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Differences in fine particle chemical composition on clear and cloudy days

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Abstract

Clouds are prevalent and alter $PM_{2.5}$ mass and chemical composition. Cloud-affected satellite retrievals are often removed from data products, hindering estimates of tropospheric chemical composition during cloudy times. We examine surface fine particulate matter ($PM_{2.5}$) chemical constituent concentrations in the Interagency Monitoring of PROtected Visual Environments network during Cloudy and Clear Sky times defined using Moderate Resolution Imaging Spectroradiometer (MODIS) cloud flags from 2010–2014 with a focus on differences in particle hygroscopicity and aerosol liquid water (ALW). Cloudy and Clear Sky periods exhibit significant differences in $PM_{2.5}$ and chemical composition that vary regionally and seasonally. In the eastern US, relative humidity alone cannot explain differences in ALW, suggesting emissions and *in situ* chemistry exert determining impacts. An implicit clear sky bias may hinder efforts to quantitatively to understand and improve model representation of aerosol-cloud interactions.

Plain Language Summary

Satellite retrievals affected by clouds are often removed from final data products, hindering knowledge of chemical composition under cloudy conditions. Much of the contiguous US (CONUS) is covered by visible clouds much of the time, and clouds alter fine particulate matter ($PM_{2.5}$) chemical composition and mass concentration. We investigate differences in $PM_{2.5}$ mass and chemical composition between cloudy and clear sky times from 2010–2014. We use surface measurements of $PM_{2.5}$ chemical constituents from the Interagency Monitoring of PROtected Visual Environments (IMPROVE) network, grouped into Cloudy and Clear Sky bins based on the cloud flag from the Moderate Resolution Imaging Spectroradiometer. We find that $PM_{2.5}$ mass and chemical composition varies regionally and seasonally across the CONUS between Cloudy and Clear Sky time periods. We find an important role for chemical composition linked to emissions and formation processes to explain Cloudy and Clear Sky differences in aerosol liquid water (ALW). This work suggests that an implicit clear sky bias hinders efforts to quantitatively understand particulate matter chemical composition during cloudy times.

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Further data including particle chemical speciation and meteorology information, estimated growth factors, region locations, and model input details can be obtained in the Supporting Information.

1 Introduction

At any given time, visible clouds cover over 60% of the Earth's surface (King et al., 2013), and a warming climate causes cloud cover to change (Norris et al., 2016). Average cloud fraction values over the contiguous US (CONUS) are ~40% year-round with higher values in the winter (44–54%) than the summer (26–34%) (Ju & Roy, 2008; Kovalskyy & Roy, 2015). Clouds act as atmospheric aqueous phase reactors, and their condensed phase oxidative chemistry generates particle mass aloft, such as sulfate (Zhou et al., 2019) and water-soluble organic carbon (Carlton et al., 2008; Duong et al., 2011) Clouds are the primary drivers of vertical transport in the atmosphere, moving trace species from the boundary layer to the free troposphere (FT) (Ervens, 2015).. The radiative impacts of aerosols in the FT are substantial, especially when located above clouds where aerosols scatter and absorb both incoming solar radiation and diffuse back scatter from clouds (Seinfeld, 2008). Aerosol-cloud interactions are complex and a critical uncertainty in model projections of the future (Fan et al., 2016).

Atmospheric chemistry laboratory studies, sampling, modeling and analysis strategies designed to minimize cloud and water influences lead to an implicit, yet persistent clear sky bias in the quantitative understanding of tropospheric composition. During atmospheric chemistry field campaigns, aircraft typically avoid clouds. There is increased error in remotely sensed aerosol optical thickness (AOT) retrieval techniques during cloudy times (Martin, 2008), and impacted retrievals are often screened from final data products to avoid measurement artifacts. Most validation of satellite-derived AOT through comparison to surface measurements, such as those from sun photometers used to retrieve AOT from the ground up, is conducted for cloud-free periods (Liu et al., 2018). Air quality models are often evaluated with cloud-free satellite retrievals (van Donkelaar et al., 2010; Guo et al., 2017; de Hoogh et al., 2016; Song et al., 2014; J. Tian & Chen, 2010) and cloud-free aircraft samples (Bray et al., 2017; McKeen et al., 2009). This biases model development and predictive skill toward cloud-free conditions. Laboratory experiments to understand particulate matter formation are conducted under dry conditions (e.g., Lamkaddam et al., 2017; Ng et al., 2007) atypical of cloudy time periods. Should differences in aerosol physicochemical properties exist between cloudy and clear sky time periods, current approaches are limited in their ability to quantitatively assess those differences. This is a key knowledge gap.

Characterization of fine particulate matter ($PM_{2.5}$) mass and chemical composition in the US primarily relies on surface measurements from relatively sparsely spaced monitors. At various locations across the CONUS, the Interagency Monitoring of PROtected Visual Environments (IMPROVE) network samples every 3 days, and the Chemical Speciation Network (CSN) samples every 3 or 6 days (US Environmental Protection Agency, 2008). To improve upon surface network spatial and temporal limitations of point measurements, data can be interpolated to describe particle mass (Li et al., 2014; G. Zhang et al., 2018) and chemical composition over larger areas (Y. Liu et al., 2009; Tai et al., 2010). Satellite information can also be used (van Donkelaar, Martin, Brauer, et al., 2015), such as the Moderate Resolution Imaging Spectroradiometer (MODIS) instruments aboard the Aqua and Terra satellite platforms. These view the entire Earth surface every 1 to 2 days and

are used to impart information about AOT translated to $PM_{2.5}$ mass concentrations for use in air quality applications (van Donkelaar, Martin, Brauer, et al., 2015; Gupta et al., 2006; Kloog et al., 2011; Sorek-Hamer et al., 2016). Many advanced satellite AOT models translate space-based radiation measurements to surface $PM_{2.5}$ (van Donkelaar et al., 2010; van Donkelaar, Martin, Brauer, et al., 2015; van Donkelaar, Martin, Spurr, et al., 2015; Gupta et al., 2006; Kessner et al., 2013; Kloog et al., 2011; Kumar et al., 2007; Y. Liu et al., 2011; Schaap et al., 2009; J. Wang et al., 2012; J. Wang & Christopher, 2003) and employ sophisticated techniques which account for aerosol size and type, vertical extinction, mass, and relative humidity (RH) (van Donkelaar et al., 2010). Evaluation of AOT-to-$PM_{2.5}$ techniques finds that monthly aggregated AOT can robustly estimate relationships spanning five years of daily mean values over North America ($R>0.77$) (van Donkelaar et al., 2010). While temporal and geospatial satellite AOT is useful for understanding trends in $PM_{2.5}$ concentrations (van Donkelaar, Martin, Brauer, et al., 2015; Sorek-Hamer et al., 2016; J. Wang & Christopher, 2003), an implicit constraint for this and other similar findings is that such agreement is for clear sky conditions.

Surface networks record $PM_{2.5}$ mass and chemical composition during clear sky and cloudy time periods alike. The difference between spatially and temporally aggregated $PM_{2.5}$ mass concentrations in the CONUS for cloudy and all sky (cloudy + clear sky) conditions is estimated to be ± 2.5 μg m^{-3} (Christopher & Gupta, 2010). Less attention has been given to clear sky and cloudy differences in $PM_{2.5}$ chemical composition, especially with regards to particle hygroscopicity and water uptake. Aerosol mass concentrations and chemical speciation including aerosol liquid water (ALW) influence AOT (Christiansen et al., 2019; Malm et al., 1994; Nguyen et al., 2016; Pitchford et al., 2007), cloud microphysics, and mesoscale convective systems (Kawecki & Steiner, 2018), including storm morphology and precipitation patterns (Kawecki et al., 2016). Particle chemical composition modulates particle size via water uptake. Particle size is a determining factor in light scattering by particles, which is important for aerosol radiative properties. An implication of this work is that if particle hygroscopicity changes from clear sky to cloudy time periods, when aerosol-cloud interactions are most important, a quantitative understanding remains unclear.

In this work, we test the hypothesis that there are quantitative differences in $PM_{2.5}$ chemical composition between cloudy and clear sky time periods in ways important for water uptake. We employ a combination of satellite products, surface measurements, and thermodynamic modeling to analyze annual and seasonal trends in different chemical climatology regions across the CONUS. We assess and quantify seasonal statistical significance (Kahn, 2005) for differences in distributions of RH, $PM_{2.5}$, and chemical speciation during cloudy and clear sky times using surface measurements from the IMPROVE network from 2010–2014 within the context of MODIS cloud flag values. Further, we examine one chemical climatology region in detail, the Mid South, as a case study. This region encompasses the location of the Atmospheric Radiation Measurement Southern Great Plains (SGP) site in an area of the CONUS that experiences varied weather patterns, a broad range of cloud conditions, and distinct seasonal variations in temperature and humidity (Sisterson et al., 2016).

2 Data and Methods

Cloudy and clear sky classifications are determined using publicly available data (National Aeronautics and Space Administration, 2018) from MODIS on the Aqua and Terra satellites. Pairing of satellite and surface $PM_{2.5}$ mass measurements typically works best in rural and vegetated locations, where the spectral properties of the background tend to be dark and vary little over the space of a satellite grid cell (Hauser, 2005; Jones & Christopher, 2010). For this reason, we use rural IMPROVE network sites that are located primarily in national parks, although improvements have been made for retrievals over bright surfaces such as deserts and urban areas (Hauser, 2005; Hsu et al., 2004, 2006, 2013; H. Zhang et al., 2016). We use 500 m resolution pixels that contain the IMPROVE sites. Retrievals are flagged as cloudy if QA flags specifically identified clouds as preventing retrieval (LAND_GOOD_FLAG=11, LAND_BAD_FLAG=3), or if 2.1-micrometer reflectance was too high (r>0.35) and the fraction of 500 m sub pixels that were cloudy was greater than 44.4% (LAND_CLOUD_FRAC>0.444). We choose 44.4% because it is a fundamental limit of the algorithm (Remer et al., 2013). IMPROVE monitors are frequently under a MODIS swath with valid retrievals even if the pixel containing the IMPROVE station is not successfully retrieved. As an alternative to the IMPROVE pixel, we employ a method for quality assurance, a 17×17 grid. This allows for any retrieval within a 50 km x 50 km area to represent the IMPROVE station. If all 17×17 pixels are not retrieved, then the state over the monitor is determined to be cloudy. The 17×17 grid approach is much more likely to attribute non-retrieved data to clouds (98.5%) than the containing pixel approach, which attributes 89.8% of non-retrieved data to clouds. Misidentifying non-retrievals as cloudy is unlikely to substantially affect interpretation, as the sample size is large (N>70,000 total observations, and N>1500 for an individual region).

IMPROVE network data were downloaded on 13 July 2015 and 26 May 2016 from public archives (http://vista.cira.colostate.edu/Improve/) (IMPROVE Network, 2019) for 132 unique sites across the CONUS with complete data records for the years 2010–2014 (Figure S1a). IMPROVE data is collected every 3 days. We investigate 24-hour average $PM_{2.5}$ mass, ALW, RH, sulfate (SO_4^{2-}), nitrate (NO_3^-), and total organic carbon (TOC) mass concentrations. We group IMPROVE sites across the CONUS into 22 chemical climatology regions defined by the IMPROVE network (Figure S1b) (Hand et al., 2011; Malm et al., 2017). $PM_{2.5}$ mass and composition is provided directly from the IMPROVE database, while ALW is estimated.

ALW is a function of RH, particle concentration, and chemical composition. We estimate ALW using a metastable assumption in the inorganic (K^+–Ca^{2+}–Mg^{2+}–NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O) aerosol thermodynamic equilibrium model ISORROPIA 2.1 (Fountoukis & Nenes, 2007). We use the reverse, open-system problem because only aerosol measurements are available. Particle mass concentration inputs of SO_4^{2-} and NO_3^- are taken from IMPROVE measurements. Because of limited measurement availability, ammonium ion is not considered. Dust and organic species are also not considered because water uptake properties are not well constrained (Jathar et al., 2016; Metzger et al., 2018), and there is large spatial heterogeneity in dust mass concentrations. Excluding dust and organics from ISORROPIA estimates alters ALW concentrations but does not affect overall interpretation

(Figure S2), consistent with an earlier sensitivity using this technique and including organic compounds (Nguyen et al., 2015). The temperature and RH were extracted from the North American Regional Reanalysis (NARR) model (Kalnay et al., 1996) similar to Nguyen et al (Nguyen et al., 2016).

Cloudy and clear sky differences in ALW are investigated in two ways. First, we compare ALW estimated using 24-hour average chemical composition and meteorology and group results into Clear Sky and Cloudy bins using the MODIS cloud flag. We use these daily values when comparing ALW within chemical climatology regions. Second, we investigate trends across the eastern US to isolate the effect of chemical composition. We select the eastern US since ALW concentrations are largest in this region (Figure S3). We group 24-hour average chemical composition and meteorology into Clear Sky and Cloudy bins and take monthly medians. We perform ALW estimations using the medians via three ISORROPIA calculation scenarios: 1) Clear Sky chemical composition and Clear Sky meteorology (“Clear Sky” scenario), 2) Cloudy chemical composition and Cloudy meteorology (“Cloudy”), and 3) Clear Sky chemical composition and Cloudy meteorology (“Mixed”) (Table S1, Figure S4). We use monthly medians to avoid complications that arise from differing numbers of Cloudy and Clear Sky days in the Mixed scenario. We perform the Mixed scenario to reproduce studies in which growth factors are measured under clear sky conditions (e.g., Brock et al., 2016) and eventually applied to models which contain cloudy meteorological conditions. When the Mixed scenario is significantly different than Cloudy, we can reject the hypothesis that RH and temperature alone explain the difference.

Growth factors used in the Mid South region are estimated from a modified Kohler equation (Brock et al., 2016; Jefferson et al., 2017) (Equation 1). We use RH from the NARR and estimate κ_d, the particle hygroscopicity, from IMPROVE-measured chemical composition mass concentrations and individual species κ values (κ_{SO4} = 0.5 and κ_{NO3} = 0.7).(Petters & Kreidenweis, 2007) Here, $gf(D)$ is the hygroscopic diameter growth.

$$gf(D) = \left(1 + \kappa_d \frac{RH}{100 - RH}\right)^{1/3} \quad (1)$$

Statistical significance for differences in measurement distributions of $PM_{2.5}$ chemical composition and properties between Cloudy and Clear Sky time periods for all seasons in the years 2010–2014 is determined using the Mann-Whitney U Test in R statistical software (R Core Team, 2013). The Mann-Whitney U Test is a non-parametric test that compares two samples to assess whether population distributions differ (McKnight & Najab, 2010). The timeframe 2010–2014 encompasses typical conditions, and coincides with several intensive observation periods including the Southeast Atmosphere Studies (SAS) (Carlton et al., 2018), the Studies of the Emissions and Atmospheric Composition, Clouds, and Climate Coupling by Regional Surveys (SEAC^{4}RS) (Toon et al., 2016), and the California Research at the Nexus of Air Quality and Climate Change (CalNex) (Ryerson et al., 2013) field campaigns,. We define cloud fraction for each region as the number of MODIS-flagged cloudy IMPROVE sampling days over the total number of IMPROVE sampling days. Further, we define winter as December, January, and February (DJF), spring as March,

April, and May (MAM), summer as June, July, and August (JJA), and fall as September, October, and November (SON).

3 Results and Discussion

3.1 Hygroscopicity and chemical composition

Distributions in monthly particle chemical composition across the eastern US in 2010–2014 are sufficiently changed between MODIS-defined Cloudy and Clear Sky times to affect hygroscopicity and alter predicted ALW mass concentrations beyond differences that would arise from changes in meteorology alone (Figure 1). The only difference between the Mixed and Cloudy ALW calculations is that the Mixed scenario employs Clear Sky chemical composition extrapolated to Cloudy meteorology. This can occur in model development or satellite validation applications when $PM_{2.5}$-AOD relationships or growth factors are unmeasured for Cloudy periods (e.g., van Donkelaar et al., 2010; de Hoogh et al., 2016; J. Tian & Chen, 2010; Brock et al., 2016). When Clear Sky chemical composition is extrapolated to Cloudy period meteorology (“Mixed”), monthly median ALW concentrations in the eastern US are significantly different from our best estimate, which employs the actual chemical composition during cloudy periods (“Cloudy”), in all seasons except winter. Interestingly, monthly median Clear Sky and Cloudy scenario ALW concentrations do not differ significantly except during winter despite higher Cloudy RH (Figure S5). This suggests chemical composition is a determining factor in ALW uptake on cloudy days because the pattern in ALW is opposite the pattern in RH. Clear Sky/Cloudy patterns in SO_4^{2-} and NO_3^- mass concentrations, which affect particle hygroscopicity, vary regionally and seasonally. When aggregated over the eastern US, ALW estimates for the Mixed case are largest during summer and spring and can be explained by elevated Clear Sky SO_4^{2-} and NO_3^- concentrations and high Cloudy RH. Generally, Mixed ALW concentrations in the eastern US are higher than for the Cloudy scenario during each season because Clear Sky chemical composition facilitates greater hygroscopicity and Cloudy RH is elevated (Table S2). A notable exception is the Ohio River Valley during winter, where Cloudy SO_4^{2-}, NO_3^-, and RH are higher than Clear Sky. In this case, Cloudy period ALW concentrations are higher than for the Mixed scenario. These findings highlight that a changing $PM_{2.5}$ chemical composition has a determining effect on ALW mass concentrations (Nguyen et al., 2016), a critical element in the estimation of aerosol-cloud interactions and particle radiative impacts. During cloudy periods, when the accurate prediction of ALW and aerosol-cloud interactions is most critical, *in situ* knowledge of $PM_{2.5}$ chemical composition is required.

Differences in daily mass concentrations of fine particle chemical constituents between Cloudy and Clear Sky periods across the CONUS are spatially and temporally different among $PM_{2.5}$ mass and its chemical constituents except in the Northwest region (Tables S3-S7, Figure S6). These patterns cannot be adequately described as a function of MODIS cloud fraction (Figures S7-S8). If meteorological processes and physical transport are the only controlling factors, then patterns in mass concentrations among $PM_{2.5}$ and constituents should not vary. However, they do, suggesting differences in emissions and/or *in situ* chemical production of $PM_{2.5}$ during Cloudy and Clear Sky time periods. Where differences

are significant for ALW, Cloudy ALW is higher than Clear Sky in all seasons, with few exceptions (Figure 2, Table S3). Water uptake contributes to particle growth with a determining impact on particle size and radiative properties. $PM_{2.5}$ mass, greater during Clear Sky times in most regions and seasons, has nearly an opposite pattern to ALW spatial and seasonal trends (Figure 3, Table S3). Although Cloudy vs. Clear Sky differences are observed across the CONUS, the largest differences are observed in the central and eastern US during winter (Figure 2, Table S2). Wintertime Cloudy SO_4^{2-} mass concentrations are greater than Clear Sky (Figure S9, Table S5), and the highest NO_3^- mass concentration differences are observed during Cloudy times in winter when temperatures are coldest (Figure S10, Table S8). This promotes thermodynamic stability of nitrate in the condensed phase, increasing particle hygroscopicity and facilitating ALW.

Outside of winter, significant SO_4^{2-} mass concentrations are typically higher on Clear Sky days in the eastern US (Figures S9 & Tables S5-S6). In the eastern US summer, SO_4^{2-} differences are greater during Clear Sky times than Cloudy. Higher Clear Sky SO_4^{2-} concentrations during these times may be associated with heat waves and stagnation events prevalent during summer, which are characterized by a lack of ventilation in high pressure systems (Jacob & Winner, 2009; J. X. Wang & Angell, 1999), higher air conditioning loads, and electricity demand (Farkas et al., 2016).

TOC mass concentrations are nearly always higher during Clear Sky times than Cloudy (Figure S11, Table S7) in all chemical climatology regions across the CONUS, with the largest differences during summer and fall. Precursor VOC emissions (e.g., biogenic) and subsequent derived PM that contributes to OC differ by season and region (Donahue et al., 2009; Gentner et al., 2017). Increased sunlight under clear sky conditions leads to higher biogenic VOC emissions (Sakulyanontvittaya et al., 2008) and enhanced photolysis rates that facilitate hydroxyl radical (OH) production important to secondary organic aerosol formation (Tang et al., 2003). We note that TOC is also influenced by primary sources of OC including wildland fires in the west and prescribed burning in the east which are not influenced by cloud presence (Spracklen et al., 2007); (D. Tian et al., 2009; Zeng et al., 2008).

3.2 $PM_{2.5}$ Mass Concentrations

Significant differences in $PM_{2.5}$ mass concentrations measured at IMPROVE monitoring locations are observed between Cloudy and Clear Sky conditions in the majority (>60%) of regions in any given season during 2010–2014 (Figure 3 and Table S4) and do not trend with MODIS cloud fraction during any season in any region (Figure S12). In all regions, Clear Sky $PM_{2.5}$ concentrations are generally higher than Cloudy. Satellite AOT products used to derive $PM_{2.5}$ may overestimate the atmospheric burden across the CONUS, particularly during summertime. Median All Sky $PM_{2.5}$ concentrations are also significantly different than Clear Sky in multiple chemical climatology regions and are typically lower than Clear Sky concentrations (Table S9). This suggests the clear sky bias in satellite data may impart a positive bias when assessing surface $PM_{2.5}$ trends in model applications for air quality, weather, and climate.

3.3 Case Study: The Mid South

ALW concentrations are significantly higher during Cloudy times than Clear Sky in the Mid South during all seasons (Table 1, Figure S13). RH in the region is high year-round during Cloudy and Clear Sky periods alike, with the median greater than 60%. Gas-phase water vapor mixing ratios are sufficiently high that water availability is not limiting for ALW in the region for any season. Aerosol mass concentrations and chemical composition vary, however, and the effects on particle hygroscopicity can be seen in contrasting Cloudy and Clear Sky ALW concentrations among the seasons. For example, during Clear Sky conditions, the highest ALW mass concentrations occur during summer and spring, which correspond to the highest SO_4^{2-} concentrations in the Mid South, and not when Clear Sky RH is highest (i.e., during winter). The largest absolute ALW concentrations and estimated growth factors occur during Cloudy times in the winter and spring, when NO_3^- mass fraction and RH are highest. This is consistent with independent humidified nephelometer measurements by Jefferson et al. who find that aerosol growth rates are highest in the winter and spring at the SGP site within the Mid South chemical climatology region, and identify nitrate and RH as determining factors (Jefferson et al., 2017).

NO_3^- concentrations are generally lower than SO_4^{2-} in the Mid South, but NO_3^- is more hygroscopic and provides influence over ALW patterns. Sulfate is traditionally considered dominant in determining absolute ALW mass concentrations in this region, and sulfate mass fraction is highest in summer. (A. G. Carlton & Turpin, 2013; Gasparini et al., 2006) Similar to other regions of the CONUS, SO_4^{2-} mass concentrations are greatest during summertime Clear Sky conditions due to transport (Parworth et al., 2015), increased rates of photochemistry (Stone et al., 2012), and increased electricity sector emissions during heat waves and stagnation events (Appel et al., 2011; Farkas et al., 2016), which generally occur on sunny days. Sulfate mass fraction is lowest in winter, when nitrate concentrations are high due to cooler temperatures and transport of precursor species from nearby agricultural and surrounding urban areas (Parworth et al., 2015). Year-round NO_3^- concentrations are higher during Cloudy conditions than Clear Sky, which are associated with lower temperatures. Under Cloudy conditions, the highest ALW concentrations and estimated growth factors occur during winter and spring, when NO_3^- mass fraction and RH are highest. In another continental location, the Po Valley in Italy, NO_3^- was found to control ALW concentrations with implications for SOA (Hodas et al., 2014). The Mid South is also a continental, agricultural area and aerosol growth may be subject to similar mechanisms.

4 Conclusions

Across the CONUS, statistically discernible differences among $PM_{2.5}$ and chemical constituent concentrations under Cloudy and Clear Sky conditions cannot be explained solely by physical mechanisms. The chemical properties of aerosol are important to explain differences in water uptake and particle composition under different meteorological conditions. While meteorological phenomena such as pressure systems, winds, and air mixing affect $PM_{2.5}$ and chemical component concentrations, they are not sufficient to explain chemical constituent differences between Cloudy and Clear Sky times. *In situ*

chemical formation processes are necessary to fully explain temporal and spatial patterns. Spatially and seasonally, $PM_{2.5}$ and particle speciation information that lends insight into water uptake, particle properties, and particle growth is incomplete when information is gathered only during clear sky time periods. The work presented here indicates aerosol growth due to water uptake is greatest during satellite periods identified as Cloudy in many regions. Satellites are unable to remotely sense particle properties and impacts during these times. This limits understanding of atmospheric particle burden and its climate-relevant physicochemical properties, which have implications for the prediction of weather (Kawecki & Steiner, 2018), air quality, and climate. This indicates that the clear sky bias affects accurate representation of ALW on cloudy days and is suggestive that without in situ chemical information, aerosol-cloud interactions and subsequent estimates of radiative forcings in models (Lin et al., 2016; Vogelmann et al., 2012) will remain inaccurate. More detailed particle chemical composition is needed to accurately predict atmospheric particulate matter and subsequent impacts on weather, climate, and air quality.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements and Data

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Key Points

- Aerosol liquid water and particle chemical constituent mass concentrations significantly differ on cloudy and clear days across the CONUS
- Physical meteorology is insufficient to fully explain the differences in fine particle chemical composition
- Aerosol liquid water differences are partly determined by composition affecting hygroscopicity in many locations including the Mid South

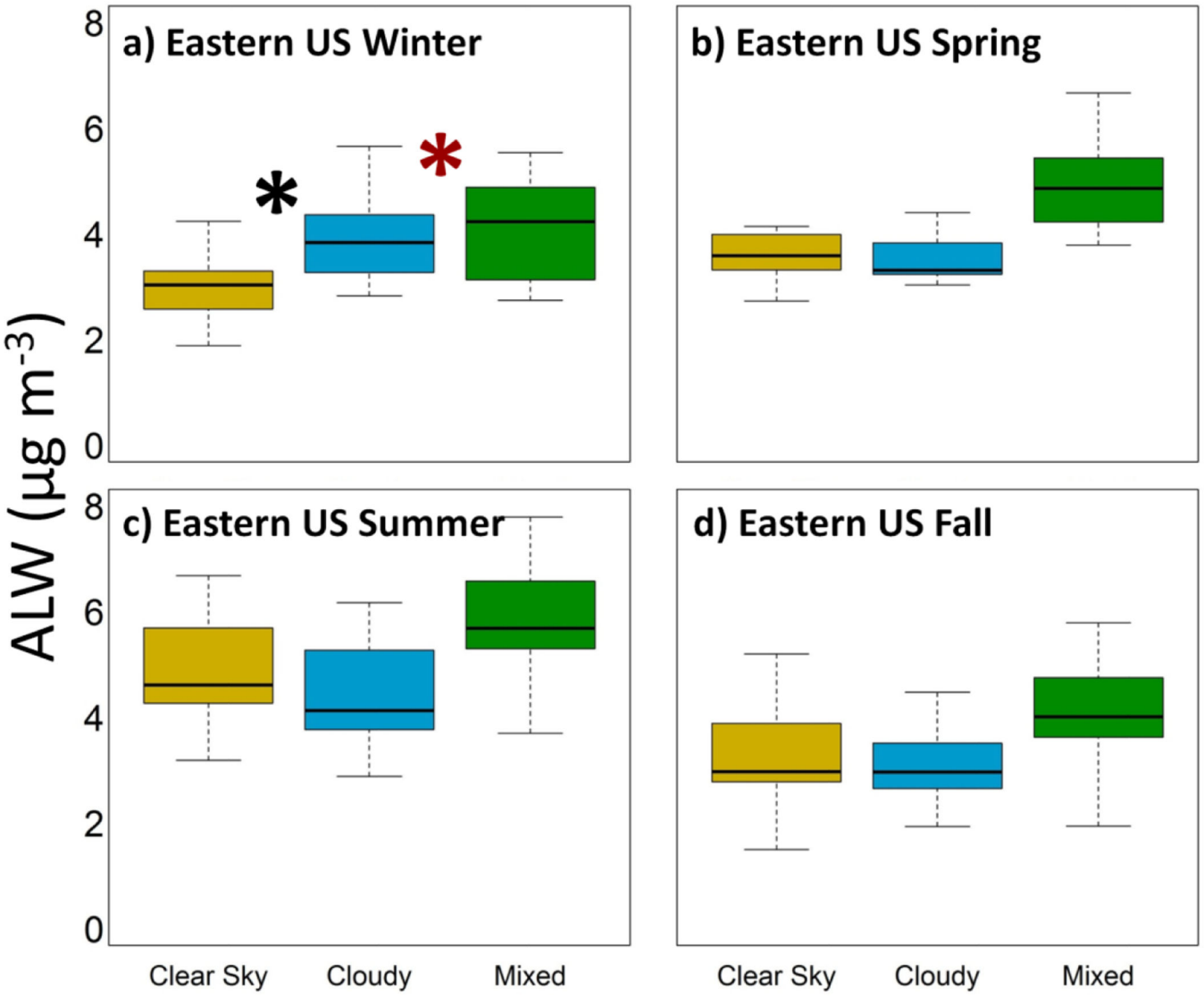

Figure 1.

ALW mass concentrations are significantly different between Clear Sky and Cloudy time periods beyond what would arise from changes solely in meteorology (e.g., RH). Monthly median estimated ALW distributions at each IMPROVE monitor in the eastern US during Clear Sky times (yellow, Clear Sky scenario), Cloudy times (blue, Cloudy scenario), and Cloudy times employing Clear Sky particle chemical composition (green, Mixed scenario). The black asterisk in (a) indicates the only situation where Clear Sky and Cloudy scenarios differ significantly. The red asterisk in (a) indicates the only situation where the Cloudy and Mixed scenarios do *not* differ significantly. The midline in the box is the median, the box boundaries are the 25th and 75th percentiles, and the whiskers are the 10th and 90th percentiles. Note that potential outliers are not shown but are used in calculations.

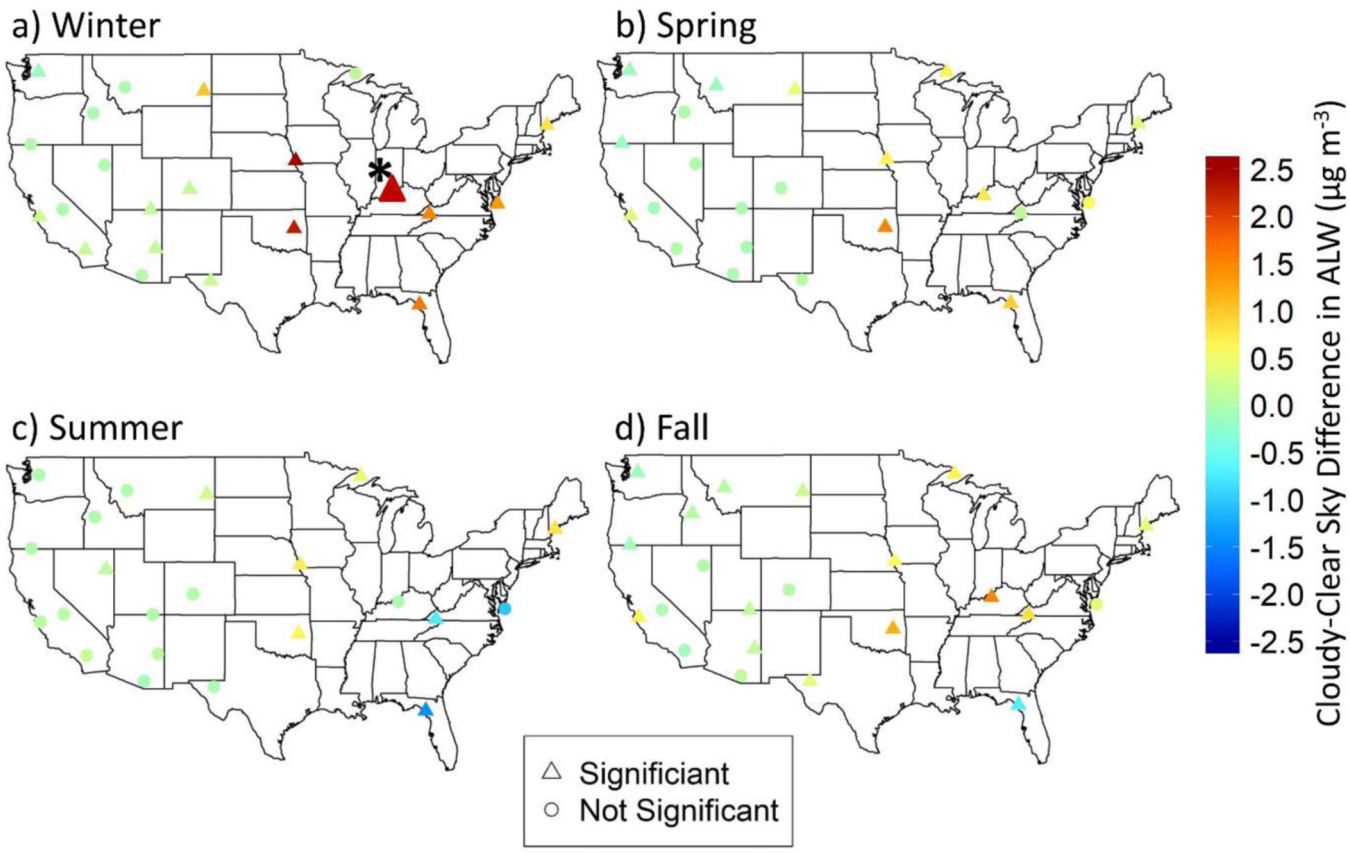

Figure 2.

Maps of the difference in ALW mass concentration medians (Cloudy-Clear Sky) for all regions from 2010–2014 for a) winter, b) spring, c) summer, and d) fall. The color of the point corresponds to the magnitude of the difference. Triangles indicate that median differences are significant by the Mann-Whitney U Test. Note that the difference in wintertime medians for daily ALW concentrations in the Ohio River Valley (denoted with asterisk) is substantially larger than other regions (Cloudy median value is 4.58 μg m^{-3} larger than Clear Sky).

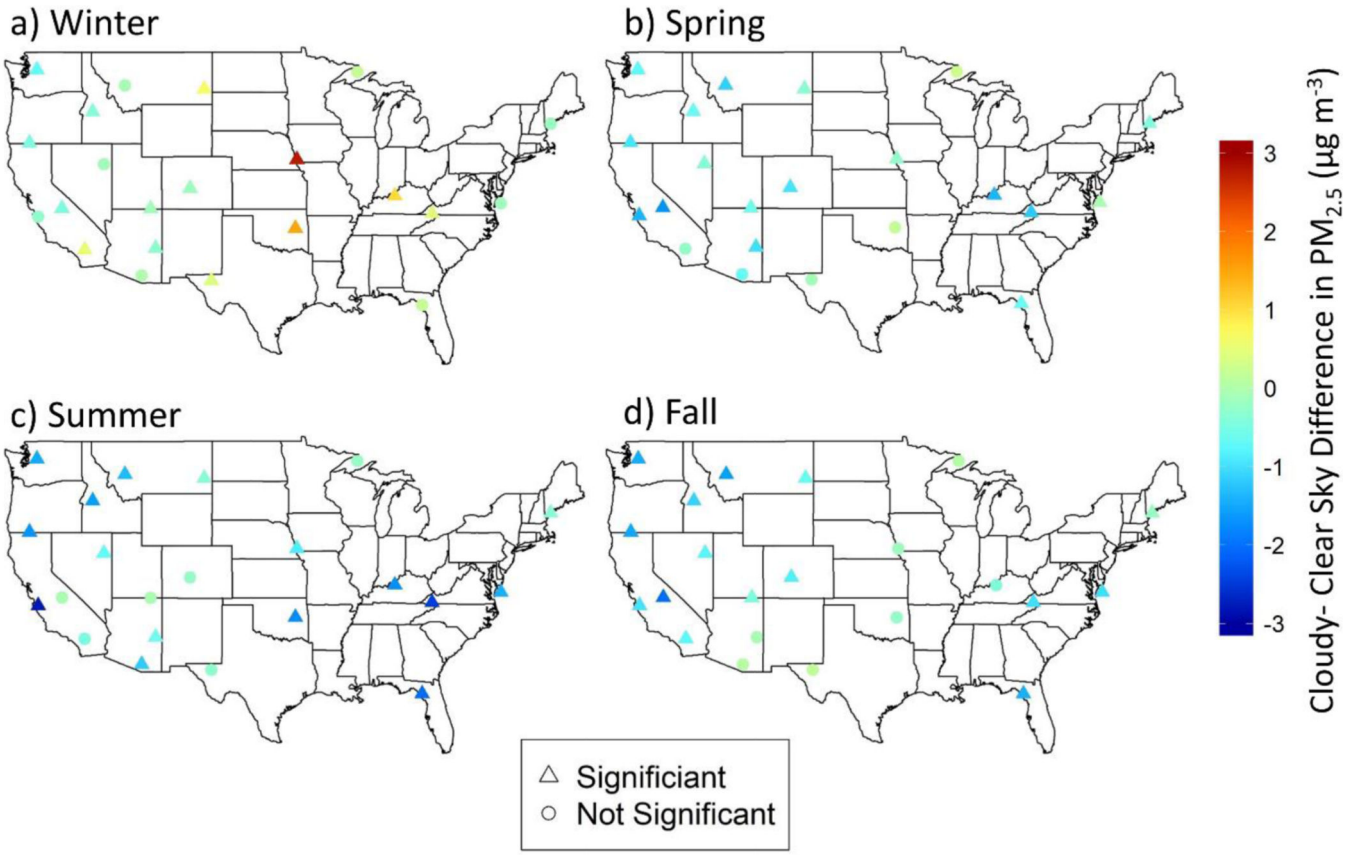

Figure 3.

Maps of the difference in $PM_{2.5}$ mass concentration medians (Cloudy-Clear Sky) for all regions from 2010–2014 for a) winter, b) spring, c) summer, and d) fall. The color of the point corresponds to the magnitude of the difference. Triangles indicate that median differences are significant by the Mann-Whitney U Test.

Table 1.

Particle chemical constituent concentrations, meteorology, and growth factors during Cloudy (Cl) and Clear Sky (CS) times in the Mid South.

	SO_4^{2-}		NO_3^-		ALW		RH		Growth Factors	
	CS	Cl	CS	Cl	CS	Cl	CS	Cl	CS	Cl
Win	0.77	1.24	0.90	1.22	1.32	3.61	0.64	0.80	1.33	1.50
Spr	1.46	1.79	0.37	0.50	2.48	4.02	0.62	0.76	1.25	1.41
Sum	1.91	1.69	0.20	0.19	2.92	3.57	0.59	0.72	1.21	1.39
Fall	1.05	1.17	0.18	0.33	1.56	2.74	0.57	0.73	1.18	1.37