cloud droplet size distribution when seeded by anthropogenic aerosols. Satellite data provide the density and diversity of observations needed for a statistical estimate of this effect. Numerous such studies have demonstrated the correlation between aerosol load and Cloud Droplet Radius (CDR) and a few have quantified the impact of aerosol on the microphysics. Here, we go one step further by using the profiles from the spaceborne CALIPSO lidar that indicates the respective position of aerosol and cloud layers. The results show that, when aerosol and cloud layers are clearly separated, there is no correlation between aerosol load and CDR. On the other hand, when the lidar profile indicates mixing, there is a strong correlation. We focus on the stratocumulus cloud fields off the coast of Namibia and Angola that are seeded by biomass burning aerosols from Africa. The log-log slope of CDR and a proxy of the condensation nuclei number are -0.24 in excellent agreement with theoretical estimate. When the vertical profile information is not used, the slope is significantly smaller. **Citation:** Costantino, L., and F.-M. Bréon (2010), Analysis of aerosol-cloud interaction from multi-sensor satellite observations, *Geophys. Res. Lett.*, 37, L11801, doi:10.1029/2009GL041828.

1. Introduction

[2] Aerosol interaction with clouds is the primary uncertainty on anthropogenic forcing [Intergovernmental Panel on Climate Change, 2007]. Submicronic aerosols act as Cloud Condensation Nuclei and change the cloud droplet size distribution (smaller droplets in a polluted atmosphere than for a clean one), which tends to increase the cloud albedo, assuming a constant liquid water content. Such aerosol cloud interaction was first theorized by Twomey [1977] and it is generally called *first aerosol indirect effect* or more simply the *Twomey effect*. Previous studies predict a linear relationship between the logarithm of cloud droplet size and the logarithm of aerosol index. Under the condition of a constant liquid water path, assuming that the number of cloud droplets is equal to the number of particles that may act as CCN (Cloud Condensation Nuclei) elevated to the power of 0.7, the slope of the linear regression is -0.23 [Bréon *et al.*, 2002; Feingold *et al.*, 2003]. With such hypothesis, the size distribution change increases the cloud albedo so that the aerosols have a

net cooling effect. In addition, the change in size distribution may impact the cloud precipitation and life cycle. Because many other factors act on the cloud life cycle, a quantitative estimate of the aerosol impact may be possible in a statistical sense only, which implies the use of a great number of data, which are uniquely provided by satellite retrievals [Nakajima *et al.*, 2001; Bréon *et al.*, 2002; Sekiguchi *et al.*, 2003; Kaufman *et al.*, 2005; Quaas *et al.*, 2006; Storelvmo *et al.*, 2006; Quaas *et al.*, 2009]. For instance, using satellite data acquired by the POLarization and Directionality of the Earth Reflectance (POLDER) instrument Bréon *et al.* [2002] analysed the relationship between CDR and the aerosol optical depth. They did find a negative relationship, demonstrating the impact of fine-scale aerosol on cloud microphysics. A quantitative relationship between aerosol optical depth and CDR was found. One uncertainty however lies in the relative altitude levels of aerosols and clouds.

[3] Even when aerosol and cloud layers are detected, they may not be mixed. Such situations may therefore bias the statistical slope. In this paper, we use the information provided by the spaceborne lidar on board the CALIPSO (Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite. Its products provide an indication of the respective height of aerosols and clouds. From this information, it is then possible to distinguish cases when aerosols and clouds are mixed from those that are clearly separated. We can therefore provide a statistical estimate of the aerosol impact on cloud microphysics, albeit limited to the “mixed case”. We focus our analysis on the Atlantic Ocean, off the coasts of Angola and Namibia. During the period from July to September, this area is under the influence of biomass burning aerosols that are advected from the African continent by persistent easterly flows (trade winds). In addition, the area is often covered by low-level stratocumulus clouds. In many cases, the aerosols are above the cloud layer, resulting in a large positive radiative forcing, due to the combination of a large aerosol absorption and a high albedo of the cloud deck beneath the aerosol layer [Hansen *et al.*, 1997; Penner *et al.*, 1998; Chand *et al.*, 2009].

2. Data

[4] The present work is based on retrievals of cloud droplet radii (CDR), aerosol index (AI) and vertical profiles of aerosols and clouds. The region of interest (4N, 30S; 14W, 18E) is mostly over the Atlantic Ocean, characterized by a persistent cover of low clouds and a strong concentration of aerosol originating from fires from Central and Southern Africa [Ichoku *et al.*, 2003]. Figure 1 shows the study area (inside the yellow square) with the active fires for July 1st, 2007. Fires are detected by the MODIS (Moderate Resolution Imaging Spectroradiometer) instrument; collected data are available

¹Laboratoire des Sciences du Climat et de l'Environnement, Unité Mixte de Recherche, UVSQ, CEA, CNRS, Gif-sur-Yvette, France.

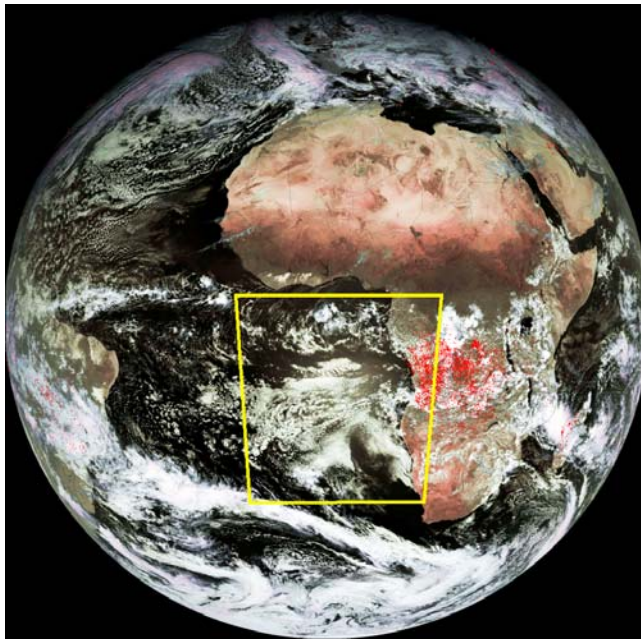


Figure 1. Meteosat-7 full disc image realized through the combination of two sensors, the broad-band visible high-resolution channel (HRV) and the far-IR channel centred on $10.8\mu\text{m}$. The visible channel has been histogram equalised and sharpened and displayed as brightness. The thermal channel has been re-sampled to the same resolution as the visible channel, converted into temperature, and used to supply the hue of the colour component of the output image. Overlaid on the original image the yellow square represents our study area, and the red spots represent all active fires seen from MODIS for July 1st 2006. Each active fire location represents the center of a 1km pixel (approximately) flagged as containing one or more actively burning fires within that pixel.

on the FIRMS (Fire Information for Resource Management System) project at <ftp://mapsftp.geog.umd.edu>. During the dry season that peaks between July and September, biomass burning injects large quantities of aerosols to the atmosphere. For the most part, aerosols remain within the boundary layer, which is several km thick as shown by lidar profiles [Labonne *et al.*, 2007]. As the aerosols are transported over the oceans by easterly trade winds, their altitude remains fairly constant. As a consequence, and because the mixing layer is much thinner over the ocean, the aerosols pass into the free troposphere and are therefore located above the stratocumulus clouds that lie at the top of the boundary layer. This process is well illustrated in Figure 2, which is a profile at 532 nm of the CALIPSO lidar for the night of 9 September 2009. The inset in Figure 2 in the upper right corner shows the location of the study area together with the CALIPSO footprint. For the vertical profile, a color scale is associated with the attenuated backscatter signal magnitude. Purple and blue colors are associated with low backscatter values, indicative of a clean atmosphere (molecular return). Light green corresponds to intermediate values and corresponds in Figure 2 to the aerosol layer. Because the aerosol layer optical depth is moderate (<1), the CALIPSO lidar can sense other scattering layers that are below the aerosol. Figure 2 clearly shows the presence of a cloud whose backscatter signal is much larger than that of the

aerosol layer. The cloud top is at an altitude of approximately 1 km. This cloud layer is dense and results in high backscatter levels, shown in red in Figure 2. Because the cloud optical depth is large, the lidar signal cannot fully penetrate the cloud, so that the backscatter profile does not contain useful information below the cloud upper layer. The yellow line in Figure 2 represents the boundary layer altitude according to ECMWF data (available with a 3-hourly and 25 km resolution at http://data-portal.ecmwf.int/data/d/interim_daily). This altitude appears in good agreement with the location of the stratocumulus cloud tops, even though the ECMWF diagnostic is some time under estimated. The black line is the ground level altitude, which is constant and equal to zero for the ocean (middle and right side of Figure 2). In the left side of Figure 2, approximately between (3.3S; 12.3E) and (10.0S; 10.9E) and an altitude of 1.5km, a clear molecular signal denotes a vertical gap of clean atmosphere between aerosol and cloud layers. On the other hand, the right side of Figure 2 represents a typical situation where aerosols appear somewhat mixed with the cloud layer, with a potential alteration of its microphysics and lifetime.

[5] In our study, we focus on the time window from June 2006 to December 2008. Because we use passive solar reflectance observations, the data are limited to daytime. The data described below are from three satellites of the A-Train constellation and are available at the ICARE center at <http://www.icare.univ-lille1.fr>.

2.1. Cloud Droplet Radius (PARASOL)

[6] Estimates of CDR are derived from the polarized measurements of PARASOL (Polarization & Anisotropy of Reflectances for Atmospheric Sciences coupled with Observations from a Lidar). This satellite was launched in December 2004 and carries the imaging radiometer/polarimeter POLDER (Polarization and Directionality of the Earth's Reflectances) which provides systematic measurements of spectral, directional and polarized properties of the reflected solar radiation, in coincidence with MODIS and CALIPSO. The CDR estimate is based on the directional signature of the polarized reflectance. Indeed, Mie computations and radiative transfer simulations show that the scattering angle for the maxima and minima of the polarized reflectance are tightly related to the cloud droplet effective radius [Bréon and Doutriaux-Boucher, 2005]. One limitation however is that the measurement is only sensitive to the very cloud top (i.e., an optical depth of ≈ 1). In addition, the estimate is only possible for fairly homogeneous cloud fields and can be retrieved at a spatial resolution of about 100 km.

2.2. Aerosol Load (MODIS)

[7] Measures of the aerosol load are provided by MODIS on board the Aqua satellite, launched in May 2002. The instrument has a wide swath of 2330 km along the orbital path, so that most of the globe is observed at least once daily, and seven wavelength bands that can be used for aerosol retrievals. For this study, we make use of MODIS Level 2 aerosol product files (MYD04_L2), which provide products corresponding to 5 minutes of observation (called *granules*) with 10 km spatial resolution. The aerosol optical depth at $0.55\mu\text{m}$ and the $0.55/0.867\mu\text{m}$ Angstrom exponent measurements are taken from the HDF Scientific Data Set *Effective_Optical_Depth_Average_Ocean_Mean* and

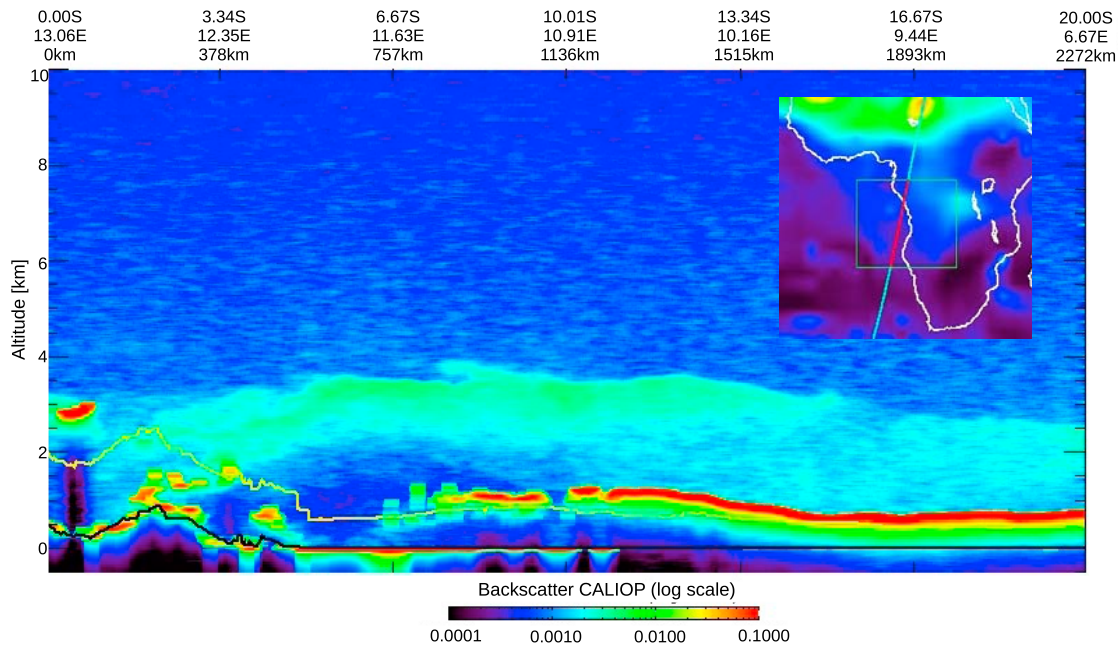


Figure 2. Profile of 532 nm backscatter signal ($\text{st}^{-1}\text{km}^{-1}$) from CALIPSO lidar Level-1 Scientific Data Set, showing the vertical distribution of aerosol and clouds for July 9th 2009 (night time), together with ECMWF boundary layer top height (yellow line). The black line represents the ground level, blue/green colors are associated with tenuous backscattering (aerosol layers) and strong backscattering is shown in red (cloud layers). The color shading of the inlay figure represents the total aerosol optical depth with the location of the CALIPSO footprint (purple/blue line), given by a back trajectory model.

Angstrom_Exponent_1_Ocean_Mean. Rather than the aerosol load, our analysis uses the so-called aerosol index, given as the product $\tau\alpha$, where τ is the aerosol optical depth and α is the Angstrom exponent. The objective is to give more weight to the fine mode aerosols that, for a given optical depth, provide the largest fraction of the condensation nuclei number concentration. Under certain assumption AI is representative of the aerosol column number [Nakajima *et al.*, 2001].

[8] MODIS is not able to provide aerosol load estimates in the presence of clouds. However, in case of broken cloudiness, its high spatial resolution allows the spectroradiometer to estimate the Aerosol index values from the measurements acquired between the clouds.

2.3. Vertical Profiles (CALIPSO)

[9] The CALIPSO satellite flies behind the Aqua satellite and carries the first lidar optimized for aerosol and cloud measurement from space, the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). It was launched in April 2006 and provides atmospheric measurements that are near-coincident with observations from MODIS and PARASOL (along track separation less than two minutes) [Winker *et al.*, 2007]. In the following, we make use of Level-2 HDF Scientific Data Set parameters *Number_Layers_Found*, *Layer_Top_Altitude*, and *Layer_Base_Altitude*, with a spatial resolution of 5 km for both aerosol and cloud products. The lidar is the only instrument that provides a reliable information about the vertical structure of the scattering layers and is used here to distinguish between cases with separated layers and those that appear to be mixed and interacting.

[10] There is an optical depth estimate within the CALIPSO Level-2 product. However, our first analysis indicated that this

parameter is very noisy and less reliable than the equivalent parameter from MODIS. Another concern with CALIPSO products is that there remain some mis-classifications between aerosols and clouds. However, these events appear rare enough to have a negligible impact on our statistical analysis.

3. Method

[11] The data analysis requires near-simultaneous measurements of AI, CDR and cloud and aerosol layer altitude. For each CALIPSO atmospheric layer retrieval, we searched for MODIS and PARASOL measurements within a 150 km radius. The temporal coincidence is ensured by the coordinated orbits of the three satellites. The optical depth estimate from MODIS is available only in clear sky, and is therefore necessarily away from our target of interest (presence of aerosol and cloud). The aerosol and cloud layers are considered well separated if the distance between the bottom of the higher layer and the top of the lower one is greater than 250 meters. On the other hand, when this vertical distance is less than 250 metres, it is assumed that the aerosol and cloud layers are interacting. The distance criteria are applied only on the highest detected cloud layer, since PARASOL CDR retrieval is only sensitive to the upper layer of the highest cloud. All lower layers are ignored in case of multilayer clouds. Similarly, for multi aerosol layers above the cloud, we consider the lower one.

[12] Finally, it should be noted that MODIS is able to detect several kinds of aerosol particles with different Angstrom exponents. Since our work focuses on the fine aerosol (in particular biomass burning) effects on cloud microphysics, we selected only cases when fine particles have been detected

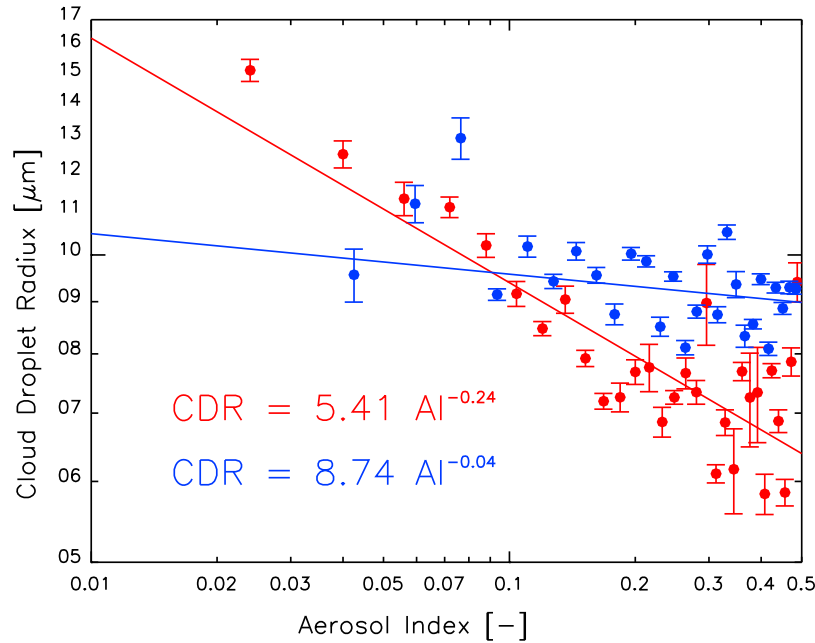


Figure 3. Statistical relationship of the Aerosol Index and the Cloud Droplet Radius for mixed (red) and non mixed (blue) cases. The error bars represent $\sigma/(n-2)^{1/2}$, where n is the number of CDR measurements within the bin and σ is their standard deviation. It represents the confidence level of the mean values if one assumes independent data.

by MODIS, i.e., those with an Angstrom exponent ranging from 1.0 to 1.5.

4. Results

[13] The lidar profile product, with near-simultaneous measurements of aerosol index and cloud droplet size, results in different data sets for two distinct atmospheric situations: mixed and well separated cloud aerosol layers. Figure 3 shows the mean value of the CDR for various bins of aerosol index. The total number of valid coincidences (i.e., with a valid atmospheric profile, an aerosol load estimate and a CDR) is 8942. Among these, a large majority (7438 or 83%) have a vertical profile where the aerosol and cloud layers are well separated according to the CALIPSO product. The corresponding datapoints are shown in blue in Figure 3. For such case, there is no significant variation of the CDR with the AI and the typical CDR is around $10 \mu\text{m}$. The observations confirm the expected behaviour with limited correlation between the two parameters.

[14] The statistics are very different for the mixed case shown in red in Figure 3. There are 1504 (17%) such cases in our dataset of CALIPSO-MODIS-PARASOL coincidences above the study of interest. On average, the CDR decreases from a maximum value of approximately $15 \mu\text{m}$ for a clean atmosphere ($\text{AI} \sim 0.02$) to a constant value between 6 and $8 \mu\text{m}$ when the aerosol index is larger than ≈ 0.3 , in good agreement with Peng *et al.* [2002].

[15] The strength of the aerosol load - cloud droplet size relationship can be quantified as the slope of the linear regression between the logarithm of CDR and AI. The computed regressions, accounting for the statistical uncertainties in the CDR means, leads to a slope of -0.04 for the unmixed cases and -0.24 for the mixed cases. For the latter case, the regression indicates a mean relationship as $\text{CDR} = 5.41 \times \text{AI}^{-0.24}$. The regression slopes confirm the observation

that the relationship is much stronger when aerosol and clouds are mixed according to CALIPSO. In addition, it is remarkable that the slope retrieved for the mixed case (-0.24) is very close to its theoretical estimate of -0.23 .

5. Conclusion

[16] In order to investigate the effects of fine aerosols on cloud microphysics, a multisensor analysis of the atmosphere has been performed. The selected area extends over the Atlantic Ocean off the coasts of Central and Southern Africa, a region frequently affected by smoke from biomass burning in Africa. Aerosol particles are injected in the free troposphere and transported by easterly flows over the ocean, where extended low shallow clouds are often present. In this context a cloud aerosol coexistence is very frequent.

[17] With the use of near-simultaneous measurements from CALIPSO, MODIS and PARASOL, it has been shown that cases of separated aerosol and cloud layers do not present any specific relationship between CDR and AI. On the other hand, when cloud and aerosol are well mixed, the Twomey effect is clearly detectable.

[18] The result of this study confirms that aerosols have a strong impact on the cloud microphysics. A quantitative analysis of the relationship between aerosol load and the impact on cloud should account for the respective position of the aerosol and cloud layer, which is now possible thanks to the CALIPSO lidar measurements. The assumption for a negative CDR - AI correlation due to the Twomey effect has been widely discussed in the literature under the condition of constant cloud liquid water path. Note however, that the present work does not take into account this condition, so that further studies should address this issue.

[19] Similar to other studies, our study has not demonstrated the impact on the cloud albedo (first indirect effect). Further work and statistical analysis is required to quantify

the albedo change induced by the aerosol, or the impact of aerosols on the cloud life cycle (second indirect effect). Indeed, there is no clear evidence that the huge amount of observations [Quaas *et al.*, 2009] which link an increase of aerosol load to an increase of cloud albedo and an increase of cloud lifetime are strictly connected by a cause-effect relationship. The role of the local meteorology maybe the greatest uncertainty for a proper quantification of the aerosol effect on clouds. In addition, the effect of cloud microphysics altering droplet number concentrations remains to be determined, since a strict CDR – AI relationship does not necessarily quantify the first indirect effect.

[20] The area under study in this paper is also particular because it is affected by strongly absorbing aerosols. These aerosols are transported above the cloud layer and have a strong warming effect that may stabilize the atmosphere and affect the cloud dynamics. It may not be possible to distinguish these various processes by a statistical analysis of the satellite observations alone.

[21] **Acknowledgments.** The data used in this paper were acquired by satellites operated by NASA and CNES. We thank the ICARE center for making the data easily available and providing computing resources.

References

- Bréon, F. M., and M. Doutriaux-Boucher (2005), A comparison of cloud droplet radii measured from space, *IEEE Trans. Geosci. Remote Sens.*, 43(8), 1796–1805, doi:10.1109/TGRS.2005.852838.
- Bréon, F.-M., D. Tanré, and S. Generoso (2002), Effect of aerosols on cloud droplet size monitored from satellite, *Science*, 295(5556), 834–838, doi:10.1126/science.1066434.
- Chand, D., R. Wood, T. L. Anderson, S. K. Satheesh, and R. J. Charlson (2009), Satellite-derived direct radiative effect of aerosols dependent on cloud cover, *Nat. Geosci.*, 2(3), 181–184, doi:10.1038/ngeo437.
- Feingold, G., W. L. Eberhard, D. E. Veron, and M. Previdi (2003), First measurements of the Twomey indirect effect using ground-based remote sensors, *Geophys. Res. Lett.*, 30(6), 1287, doi:10.1029/2002GL016633.
- Hansen, J., M. Sato, and R. Ruedy (1997), Radiative forcing and climate response, *J. Geophys. Res.*, 102(D6), 6831–6864, doi:10.1029/96JD03436.
- Ichoku, C., L. A. Remer, Y. J. Kaufman, R. Levy, D. A. Chu, D. Tanré, and B. N. Holben (2003), MODIS observation of aerosols and estimation of aerosol radiative forcing over southern Africa during SAFARI 2000, *J. Geophys. Res.*, 108(D13), 8499, doi:10.1029/2002JD002366.
- Intergovernmental Panel on Climate Change (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon *et al.*, Cambridge Univ. Press, Cambridge, U. K.
- Kaufman, Y. J., I. Koren, L. A. Remer, D. Rosenfeld, and Y. Rudich (2005), The effect of smoke, dust and pollution aerosol on shallow cloud development over the Atlantic Ocean, *Proc. Natl. Acad. Sci. U. S. A.*, 102, 11,207–11,212, doi:10.1073/pnas.0505191102.
- Labonne, M., F.-M. Bréon, and F. Chevallier (2007), Injection height of biomass burning aerosols as seen from a spaceborne lidar, *Geophys. Res. Lett.*, 34, L11806, doi:10.1029/2007GL029311.
- Nakajima, T., A. Higurashi, K. Kawamoto, and J. E. Penner (2001), A possible correlation between satellite-derived cloud and aerosol microphysical parameters, *Geophys. Res. Lett.*, 28, 1171–1174, doi:10.1029/2000GL012186.
- Peng, Y., U. Lohmann, R. Leaith, C. Banie, and M. Couture (2002), The cloud albedo-cloud droplet effective radius relationship for clean and polluted clouds from RACE and FIRE, ACE, *J. Geophys. Res.*, 107(D11), 4106, doi:10.1029/2000JD000281.
- Penner, J. E., C. Chuang, and K. Grant (1998), Climate forcing by carbonaceous and sulfate aerosols, *Clim. Dyn.*, 14, 839–851, doi:10.1007/s003820050259.
- Quaas, J., O. Boucher, and U. Lohmann (2006), Constraining the total aerosol indirect effect in the LMDZ and ECHAM4 GCMs using MODIS satellite data, *Atmos. Chem. Phys.*, 6, 947–955, doi:10.5194/acp-6-947-2006.
- Quaas, J., B. Stevens, P. Stier, and U. Lohmann (2009), Interpreting the cloud cover—Aerosol optical depth relationship found in satellite data using a general circulation model, *Atmos. Chem. Phys. Discuss.*, 9, 26,013–26,027, doi:10.5194/acpd-9-26013-2009.
- Sekiguchi, M., T. Nakajima, K. Suzuki, K. Kawamoto, A. Higurashi, D. Rosenfeld, I. Sano, and S. Mukai (2003), A study of the direct and indirect effects of aerosols using global satellite data sets of aerosol and cloud parameters, *J. Geophys. Res.*, 108(D22), 4699, doi:10.1029/2002JD003359.
- Storelvmo, T., J. E. Kristjánsson, G. Myhre, M. Johnsrud, and F. Stordal (2006), Combined observational and modeling based study of the aerosol indirect effect, *Atmos. Chem. Phys.*, 6, 3583–3601, doi:10.5194/acp-6-3583-2006.
- Twomey, S. (1977), The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci.*, 34, 1149–1152, doi:10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2.
- Winker, D. M., W. H. Hunt, and M. J. McGill (2007), Initial performance assessment of CALIOP, *Geophys. Res. Lett.*, 34, L19803, doi:10.1029/2007GL030135.

F.-M. Bréon and L. Costantino, Laboratoire des Sciences du Climat et de l'Environnement, Unité Mixte de Recherche, UVSQ, CEA, CNRS, F-91191 Gif-sur-Yvette CEDEX, France. (lore.costantino@gmail.com)