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Long-term variation of cloud droplet number concentrations from space-based Lidar

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A R T I C L E I N F O

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A B S T R A C T

This study presents a new 10 year of liquid water cloud droplet number concentration (N_d) climatology, and analyzes its long-term variation on both regional and global scales based on accurate depolarization ratio measurement from CALIPSO and 3.7 μm cloud effective radius retrieval from MODIS. Compared with the widely used passive retrieval method (e.g., MODIS retrieval), which considers N_d as function of cloud optical depth, geometry thickness and effective radius, retrieval method of the new N_d dataset has a weak dependence upon the cloud adiabatic assumption and eliminates the possible bias caused by multilayer clouds. Statistical results show that the annual cycle and long-term variability of N_d retrieved by CALIPSO agree reasonably well with those obtained from MODIS retrieval method, especially over the stratocumulus regions (correlation coefficient > 0.9). Multiple regression models and contribution calculation verify that the variability of sulfate mass concentration dominates the long-term variation of N_d over most regions, even though the contribution factors and rates vary with different regions, temperatures and methods. In addition, our study also indicates that the impact of BC and OC on N_d should not be ignored, especially for supercooled water clouds over those important biomass burning regions. These results demonstrate the temperature-dependent N_d climatology derived from CALIOP has potential to be beneficial to climate research and reduce the uncertainties in estimates of the aerosol indirect effect in the model simulations.

1. Introduction

Liquid water clouds (e.g., stratiform boundary layer) play a key role in modulating the earth's climate by changing their radiative (e.g., shortwave reflection and infrared emission) (Brenguier et al., 2000; Garrett and Zhao, 2006; Klein and Hartmann, 1993) and precipitation properties (Lohmann and Feichter, 2005). Their formations and variations are closely controlled by the relevant dynamical (Klein and Hartmann, 1993; Myers and Norris, 2016; Seethala et al., 2015; Wood, 2012) and microphysical processes (McCoy et al., 2015, 2017a; Quaas et al., 2009). Higher atmospheric aerosol loading from anthropogenic activities (e.g., rapid industrialization) and natural processes (e.g., volcanic eruptions) may influence cloud properties in various ways. Among many others, the most direct effect of aerosols on clouds is that aerosols serve as cloud condensation nuclei (CCN), increasing the cloud droplet number concentration (N_d) and decreasing the effective radius, thereby enhancing the reflectivity of solar radiation by clouds for a given cloud liquid water content (i.e., "the first aerosol indirect effect" or "Twomey effect") (Twomey, 1977). While any disturbance of N_d caused by increased aerosol concentrations may significantly influence cloud albedo and possibly regionally counteract greenhouse warming, the strength of the first indirect aerosol effect is still a highly uncertain component of the overall global radiative forcing estimation made using global climate models (Ramaswamy et al., 2001). One of the prominent problems is that models fail to capture all the key controls of N_d; thus, they usually employ distinctly different values of N_d and its lower bounds, such that they exhibit a wide range of uncertainty in the simulated magnitudes of the first indirect effect (Lohmann et al., 2007; Quaas et al., 2008). A previous study has shown that the simulated indirect aerosol effect can be reduced by up to 80% when models

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constrain the lower bounds of N_d without regard for the simulated concentrations of activated aerosols (Hoose et al., 2009).

In recent decades, many efforts have been made to decrease the uncertainties of the first indirect effects of the model simulations based on satellite observations, in situ measurements and field campaigns (e.g., Chubb et al., 2016; Huang et al., 2014; Garrett et al., 2004; Lohmann et al., 2000, 2007; Schmidt et al., 2013, 2014; Wang et al., 2010). Ground-based lidar observations and airborne measurements may provide more accurate N_d values, but only limited temporal and spatial coverages are possible (Allen et al., 2011; Donovan et al., 2015; Lu et al., 2007; Schmidt et al., 2015). Thus, the results from in situ observational measurements are commonly used to validate and evaluate satellite-derived N_d (e.g., Ahmad et al., 2013; Painemal and Zuidema, 2011). Until now, the satellite retrieval of N_d has been challenging, and different methods have been presented to derive the climatology of N_d or its precursor (that is, cloud condensation nuclei) (e.g., Bennartz, 2007; Bennartz and Rausch, 2017; Brenguier et al., 2000; Han et al., 1998; Hu et al., 2007a; Rosenfeld et al., 2012, 2016; Schuller et al., 2005). The one method that has been widely used is based on the assumption of an "adiabatic cloud model" and considers the cloud droplet number concentration as a function of cloud optical depth (τ), cloud geometry thickness (H) and effective radius (r_e) at the cloud top (Bennartz, 2007; Brenguier et al., 2000; Schuller et al., 2005). However, most of the clouds in the atmosphere are not strictly adiabatic. Precipitation processes or other factors (e.g., cloud top entrainment) may lead to the clouds being under the sub-adiabatic condition (Wood, 2012; Wood et al., 2012). In contrast to the passive method, Hu et al. (2007a) developed a novel approach to evaluate N_d by combining the lidar depolarization ratio measurements from CALIPSO and the cloud effective radius from MODIS. This method has a weak dependence on the adiabatic assumption and is independent of cloud type. By using one year of data from CALIPSO and MODIS, Zeng et al. (2014) found similar geographical distributions and seasonal variations of N_d between the above two methods. As a result, the advantage of CALIPSO is that it allows us to build a new N_d climatology and further analyze the consistency of the long-term variations between the two N_d datasets.

Such a long-term N_d dataset will be beneficial to determine the factors that contribute to this temporal variability of N_d at the global and regional scales. Many observations and model simulations have verified that increased aerosol concentrations may result in increased N_d (e.g., Bennartz, 2007; Bennartz et al., 2011; Snider et al., 2003). In addition to aerosol concentrations, N_d is also associated with the aerosol size distribution, chemical composition and meteorological conditions (e.g., updraft velocity at cloud base) (Chubb et al., 2016; Reutter et al., 2009; Wood et al., 2012). Reutter et al. (2009) used a cloud parcel model to investigate the dependence of N_d on the aerosol number concentrations and updraft velocities, and found that the sensitivity of N_d to aerosols and velocity varies with region. Karydis et al. (2012) tested the adjoint sensitivity of global N_d values to aerosol and dynamic parameters. Their simulation showed that N_d is more sensitive to updraft velocities and water uptake coefficients (aerosol number concentration and hygroscopicity) over polluted (pristine) areas. Over the southern oceans, McCoy et al. (2015) analyzed the correlations between N_d and aerosols, and noted that natural aerosols affect the spatiotemporal variability of N_d and may explain the seasonal and spatial patterns of the Southern Ocean cloud albedo, which is consistent with the results of the study by Karydis et al. (2012). However, a recent model simulation demonstrated that the updraft velocity is the primary driver of N_d variability for 45.5% of the grid, and the sensitivity of the temporal variability of N_d to the velocity cannot be neglected over the southern oceans (Sullivan et al., 2016). Thus, to reconcile such an inconsistency between model simulations, we perform an adjoint sensitivity analysis of N_d to aerosol type and vertical velocity by using two satellite-observed N_d datasets derived from CALIPSO and MODIS, the aerosol properties from the Modern-Era Retrospective Analysis for Research and Applications Version 2 (MERRA2) and the updraft velocities from the ERA-interim and MERRA2 datasets. Based on this investigation, we attempt to focus on two key points: (1) What factors drive the temporal variability of N_d at regional and global scales? (2) Which one is the dominant factor? Although some statistical results agree reasonably well with previous studies, new insights are also presented.

This paper is organized as follows. A brief introduction to all the datasets and retrieval methods used in this study is given in Section 2. Section 3.1 prescribes the comparisons of the geographical, annual and long-term variations of the N_d between the two retrieval methods. Further analyses of the contributions of the aerosols and vertical velocities to the long-term variabilities of regional N_d are provided in Section 3.2. Finally, the conclusions are presented in Section 4.

2. Datasets and methodology

In this study, 10 years (2007–2016) of data from the Aqua-MODIS collection 6 level-2 cloud product (MYD06), the CALIPSO Lidar level-2 cloud layer products, and the daily 3-hour aerosol product from the MERRA2 reanalysis were collected. Then, these datasets are used to retrieve the liquid water cloud droplet number concentrations during the daytime and to discuss the contributions from different factors on its temporal variability.

2.1. Satellite products and reanalysis dataset

The effective cloud radius of 3.7 μm (r_e), cloud optical thickness (τ), cloud multi-layer flag (CMLF) with a spatial resolution at the nadir of 1 × 1 km, and cloud fraction (CF) with a spatial resolution at 5 × 5 km from the Aqua-MODIS level-2 collection 6 cloud products (MYD06) (Platnick et al., 2015, 2017) during the daytime were used in our study. Compared with the MODIS collection of 5 cloud products, several improvements have been made (Rausch et al., 2017), such as significant improvements in the forward radiative transfer models.

The collocated CALIPSO level-2 1 km (v4.10) cloud layer product provides essential cloud thermodynamic phases (e.g., water, randomly oriented ice, horizontally oriented ice or unknown phase) at the cloud top, the cloud top and base height (temperature and pressure) information, the layer-integrated volume depolarization ratio and the number of cloud layers in a given Lidar profile (Hu et al., 2009). Compared with the earlier CALIOP version 3 products, there have been several substantial improvements made to increase the retrieval accuracies of the parameters needed to determine the N_d from Lidar (e.g., improved cloud subtypes and ice-water phase determination).

In addition, the Modern-Era Retrospective Analysis for Research and Applications Version 2 (MERRA2) combines measurements of the atmospheric states and remotely sensed aerosol optical depths to provide the aerosol reanalysis (Buchard et al., 2015; Molod et al., 2015), which has been evaluated by CALIOP measurement in recent studies (Buchard et al., 2017; Nowottnick et al., 2015; Li et al., 2016). Here, the daily 3-hour aerosol and meteorological products from the MERRA2 reanalysis, which have gridded resolutions of 0.5° × 0.625°, are also used to provide the related information of the updraft velocity (w) and mass concentration of different aerosol species at several pressure levels. The MERRA2 product can supply the mass mixing ratios of eight aerosol types, including black carbon (BC), dimethyl sulfide (DMS), dust (DU), methane sulfonic acid (MSA), organic carbon (OC), sulfate aerosol (SO_4), sulfur dioxide (SO_2) and sea salt (SS). Some studies have addressed the effects of MSA, OC, BC, sulfate and SS aerosols as cloud condensation nuclei (CCN) (Ayers and Gras, 1991; Lammel and Novakov, 1995; O'Dowd et al., 1997; Ruehl et al., 2016; Sun and Ariya, 2006). Following the studies of Sullivan et al. (2016) and McCoy et al. (2017b), this investigation uses only the mass mixing ratios of hydrophilic OC, BC, SO_4, SO_2 and the smallest particles of SS (that is, 0.03–0.1 μm size bin) and dust (that is, 0.1–1 μm size bin) to calculate their mass concentrations at different pressure levels. In addition, the daily 6-hour vertical velocities from the ERA-interim reanalysis