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# Multi-year ground-based observations of aerosol-cloud interactions in the Mid-Atlantic of the United States



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#### ABSTRACT

The U.S. Mid-Atlantic region experiences a wide variability of aerosol loading and frequent episodes of elevated anthropogenic aerosol loading associated with urban pollution conditions during summer months. In this study, multi-year ground-based observations (2006 to 2010) of aerosol and cloud properties from passive, active and in situ measurements at an atmospheric measurement field station in the Baltimore-Washington corridor operated by Howard University were analyzed to examine aerosol indirect effect on single-layer warm clouds including cloud optical depth (COD), liquid water path (LWP), cloud droplet effective radius  $(R_e)$  and cloud droplet number concentration  $(N_d)$  in this region. A greater occurrence of polluted episodes and cloud cases with smaller Re  $(<7 \mu m)$  were found during the polluted year summers (2006, 2007 and 2008) than the clean year summers (2009 and 2010). The measurements of aerosol particulate matter with aerodynamic diameter  $\leq 2.5 \,\mu m$  (PM2.5) were used to represent the aerosol loading under cloudy conditions. Significant negative relationships between cloud droplet  $R_e$  and PM2.5 were observed. Cloud cases were separated into clean and polluted groups based on the value of PM2.5. The cloud droplet R<sub>e</sub> was found proportional to LWP under clean conditions but weakly dependent on LWP under polluted conditions, The  $N_d$  was proportional to LWP under polluted condition but weakly dependent on LWP under clean conditions. Moreover, the effects of increasing fine aerosol particles on modifying cloud microphysical properties were found more significant under large LWP than small LWP in this region.

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### 1. Introduction

Investigation of the radiative forcing from aerosol-cloud interactions (RFaci) is crucial to estimates and interpretations of the Earth's changing energy budget [49]. The changes of cloud active aerosols can impact cloud microphysical properties, precipitation and the meteorological and radiance responses of clouds. A growing list of studies with space- and ground-based observations provides convincing evidences of

\* Corresponding author. Tel.: +1 202 865 8678. E-mail address: siwei.li@howard.edu (S. Li). RFaci. By using satellite remote sensing, Han et al. [11] showed that cloud-droplet concentrations correlate to the cloud condensation nuclei (CCN) at different regions. Feingold et al. [7] defined the ratio of logarithmic  $R_{\rm e}$  and aerosol optical depth (AOD) to represent the RFaci and Feingold et al. [8,9] reported the observed RFaci by using ground-based observations of cloud and aerosol properties provided by the Atmospheric Radiation Measurement (ARM) at the Southern Great Plains (SGP) site in Oklahoma. Also with ARM SGP observations Kim et al. [17–19] demonstrated the positive relationship between COD and LWP, an inverse relationship between  $R_{\rm e}$  and aerosol scattering coefficient and the role of adiabaticity in RFaci. Nzeffe et al. [31] showed that  $R_{\rm e}$  reduced under polluted

airmasses for given LWP based on the ground-based observations from Howard University Beltsville Campus (HUBC) facility.

However untangling aerosol effects on clouds and precipitation is still challenging. The extent to which aerosols impact clouds can be different or even opposite under different cloud regimes. The RFaci can be buffered by compensation between different cloud responses to aerosols [33,36]. That explains why the statistical effects of aerosol on clouds and precipitation are not agreed upon. But the RFaci is still evident in specific circumstance or regimes [36,16,32]. The complexity of the climate system and the inadequacy of measurements and methodologies have made it very challenging to obtain a more detailed understanding of aerosolcloud interactions and their effects on climate [24,36]. Part of the challenge is reducing the uncertainty in estimates of RFaci, which as pointed out by McComiskey and Feingold [24] requires small scale studies. Though field experiments that produce "process scale" observations of aerosol-cloud interaction continue to occur, historically they have been insufficient in number, regional diversity and duration. Current satellite-based sensors can provide global coverage and longterm measurements of aerosols and clouds. However detailed understanding of the RFaci is limited by the large scale, coarse time resolution and inherent limitations in the retrieval algorithms and sensors. Ground-based sensors can provide long-term measurements which are more accurate, stable, smaller scale and higher temporal resolution at specific region of interest compared to satellite sensors. A number of groundbased observation facilities (e.g. ARM sites) have been developed to study aerosol, clouds, precipitation and their influences on global climate change. Independent from ARM the HUBC station (39.054°N and 76.877°W) was established for atmospheric measurements in the U.S. Mid-Atlantic region. This region experiences a wide variability of aerosol loading and frequent episodes of elevated anthropogenic aerosol loading associated with urban pollution conditions during summer months as seen in analyses of derived AOD from Aerosol Robotic Network (AERONET) and a collocated air quality monitoring operated by the Maryland Department of the Environment (MDE) [14]. Thus, the observations of aerosol and cloud macro- and micro-physical properties in this region are valuable for investigating aerosol-cloud interaction and its impacts on weather and climate. Nzeffe et al. [31] previously observed aerosol indirect effect based on six months of observations at HUBC site in 2005. This study extends their work to analysis five years (from 2006 to 2010) of ground-based observations of aerosol and cloud properties for systematically investigating aerosol impacts on variability of cloud properties including COD, LWP,  $R_e$  and  $N_d$  during summer in this region.

### 2. Measurements

The HUBC facility in Beltsville, MD is situated in a ruralsuburban transition region between Washington, DC and Baltimore, MD urban centers. It has a wide range of sensors deployed to observe atmospheric radiation, surface fluxes, aerosol, cloud properties and other climate and weather processes [31].

Among the sensor observations, LWP is determined from a dual frequency (23.8 and 31.4 GHz) Microwave radiometer (MWR) [46]. The error of the MWR LWP retrieval consists of instrument error, errors associated with the climatological profiles used for the retrieval (given day to day variability of atmospheric temperatures from this climatology), and errors from the absorption model used to develop parameters for the retrieval algorithm [40]. The errors associated with instrument and climatological profiles used are considered random errors and thus are minimized with increasing sample size of data. The error associated with absorption model is considered a systematic bias and represents the preponderance of the total error. Instrument uncertainty and retrieval errors associated with the climatological profiles and the microwave absorption model results in a total uncertainty of retrieved LWP around 20 g/m<sup>2</sup> [40] but the consequence of the systematic error is minimized while relative differences are investigated (e.g cloud droplet Re vs. LWP).

AOD is measured with a MultiFilter Rotating Shadowband Radiometer (MFRSR) which is a sensor with a shading band that rotates, measuring global downwelling irradiance, diffuse irradiance and direct beam irradiance calculated from global and diffuse irradiance. More detail on the instrument design can be found in Harrison et al. [13]. The MFRSR is calibrated using data acquired on clear sky days via the Langley regression which is based on linear regressions of the log of direct beam irradiance versus airmass and the calibration constant  $I_0$  is used to compute transmittances during cloudy conditions [13,12]. The AOD is retrieved based on the algorithm developed by Harrison and Michalsky [12]. For quality control, the retrieved AOD is compared with AERONET observation at NASA Goddard Space Flight Center (GSFC), around 5 miles southeast of the HUBC facility. High correlation coefficient (0.94) is found between them during the study period (from 2006 January to 2010 December).

However retrieval of AOD is not available from any passive remote sensor during cloud periods, so hourly in situ measurements of particulate matter with aerodynamic diameter  $\leq 2.5~\mu m$  (PM2.5) are obtained from samplers operated by the MDE at the HUBC site to estimate aerosol loading under cloudy conditions. Although surface PM2.5 is related more to small particles within boundary layer while AOD presents the total column aerosol loading, surface PM2.5 had a good correlation with AOD in the District of Columbia–Maryland area based on the study of PM2.5-AOD relationship in the United States [23]. We also found that the measured PM2.5 has a significant positive relationship with MFRSR retrieved AOD with correlation coefficient of 0.63 during clear-sky conditions in summer as seen in Fig. 1.

In situ aerosol size distributions are measured by a Fast Mobility Particle Sizer (FMPS) which was developed based on electrical aerosol spectrometer technology from Tartu University [38,39]. The FMPS measures particle size distributions in the range from the 6 nm to 560 nm with 16 channels per decade every second. However the FMPS measurements at HUBC are only available during the NASA DISCOVER-AQ (a field campaign for deriving information on surface conditions from column and vertically resolved observations relevant to air quality) field campaign in the

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