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Significant Climate Impact of Highly Hygroscopic Atmospheric Aerosols in Delhi, India

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Key points:

1. For the first time, we estimate the hygroscopicity of aerosols in Delhi (κ=0.42±0.07), based on 3-year (2016-2018) ground observations.
2. Hygroscopicity of aerosols in Delhi is much higher than Beijing and Asian average, therefore leading to remarkable climate effects.
3. We demonstrate a valuable method for deriving bulk-averaged hygroscopicity of aerosol based on publicly available datasets.

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Abstract:

Hygroscopicity of aerosol (κ_{chem}) is a key factor affecting its direct and indirect climate effects, however, long-term observation in Delhi is absent. Here, we demonstrate an approach to derive κ_{chem} from publicly available datasets, and validate it (bias of 5-30%) with long-term observations in Beijing. Using this approach, we report the first estimation of κ_{chem} in Delhi and discuss its climate implications. The bulk-averaged κ_{chem} of aerosols in Delhi is estimated to be 0.42±0.07 during 2016-2018, implying a higher activation ability as cloud condensation nuclei in Delhi compared with Beijing and continental averages world-wide. To activate a 0.1 μm particle averagely requires just a supersaturation of ~0.18±0.015% in Delhi, but ~0.3% (Beijing), 0.28-0.31% (Asia, Africa and S. America) and ~0.22% (Europe and N. America). Our results imply that representing κ_{chem} of Delhi using Asian/Beijing average may result in a significant underestimation of aerosol climate effects.

Plain Language Summary:

Hygroscopic water uptake of aerosols can enhance its light extinction and cloud activation. Therefore, hygroscopicity of aerosol (κ_{chem}) is a key factor affecting its direct and indirect climate effects, however, long-term observation of κ_{chem} in Delhi is absent. Here, we demonstrate an approach to retrieve κ_{chem} from publicly available datasets of $PM_{2.5}$ and meteorology, and report the first long-term estimation of κ_{chem} in Delhi is 0.42±0.07 during 2016-2018. This value indicates only a supersaturation of ~0.18±0.015% is required to activate a particle with 0.1 μm diameter in Delhi, in contrast to ~0.3% supersaturation is required for Beijing and Asian average. It implies a higher water uptake and cloud activation ability for Delhi aerosols. Therefore, using Asian/Beijing averaged κ_{chem} to represent Delhi aerosols would lead to a significant underestimation of aerosol climate effects.

Keywords: long-term, kappa value, light extinction enhancement, $PM_{2.5}$, CCN

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1. Introduction

Aerosol particles in the atmosphere exert direct radiative forcing via scattering and absorbing solar radiation [*Charlson et al.*, 1992], also exert indirect radiative forcing and impact hydrologic cycle via serving as cloud condensation nuclei (CCN, [*Tang et al.*, 2016; *Twomey*, 1974; *Wex et al.*, 2007; *Wex et al.*, 2009; *Yin et al.*, 2000; *Zhao et al.*, 2006]). The influences of anthropogenic aerosols on these direct and indirect effects contribute the largest uncertainty in climate change assessment [*IPCC*, 2013; *Attwood et al.*, 2014; *Nemesure et al.*, 1995]. Hygroscopicity of aerosol, i.e., interaction between aerosol and atmospheric water vapor, is one of the most important factors affecting these uncertainties [*Kuang et al.*, 2016; *Zhao et al.*, 2006]. In addition to the climate concerns, hygroscopic water uptake increases aerosol water content as relative humidity (RH) increases. This can significantly influence the secondary particle formation [*Chen et al.*, 2018b; *Cheng et al.*, 2016; *Ervens et al.*, 2011; *Hennigan et al.*, 2008; *Wu et al.*, 2018], visibility [*Charlson et al.*, 1967; *Malm et al.*, 2000; *Mukherjee and Toohey*, 2016], aerosol optical depth and remote sensing measurements [*Brock et al.*, 2016a; *Brock et al.*, 2016b; *Crumeyrolle et al.*, 2014; *Esteve et al.*, 2012], as well as directly influence the measurements of aerosol loading and chemical compositions [*Chen et al.*, 2018a].

Traditionally, the hygroscopic property of aerosol can be described as the enhancement of light extinction/scattering [*Wright*, 1939] and the growth of geometrical size [*Köhler*, 1936] due to water uptake. The enhancement factor of aerosol light extinction/scattering coefficient (σ), defined as $f(RH) = \sigma(RH)/\sigma(RH_{ref})$, is a common way to describe aerosol hygroscopicity [*Titos et al.*, 2016; *Brock et al.*, 2016b]. In this definition, $\sigma(RH)$ and $\sigma(RH_{ref})$ represent the σ at a certain RH and at the reference RH in low/dry humid condition (RH_{ref}), respectively. Humidified nephelometer system is commonly used to directly measure f(RH) [*Covert et al.*, 1972; *Pilat and Charlson*, 1966]. In term of geometrical growth, *Petters and Kreidenweis*

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[2007] introduced the κ-Köhler theory to describe hygroscopic growth of particle diameter using a single parameter (κ), on the basis of the original Köhler theory [*Köhler*, 1936]. This single parameter represents the dependence of hygroscopicity on chemical composition of particles, referred to as κ_{chem} in the following. The κ_{chem} of a multicomponent particle can be calculated as volume-weighted average of each component, i.e., the Zdanovskii–Stokes–Robinson rule [*Stokes and Robinson*, 1966; *Zdanovskii*, 1948]. The parameter κ_{chem} is widely used in laboratorial, field-observational and modelling studies, because it harmonizes the comparisons of hygroscopicity derived from different techniques and environments. The parameter κ_{chem} can be derived from diameter growth factor measured by Hygroscopic Tandem Differential Mobility Analyser (HTDMA) or CCN activity following the κ-Köhler theory [*Liu et al.*, 2018; *Liu et al.*, 2011; *Petters and Kreidenweis*, 2007; *Wang et al.*, 2018; *Wex et al.*, 2010], and can also be calculated with measurements of chemical components [*Petters and Kreidenweis*, 2007]. A drawback of HTDMA method is missing the information of coarse particles [*Titos et al.*, 2016], which could be highly hygroscopic (e.g., sea salt) and greatly contribute to hygroscopic growth [*Chen et al.*, 2018a]. The previous closure studies usually show reasonable agreements between HTDMA-derived, CCN-derived and chemical-derived κ_{chem} values [*Hansen et al.*, 2015; *Wu et al.*, 2016; *Yeung et al.*, 2014]. The strong relationship between f(RH), hygroscopicity (κ_{chem}), particle composition and CCN activation has been investigated in lots of previous studies since the works of *Charlson et al.* [1967], *Covert et al.* [1972], *Ervens et al.* [2007] and *Pilat and Charlson* [1966].

Hygroscopicity (κ_{chem}) measurements have been carried out world-wide during the past two decades, the observational results are compiled in previous works [*Bhattu et al.*, 2016; *Kreidenweis and Asa-Awuku*, 2014; *Swietlicki et al.*, 2008]. Hygroscopicity of aerosols was mostly measured during short-intensive field campaigns due to high financial cost and complicated maintenance. A few previous long-term observational studies mainly focused on

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clean environments [*Fors et al.*, 2011; *Holmgren et al.*, 2014; *Kammermann et al.*, 2010] and one long-term study focused on Beijing [*Wang et al.*, 2018]. To the best of our knowledge, no long-term observation of aerosol κ_{chem} in Delhi and National Capital Region of India was reported. Given the intensive solar radiation and the strong influence of the South Asia monsoon over Indian subcontinent, aerosol hygroscopicity assessment, especially based on long-term observations, is urgent and critical for the studies of radiative forcing and hydrologic cycle.

In this study, we demonstrate an approach for assessing long-term bulk-averaged aerosol hygroscopicity, based on datasets publicly available in a large spatial and temporal coverage. The bulk-averaged κ_{chem} of aerosols in Delhi is reported based on 3-year (2016-2018) ground observations. The corresponding climate implications are also discussed. The approach demonstrated here is also valuable for studies in the other regions where high-quality long-term observations of aerosol hygroscopicity are not available.

2. Materials and Methods

2.1 Observations

$PM_{2.5}$ mass loading is measured by a beta attenuation monitor (BAM-1020, MetOne) at the U.S. Embassy in Delhi during 2016-2018. BAM is a U.S. EPA (Environmental Protection Agency) equivalent reference method for continuous $PM_{2.5}$ monitoring and is used for over 80% of the state and local level observations in U.S. [*EPA*, 2015; *Mukherjee and Toohey*, 2016]. $PM_{2.5}$ measured with BAM is not strongly influenced by aerosol associated water [*Mukherjee and Toohey*, 2016]. The instruments are well maintained and calibrated, details of instrument technique, operation and calibration are given in *EPA* [2009; 2015]. Hourly $PM_{2.5}$

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concentrations in Delhi are available from the AirNow platform (https://www.airnow.gov/) maintained by the U.S. EPA.

The hourly visibility and meteorological conditions are recorded at the Indira Gandhi International Airport (DEL) in Delhi. The hourly visibility is observed by a transmissometer (Drishti, CSIR-National Aerospace Laboratories, [*Khare et al.*, 2018]), which is well calibrated and performs well at the airport as reported by India Meteorological Department (http://metnet.imd.gov.in/mausamdocs/16644_F.pdf). RH is calculated as the ratio between water vapor pressure and saturation vapor pressure, which are respectively derived from dew-point temperature and temperature using the Magnus formula [*WMO*, 2008]. As one of the Integrated Surface Database (ISD) stations, the measurements at DEL are well calibrated and quality controlled according to the regulation of National Oceanic and Atmospheric Administration, National Climatic Data Center (NOAA-NCDC, [*Neal Lott*, 2004]). These datasets are available from the NOAA-NCDC website (https://www.ncdc.noaa.gov/).

A limited spatial inhomogeneity is expected in $PM_{2.5}$ concentrations and visibility between the U.S. Embassy and DEL. As shown in Fig. S1, the distance between them is only ~7 km, which is in the visibility measuring range. Furthermore, there is very slight variation in topography and anthropogenic $PM_{2.5}$ emission flux over the region between DEL and the U.S. Embassy in Delhi [*Marrapu et al.*, 2014; *Sahu et al.*, 2011].

2.2 Assessment of aerosol hygroscopicity

The f(RH) and κ_{chem} are parameters describing aerosol hygroscopicity. Here, we briefly describe the approach in this study for deriving f(RH) and κ_{chem} using publicly available long-term datasets. The approach consists of two steps. First, estimate bulk-averaged f(RH) as a function of RH from the datasets of $PM_{2.5}$ loading and meteorology [*Mukherjee and Toohey*, 2016]. Second, derive κ_{chem} from the function between f(RH) and RH [*Brock et al.*, 2016b;

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Kuang et al., 2017]. We firstly validate the approach by measurements in Beijing, where extensive datasets of field campaigns have been published in recent years. And then the approach is applied to conduct the first estimation of aerosol hygroscopicity in Delhi.

First step, a recent study [*Mukherjee and Toohey*, 2016] demonstrated a method to derive the bulk-averaged f(RH) based on publicly available datasets: i) $PM_{2.5}$ loading (units: [$\mu g/m^3$]) from U.S. Embassy, and ii) RH (units: [%]) and visibility (units: [km]) from NOAA-NCDC. The total light extinction coefficient can be derived using Koschmieder's equation from visibility [*Koschmieder*, 1924]. As shown in Eq. 1, the $PM_{2.5}$ associated extinction coefficient (σ_{PM}, with units of [km^{-1}]) can be estimated as total σ deducted by air extinction (σ_{air}) and other factors (σ_{other}). As recommended by *Mukherjee and Toohey* [2016]: i) a constant empirical factor σ_{other}=0.064 km^{-1} is adopted to represent the influences of gaseous pollutants and coarse particles; and ii) σ_{air}=0.056 km^{-1} is adopted in our study, corresponding to a maximum visibility of 70 km under clear-sky condition [*Mukherjee and Toohey*, 2016]. Therefore, the dataset consisting pairs of RH, $PM_{2.5}$ and σ_{PM} can be prepared for analysis. Although the value of σ_{other} is adopted from an estimation for Beijing [*Mukherjee and Toohey*, 2016], this only introduces uncertainty to κ_{chem} estimation by less than 5% in general (details in Text S1). In the study of *Mukherjee and Toohey* [2016], Beijing dataset during 2009-2014 was prepared and projected to 10 RH bins with 280-320 pairs per bin. The slope between σ_{PM} and $PM_{2.5}$ ($\sigma_{PM}/PM_{2.5}$ with units of [m^2/g]) can be obtained for each RH bin using least squares fit linear regression, referred to as slope(RH) in the following. The slope at RH_{ref} (median RH at the lowest RH bin) is used to assess dry mass extinction efficiency of $PM_{2.5}$. The ratios between slope(RH_{ref}) and the slopes of higher RH bins represent the enhancements of light extinction by aerosol liquid water. Finally, the unitless light extinction enhancement factors are derived by normalizing the slopes with slope at RH_{ref}, i.e., f(RH)=slope(RH)/slope(RH_{ref}). In our study and *Mukherjee and Toohey* [2016], we use

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median RH in the bin between 30-40% as $RH_{ref,}$, since *WMO/GAW* [2016] recommends a reference RH of 30-40% for nephelometer and 40% as a maximum RH for the sampling flow. *Mukherjee and Toohey* [2016] validated this approach with other independent observation-based estimations. The slope at RH_{ref} (3.7±0.4 m^2/g) is in a good agreement with an independent estimation [*Wang et al.*, 2015] using IMPROVE algorithms I (3.2 m^2/g) and II (4.1 m^2/g) [*Pitchford et al.*, 2007]. The derived f(RH) values are also in a good agreement with the estimations in other studies, details shown in the Figure 6d of *Mukherjee and Toohey* [2016].

Second step, we further derive κ_{chem} from f(RH), following the works of *Brock et al.* [2016b] and *Kuang et al.* [2017]. Recently, *Brock et al.* [2016b] proposed a single parameter ($\kappa_{opt,}$ refer to κ value directly derived from optical method/datasets) to describe f(RH), and *Kuang et al.* [2017] further developed this parameterization with RH_{ref} included, as shown in Eq. 2. They demonstrated that κ_{opt} can better describe f(RH) than the widely used 'gamma' power-law approximation [*Kasten*, 1969]. Following the works of *Brock et al.* [2016b] and *Chen et al.* [2014], which are based on κ-Köhler and Mie theories, *Kuang et al.* [2017] proposed a physically based approach to derive the equivalent κ_{chem} from κ_{opt} with R^2=0.97. The derived κ_{chem} values ($\kappa_{f(RH)}$ in *Kuang et al.* [2017]) agree well (R^2=0.77) with measurements in Beijing using HH-TDMA, which is similar to HTDMA with capability of operating under higher RH. The ratio between κ_{opt} and κ_{chem} (R_κ) is influenced by particle number size distribution (PNSD) and chemical composition to some extent. R_κ is in a range of 0.58-0.77 (0.69 on average) based on Beijing observations [*Kuang et al.*, 2017]. Furthermore, they simplified the influences of PNSD and chemical composition on R_κ as a function of Ångström exponent and $\kappa_{opt,}$ and provided a 2-D look-up table for R_κ (Fig. S2).

To validate our approach for deriving κ_{chem} from datasets of $PM_{2.5}$ loading and meteorology, we estimate a bulk-averaged κ_{chem} of 0.18-0.24 (0.2 on average, considering the

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variation of R_κ) using the estimated f(RH) values in Beijing 2014, which is adopted from *Mukherjee and Toohey* [2016]. Our results agree well with a long-term observation of κ_{chem} in Beijing 2014 [*Wang et al.*, 2018]. They conducted a 9-month HTDMA field measurement and reported that the averaged κ_{chem} in Beijing is in a range of 0.14-0.23 for dry particles with diameters of 50-350 nm, details in the Table 2 of *Wang et al.* [2018]. An increase of κ_{chem} as particle size increases was found in their study. This may explain the slight overprediction of κ_{chem} (bias of 0.01-0.04, about 5-30%) in our approach. Since, HTDMA can only measure the κ_{chem} of particles at a certain size (usually smaller than 350 nm), however, our approach estimates a bulk κ_{chem} of the whole $PM_{2.5}$ population. These results strongly suggest that the approach we demonstrated here can estimate κ_{chem} value in a reasonable range.

$$\sigma_{PM} = \frac{3.912}{Visibility} - \sigma_{air} - \sigma_{other} \quad (1)$$

$$f(RH) = \frac{1 + \kappa_{opt}\frac{RH}{100 - RH}}{1 + \kappa_{opt}\frac{RH_{ref}}{100 - RH_{ref}}}, \qquad \kappa_{chem} = \frac{\kappa_{opt}}{R_\kappa} \quad (2)$$

where f(RH), κ_{chem}, κ_{opt} and R_κ are unitless variables.

The $PM_{2.5}$ and meteorological datasets during 2016-2018 in Delhi are used in this study for the assessment of κ_{chem}. We conduct the analysis using the visibility records in the range of 0-9 km, as recommended by *Mukherjee and Toohey* [2016]. This makes the analysis of f(RH) more reliable, since all visibility with values greater than 10 km are recorded as 10 km. The data pairs with wind speed larger than 6.5 m/s [*Kurosaki and Mikami*, 2007; *Tegen and Fung*, 1994; 1995] alongside $PM_{2.5}$ concentration higher than 500 $\mu g/m^3$ are excluded from analysis to minimize the uncertainties induced by dust. Additionally, we exclude the period with RH higher than 90%. This can minimize the uncertainties from noise signals caused by fog, cloud, precipitation and low accuracy of RH-senser under high RH conditions. We project the data pairs of RH, $PM_{2.5}$ and σ_{PM} to 8 RH bins (with borders of 30%, 40%, 50%,

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60%, 70%, 75%, 80%, 85% and 90%), and estimate the bulk-averaged f(RH) of each RH bin where more than 300 pairs of data are available. Then κ_{opt} can be derived from the function between f(RH) and RH (Eq. 2), and κ_{chem} can be estimated as R_κ is given. We identify the R_κ value for Delhi using the 2-D look-up table (Fig. S2, [*Kuang et al.*, 2017]), and perform Monte Carlo calculation (1 million random samples) to estimate the uncertainties of R_κ (Fig. S3a) and κ_{chem} (Fig. S3b). Uncertainty estimation is detailed in Text S2 (long-term Ångström exponent for Delhi refer to *Lodhi et al.* [2013]). Finally, the potential of CCN activation in Delhi is estimated using κ_{chem} and κ-Köhler theory [*Petters and Kreidenweis*, 2008].

3. Results and Discussion

As shown in Fig. 1, increased $PM_{2.5}$ loading and RH can lead to higher light extinction. The σ_{PM} shows a clear increase trend with increase of $PM_{2.5}$, and also progressively increases as increase of RH for a given $PM_{2.5}$. This is because hygroscopic growth of particle significantly enhances the light extinction. In order to estimate this enhancement effect, we derive the f(RH) as a funtion of RH (see Method) as shown in Fig. 2. The long-term bulk-averaged f(RH) monotonically increases with RH in general. The shapes of f(RH) curves are similar for each year during 2016-2018. In line with the works of *Brock et al.* [2016b] and *Kuang et al.* [2017], the pattern of f(RH) follows Eq. 2 well with R^2>0.95. The f(RH=80-85%) in Delhi is in the range of 1.7-2.3 during 2016-2018, with an average of ~2.0. This light extinction enhancement factor in Delhi is higher than the values measured in urban and rural regions of Beijing, where show a f(RH=80-85%) of 1.3 in the clean conditions and 1.5 in the polluted conditions [*Titos et al.*, 2016]. But the f(RH=80-85%) in Dehi is lower than the values measured over clean marine environments, e.g., NY-Alesund (2.5-3.8), east Asia (2.2-2.8, clean) and Cabauw (~3.5, clean); whereas, similar to the polluted or dust dominant marine environments, e.g., Gosan, Jeju Island in Korea (1.8-2.2, dust), east Asia (2.0-2.3,

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polluted) and Cabauw (~2.0, polluted) [*Titos et al.*, 2016]. These results indicate that the urban pollutants may moderate the hygroscopicity of marine aerosols, however, may enhance the hygroscopicity over inland regions, such as Delhi and Beijing. The higher hygroscopicity of aerosols in Delhi may also imply a more severe anthropogenic pollution than Beijing. This is in line with the database of WHO (http://www.who.int/airpollution/data/cities/en/) which shows a twice higher $PM_{2.5}$ loading in Delhi compared with Beijing. Furthermore, lots of previous studies (e.g., [*Titos et al.*, 2016; *Wang et al.*, 2007; *Zhang et al.*, 2015]) reported that f(RH=80-85%) is inversely propotional to the mass fration of organic matter (F_{OM}). Higher f(RH=80-85%) in Delhi may indicate a lower F_{OM} compared to Beijing. This is consistent with a recent long-term observational study in Delhi [*Sharma et al.*, 2018], which reported an annual averaged F_{OM} in $PM_{2.5}$ is in a range of 15-20% during 2012-2016 (mass of organic matter is usually calculated as 1.4 times of organic carbon). However, the F_{OM} in Beijing is usually in a range of 20-40% [*Hu et al.*, 2015; *Huang et al.*, 2014; *Tao et al.*, 2017; *Yang et al.*, 2017], where more than half of the organic matter originates from secondary organic aerosol (SOA) [*Hu et al.*, 2015; *Huang et al.*, 2014; *Jimenez et al.*, 2009]. Stronger solar radiation in Delhi may increase photochemical reactions and oxidation of volatile organic compounds, therefore may enhance SOA formation [*Hu et al.*, 2019; *McFiggans et al.*, 2019; *Zhang et al.*, 2015; *Guo et al.*, 2014; *Zhu et al.*, 2011]. However, hotter weather in Delhi compared with Beijing could suppress the condensation of semi-volatile organic compounds and compensate the enhancement of SOA formation. The lower F_{OM} in Delhi may be due to less SOA, resulting from the competition between the two effects above; however, more observational evidences are required. Moreover, in constract to the rapid decrease of SO_2 emission in China over the past decade, the significant increase of SO_2 emission in India [*Li et al.*, 2017] could lead to a great formation of highly hygroscopic particulate sulfate. This could be another reason of higher hygroscopicity and larger light extinction enhancement of

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aerosol in Delhi than in Beijing. The intensive field measurements of physicochemical properties of particulate matter and gaseous pollutants are scarce in Delhi, we hightlight the urgence of these observational studies for better understandings of physical and chemical properties of aerosols in Delhi.

To facilitate the assessment of climate impact and comparison with other studies, we derive the κ_{chem} of aerosols in Delhi from f(RH) using Eq. 2. The annual bulk-averaged κ_{chem} in Delhi is about 0.42±0.07 during 2016-2018. In line with above discussion, this value indicates higher (by ~100%) hygroscopicity in Delhi than in Beijing. The long-term HTDMA field observation in Beijing reports an averaged κ_{chem} in the range of 0.14-0.23 for particles within a size range of 50-350 nm [*Wang et al.*, 2018]. Given the absence of direct hygroscopicity measurements in Delhi, we compare our observation-based estimation with a global model study [*Pringle et al.*, 2010]. They show reasonable model results, with deviations between the modelled and observed κ_{chem} values less than 0.05 at 10 out of the 14 locations over the world. In line with our study, their model result of κ_{chem} in Delhi is about 50-100% higher than the result in Beijing. Our estimated κ_{chem} in Delhi is much higher than averaged values of Asia (0.22), Australia (0.21), S. America (0.17) and Africa (0.15), howerver, much lower than the averaged values of N. Atlantic (0.59) and Southern Ocean (0.92) [*Pringle et al.*, 2010]. The κ_{chem} in Delhi is much higher (by about 100%) than Asian averages and Beijing observations. As discussed above, this is possibly resulting from less SOA or abundant anthropogenic sulfate aerosol in Delhi, which is also implied by *Pringle et al.* [2010].

4. Implication of Finding

Cloud formation exerts a significant impact on the radiative balance of the earth system (indirect radiative forcing) and hydrologic cycle. Cloud droplet number plays a crucial role in

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determining albedo and lifetime of cloud [*Ming et al.*, 2006], and is very sensitive to κ_{chem} [*Reutter et al.*, 2009]. To further investigate the impact of κ_{chem} on aerosol-cloud interaction, we estimate the CCN activation ability of aerosols in Delhi using κ_{chem} following the works of *Petters and Kreidenweis* [2007; 2008], and compare it with the activation ability of other regions over the world and some typical constituents of atmospheric relevance (Fig. 3). It is worth noting that κ_{chem} can be size-dependent, bulk-averaged κ_{chem} values are adopted and could introduce uncertainty in the following estimation. Long-term size-resolved particle hygroscopicity observations are required in future studies to quantify this uncertainty. The activation ability of aerosols in Delhi is much higher than some organic matters of atmospheric relevance, e.g., oxidized dihexylethyle sebacate, fractionated fulvic acid, fulvic acid, mixture of levoglucosan with succinic and fulvic, and pure levoglucosan (Fig. 3a) [*Svenningsson et al.*, 2006]. However, the activation ability is lower than some typical inorganic matters of atmospheric relevance, e.g., ammonium nitrate (Fig. 3a). The activation ability of aerosols in Delhi is close to continental polluted aerosol represented by a mixture of inorganic (70%) and organic matters (30%), detailed information of mixture is given in *Petters and Kreidenweis* [2007] and *Svenningsson et al.* [2006]. This result may imply that the aerosol in Delhi is a mixture containing majority of inorganic and minority of organic species, and this is consistent with long-term measurements in Delhi [*Khare et al.*, 2018; *Sharma et al.*, 2018]. In order to emphasize the importance of climate impacts of aerosols in Delhi (Fig. 3b), we compare its activation ability with averaged values of Beijing [*Wang et al.*, 2018] and continental averages world-wide [*Pringle et al.*, 2010]. A 0.1 μm particle can activate as a cloud droplet under a supersaturation of ~0.22% for Europe and North America, about 0.28-0.31% for Asia, Australia, South America and Africa, and ~0.3% for Beijing. However, only a supersaturation of ~0.18±0.015% is required to activate 0.1 μm particles in Delhi on average. To activate a smaller particle possessing a diameter of 0.05 μm requires a

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supersaturation of ~0.51±0.04% (Delhi), ~0.70% (Europe and North America), 0.80-0.92% (Asia, Australia, South America and Africa), and ~0.85% (Beijing), respectively. Therefore, the CCN activation ability of aerosols in Delhi is much higher than the continental averages and another Asian megacity, Beijing. This indicates a larger impact of aerosols in Delhi on climate and hydrologic cycle, even if under same meteorologic conditions and same particle number concentration. Additionally, the frequent influence of monsoon and great $PM_{2.5}$ loading in Delhi make its climate impacts more remarkable (~125 μg/m^3 on average during 2016-2018, and ~110 μg/m^3 in 2015 as details given in Fig. S1 [*van Donkelaar et al.*, 2015]). Our results imply that using Asian average or measurements in other Asian megacities (e.g., Beijing) to represent the κ_{chem} in Delhi would lead to significant underestimation of its climate impacts.

Various parameterizations of cloud droplet nucleation are applied in general circulation models (GCMs) (e.g., [*Ghan et al.*, 2011; *Jiang et al.*, 2010; *Jiang et al.*, 2012; *Roelofs et al.*, 2006; *Zhang et al.*, 2016]). Some earlier cloud microphysical schemes empirically diagnose cloud droplet number concentration from aerosol mass (e.g., [Boucher and Lohmann, 1995; Lohmann and Feichter, 1997; Menon et al., 2002]) or aerosol number (e.g., [Gultepe and Isaac, 1996]) to account for aerosol-cloud interaction. However, these empirical relationships can vary largely over different regions [Ramanathan et al., 2001] and lead to substantial uncertainty. Later on, various Köhler-theory-based [Köhler, 1936] parameterizations (e.g., [Abdul-Razzak and Ghan, 2000; Fountoukis and Nenes, 2005; Ming et al., 2006; Nenes and Seinfeld, 2003]) have been applied in GCMs. For example, CMIP5-cm3 (Coupled Model Intercomparison Project, [Jiang et al., 2012; Ming et al., 2006]), CAM5 (Community Atmosphere Model, [*Abdul-Razzak and Ghan*, 2000; *Zhang et al.*, 2016]), UKCA (UK Chemistry and Aerosols community model, [Abdul-Razzak and Ghan, 2000; West et al., 2014]) and MRI-ESM1 (Meteorological Research Institute Earth System Model Version 1,

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[Abdul-Razzak and Ghan, 2000; Yukimoto et al., 2012]). However, the chemical complexity has an important impact on cloud activation and incorporating such complexity into these parameterizations is difficult [Fountoukis and Nenes, 2005]. By introducing a new concept of 'population splitting', *Fountoukis and Nenes* [2005] and *Nenes and Seinfeld* [2003] take the soluble, slightly soluble, insoluble species and organic surfactants into consideration. Recently, *Chang et al.* [2017] applied a κ-Köhler-based [Petters and Kreidenweis, 2007] parameterization in EMAC (ECHAM5-MESSy Atmospheric Chemistry model) to diagnose cloud activation efficiently and robustly, with consideration of aerosol chemical complexity but without the need of aerosol-specific information (e.g., Van't Hoff factor and osmotic coefficient), which is required by Köhler-theory-based parameterizations. *Chang et al.* [2017] shows that using the model predicted region-dependent κ values can improve the cloud and climate simulations over polluted regions (e.g., India), compared with using prescribed κ value (continental average value). This indicates the regional variation of κ can substantially influence climate simulation, and using our long-term observation-based κ estimation to constraint climate models would improve the assessment of climate change.

5. Summary

Hygroscopicity of aerosol is an important parameter affecting its climate effects, however, the long-term observation of it in Delhi, one of the biggest cities in the world, is absent. In this study, we demonstrate an approach to derive the hygroscopicity (κ_{chem}) of aerosol in Delhi from publicly available datasets. This approach is well validated, and shows a good agreement (bias of 0.01-0.04, 5-30%) with long-term observations in Beijing.

We analyze the Delhi observations during 2016-2018, and estimate a long-term bulk-averaged κ_{chem} of 0.42±0.07. This value is much higher (by about 100%) than the κ_{chem} of

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Beijing as reported from previous modelling and observational studies. This implies the difference in aerosol chemical composition between these two Asian megacities, Delhi and Beijing. The possible reasons could be higher contribution from anthropogenic sulfate or lower contribution from SOA in Delhi; however, further evidences are still needed from direct measurements. To activate particles of 0.1 μm (0.05 μm) as cloud condensation nucleis, a supersaturation of ~0.18±0.015% (0.51±0.04%) is required in Delhi, which is much lower than that in Beijing and the Asian average. Furthermore, the hygroscopicity-induced light extinction enhancement of aerosols in Delhi, i.e., f(RH=80-85%), is estimated to be in the range of 1.7-2.3, which is much higher than Beijing (1.3-1.5). The higher light extinction enhancement and easier cloud activation imply larger direct and indirect radiative forcing of aerosols in Delhi. These climate effects can be significantly underestimated if a hygroscopicity of Beijing or Asian average is used to represent the condition of Delhi. We highlight the urgence of direct hygroscopicity measurements in Delhi for a deeper understanding of human's influences on cloud formation, climate change and global hydrologic cycle. The approach we demonstrated in this study is also valuable for estimating aerosol hygroscopicity and its climate effects in other parts of the world where high-quality direct measurements are not available.

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Author contributions

Y. C. conceived the study. Y. C. and Y. W. performed the analysis and interpreted the results. All authors discuss the results and co-write the manuscript.

Notes

The authors declare no competing financial interest.

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The archived hourly measurements of $PM_{2.5}$ recorded at the U.S. Embassy in Delhi are available through the AirNow platform maintained by the U.S. Department of State and the U.S. Environmental Protection Agency at https://www.airnow.gov/. The archived hourly measurements of visibility and meteorology variables recorded at the Delhi Indira Gandhi International Airport are available through the Integrated Surface Database—Surface Data Hourly Global data product maintained by the U.S. National Oceanic and Atmospheric Administration—National Climatic Data Center at https://www.ncdc.noaa.gov/. Y. W. would like to thank the support of the joint scholarship of China Scholarship Council and University of Manchester. Y. C. would like to thank the project funded by NERC, UK (NE/P01531X/1). The paper is based on interpretation of scientific results and in no way reflect the viewpoint of the funding agencies.

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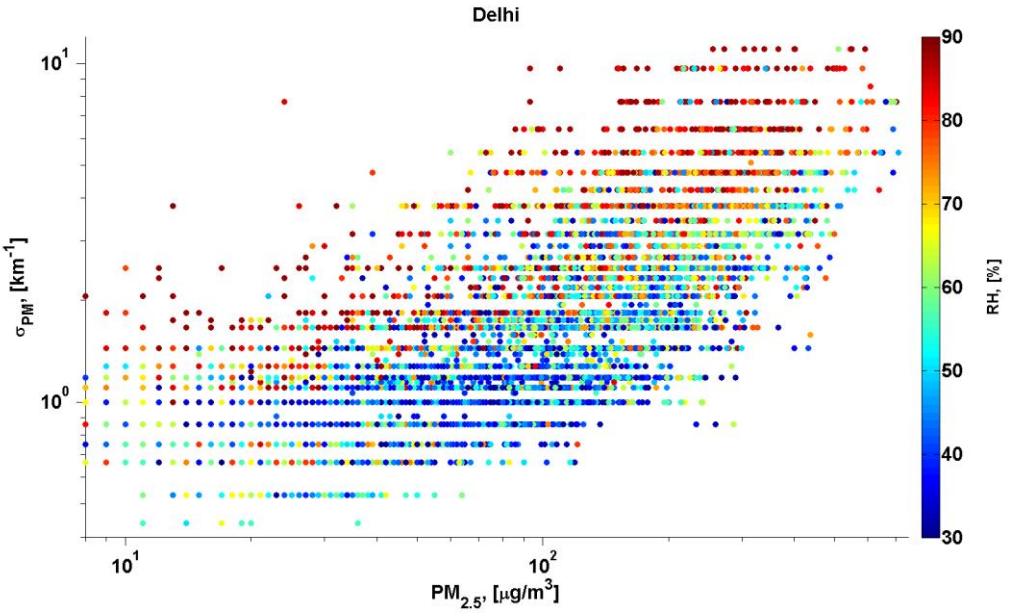

Figure 1. Light extinction efficiency as a function of $PM_{2.5}$ loading, colored by RH. Hourly values during 2016-2018 are presented.

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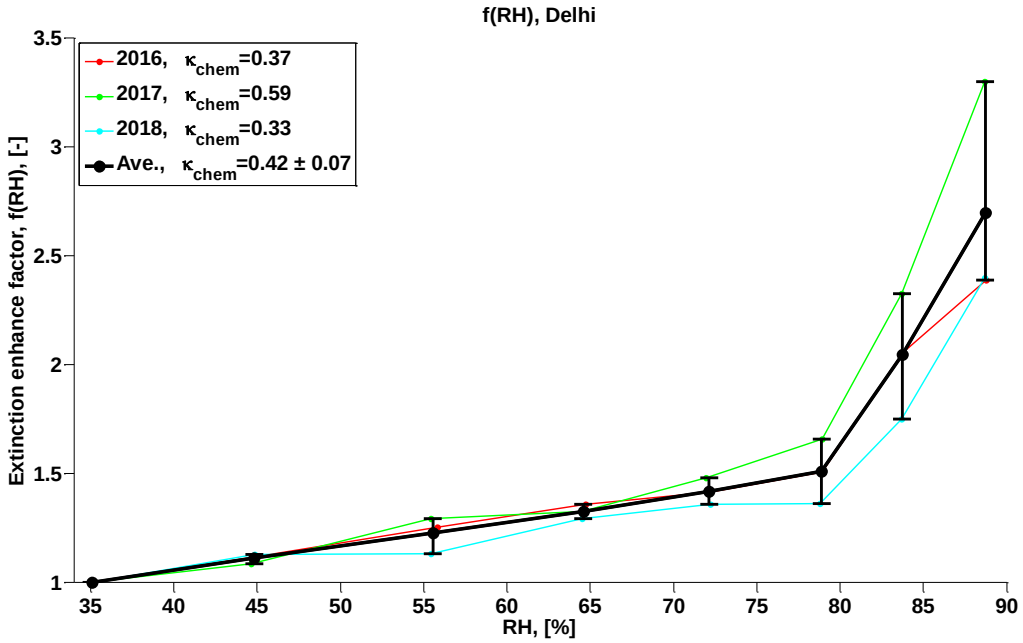

Figure 2. Estimation of κ_{chem} and light extinction enhancement factor as a function of RH. The hourly values are projected to 8 RH bins (see Method) for analysis. The annual bulk-averaged values are presented. The black line shows the mean results with variation range indicated by error bars. The estimated κ_{chem} (average ± standard deviation) are marked, uncertainty estimation is detailed in Text S2.

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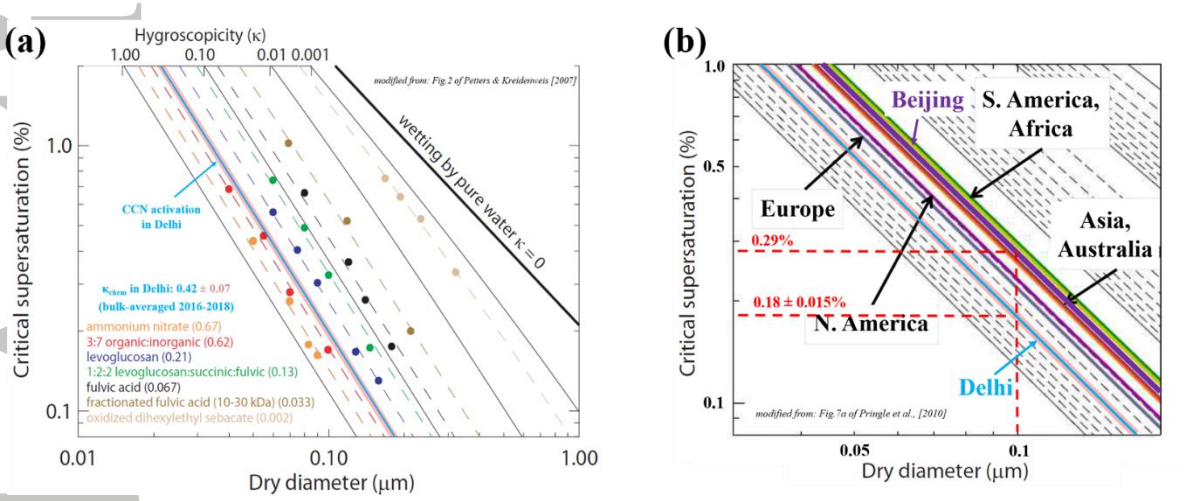

Figure 3. Critical supersaturation for cloud condensation nuclei (CCN) activation as a function of particle dry diameter. The estimated bulk-averaged values for Delhi (blue line with standard deviation in the pink shading area) is compared with values of other compounds (**a**) and values of continental regions world-wide (**b**). Panel (a) is modified from the figure 2 of Petters & Kreidenweis [2007], dots indicate the experiment results taken from literatures therein and the dashed lines indicate the best fit for each particle type. Panel (b) is modified from the figure 7a of Pringle et al. [2010]. The figures are reused under the CC Attribution 3.0 License.

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