Aerosol Total Volume Estimation From Wavelength- and Size-Resolved Scattering Coefficient Data: A New Method

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Abstract While the importance of supermicron particles on human health and climate is well recognized, knowledge of their size-related properties remains elusive. Many routine near-surface in situ measurements of aerosol properties include size spectra of submicron particles and aerosol total scattering coefficient at three visible wavelengths and two size cutoffs. These properties are collected, for example, at the U.S. Department of Energy’s Atmospheric Radiation Measurement (ARM) user facility. Our study illustrates how these conventional measurements can be used to predict total particle volume (particle size <10 μm). The well-known fact that small and large particles scatter sunlight very differently forms the basis of a new method. Our study demonstrates a good agreement between estimated and measured total volumes for five climate-important locations. The agreement suggests that the new method can be used to predict the total particle volume from the routine data collected at numerous sites around the world.

Plain Language Summary Particles in the air are astonishingly diverse in terms of size. Determining their size distribution, which characterizes the number of particles for every size of interest, is a difficult technical problem due to an extremely wide range of sizes (several orders of magnitude) and lack of a single instrument that can measure over the entire size range. For example, most conventional particle sizing techniques cannot measure large particles (size >1 μm) despite their recognized importance for human health, environment, and climate. Here, a new method is introduced to indirectly estimate the volume of large particles, which is closely related to their size distribution. This method combines (1) routinely measured size distributions of small particles and (2) the well-known fact that small and large particles scatter sunlight very differently as a function of wavelength. The performance of this method is demonstrated for data collected at five sites characterized by continental, alpine, coastal, and marine environments. The good agreement between the estimated and the measured values of the total volume of small and large particles demonstrates the credibility of this method and highlights its potential application to routine aerosol measurements made at numerous locations around the world.

1. Introduction

Supermicron particles have several essential impacts on human health, environment, and climate (e.g., Atwood et al., 2017; Feng et al., 2017; Hand et al., 2017; Mahowald et al., 2014). Moreover, concentration of supermicron particles appears to exceed substantially the air-quality guidelines over several regions in the future due to a changing climate (e.g., Monteiro et al., 2018). The comprehensive global study of supermicron particles is hindered by the lack of size-resolved measurements at established air-quality networks supported by international and national organizations (e.g., Vahlsing & Smith, 2012), including the United Nations Environment Programme and the United States Environmental Protection Agency (US EPA), and the relatively sparse coverage of these sites. These routine air-quality data are complemented by measurements of surface-level aerosol properties at instrumented sites focusing on other aspects of the atmospheric research. For example, the U.S. Department of Energy’s Atmospheric Radiation Measurement (ARM) user facility (e.g., Mather & Voyles, 2013; Miller et al., 2016) and many Global
Atmosphere Watch stations (e.g., Andrews et al., 2019) measure aerosol size distributions of submicron particles (PM$_{10}$, particulate matter with aerodynamic particle diameter smaller than 1 μm) and total scattering coefficients (SCA$_1$ and SCA$_{10}$) measured for two size cutoffs (aerodynamic particle diameters below 1 and 10 μm) and at three visible wavelengths (0.45, 0.55, and 0.7 μm).

Measurements of the size spectra and scattering of both submicron and supermicron particles (PM$_{10}$, particulate matter with aerodynamic particle diameter smaller than 10 μm) are less common relative to those for submicron particles mostly due to well-known sampling issues associated with supermicron particles (Baron & Willeke, 2001). It should be emphasized that PM$_{10}$ volume is proportional to PM$_{10}$ mass, which represents one of the main health and environmental concerns (e.g., Barthel et al., 2019; Rodriguez et al., 2019). Important qualitative information on the supermicron particles can be provided by the wavelength- and size-resolved values of the total scattering coefficient (e.g., Hansen & Travis, 1974; Schuster et al., 2006).

To illustrate this, a strong spectral dependence of SCA$_{10}$ suggests that the light scattering is dominated by submicron particles, while the lack of a wavelength dependence of SCA$_1$ is a good indicator of a large contribution of supermicron particles to the light scattering. The same is true for the size-resolved values of the total scattering coefficient. When comparable values of SCA$_1$ and SCA$_{10}$ at a given wavelength are observed, it suggests that the light scattering is dominated by submicron particles, while SCA$_{10}$ values significantly larger than SCA$_1$ indicate that supermicron particles play a major role in the light scattering.

The outlined qualitative relationship between submicron and supermicron particles and the wavelength- and size-resolved values of the total scattering coefficient is well understood. However, there is a large gap between such understanding and practical quantitative application of this relationship. The main goal of our study is to bridge the gap by taking advantage of available data sets of aerosol size spectra and total scattering coefficients. The selected data sets represent short-term integrated measurements at five sites with distinct aerosol types and environmental conditions. The two major objectives of our study are to (1) introduce a new method for predicting PM$_{10}$ volume using two parameters describing the wavelength- and size-resolved measurement of total scattering coefficient and PM$_1$ volume calculated from measured size spectra and (2) demonstrate an initial application of this method for five representative sites.

## 2. Approach

There are two main challenges associated with estimating aerosol volume from the light scattering data. First, the aerosol volume defines the third moment of the aerosol size distribution, while the scattering coefficient is proportional to the second moment of the same distribution (e.g., Sayer et al., 2012). Thus, the aerosol volume and total scattering have different sensitivity to the fraction of supermicron particles. This fraction can modify the spectral behavior of the total scattering coefficient significantly (e.g., Hansen & Travis, 1974; Schuster et al., 2006). Second, the scattering coefficient is influenced by the complex refractive index (RI), which can exhibit strong spectral dependence on aerosol type, mixing state, and composition (e.g., Eck et al., 1999). Therefore, the spectral behavior of the total scattering coefficient is governed by combined impact of two factors considered here (the RI and fraction of supermicron particles). As a result, these two factors must be considered for accurate interpretation of the light scattering spectral behavior.

These challenges suggest that the relationship between aerosol total volume and light scattering data is complicated. Nevertheless, it may still be possible to approximate this relationship by a set of statistical models and demonstrate their practical value as a useful predictive tool. For example, a regression model could express one parameter (the response variable), describing total volume of submicron and supermicron particles, as a function of two other parameters (the independent variables), expressing the wavelength- and size-dependent values of the total scattering coefficient. With this assumption in mind, we introduce the following three dimensionless parameters. The first parameter (the response variable) is a ratio of coarse and fine aerosol volumes:

\[
PM_{\text{ratio}} = (\text{PM}_{10,v} - \text{PM}_{1,v})/\text{PM}_{1,v}
\]

where PM$_{10,v}$ and PM$_{1,v}$ are volumes of PM$_{10}$ and PM$_1$, respectively. The prediction of PM$_{10,v}$ is the main goal of our approach. The second parameter (the first independent variable) is a ratio of total scattering coefficients measured at two wavelengths:
SCA_ratio = SCA_{10 \ (0.45 \ \mu m)} / SCA_{10 \ (0.7 \ \mu m)}   \ \ \ \ (2)

This parameter defines the wavelength dependence of the total scattering coefficient for a given size cutoff (aerodynamic particle diameters below 10 \ \mu m). Although the corresponding Ångström exponent can also be applied for describing this wavelength dependence (e.g., Delene & Ogren, 2002), such an alternative is not considered in our initial analysis. It should be noted that the SCA_ratio represents a quite narrow spectral range (0.45–0.7 \ \mu m); thus, it is mostly sensitive to the submicron particles (e.g., Atkinson et al., 2018). In other words, the SCA_ratio alone is not well suited for describing a link between the light scattering and PM_{10,v}. To address this limitation, we include the third parameter (the second independent variable). This parameter is a coarse-mode fraction (CMF) of the total scattering coefficient at a given wavelength:

CMF_{SCA} = (SCA_{10} - SCA_{1}) / SCA_{10}  \ \ \ \ (3)

Two parameters (SCA_ratio and CMF_{SCA}) incorporate both wavelength and size dependence of the total scattering coefficient required for the PM_{10,v} prediction (section 4).

The proposed PM_{10,v} prediction includes three steps. The first step is to establish a statistical relationship between the response variable and other two independent variables using a regression model: 

PM_{ratio} = \alpha + \beta \times SCA_ratio, where the regression coefficients \alpha and \beta depend on CMF_{SCA} (section 4).

To do this, we utilize data sets of fine and coarse aerosol size spectra and total scattering coefficient collected at five climate-important locations (section 3). The second step is to apply the established relationship and the independent variables (SCA_ratio and CMF_{SCA}) for estimating the response variable (PM_{ratio}). The third step is to calculate the PM_{10,v} from the estimated PM_{ratio} and the complementary measurement-based PM_{1,v} as PM_{10,v} = PM_{1,v} \times (PM_{ratio} + 1) (section 4).

3. Observations

We use data collected during four Intensive Observational Periods (IOPs) supported by the ARM user facility (https://www.arm.gov). The selected IOPs with relatively short duration (4–6 weeks) were part of four major atmospheric field campaigns, which examined the temporal and spatial variability of the aerosol microphysical and optical properties over climate-important continental, alpine, coastal, and marine locations (Figure 1 and Table 1), among other important topics. Similar suites of instruments, with state-of-the-art capabilities, sampled properties of the atmospheric aerosol, such as aerosol size distributions and total scattering coefficient (Table 1). In addition to the IOP-based aerosol properties, we include in our analysis related aerosol data with similar duration collected during summer and winter at the midcontinental ARM’s Southern Great Plains (SGP) site (Table 1). Detailed reviews of the integrated aerosol measurements and corresponding instruments are given in the corresponding papers (Table 1). Here, we use the data to show site-dependent contribution of supermicron particles into aerosol volume and total scattering coefficient and to illustrate the strong covariability of parameters representing the coarse aerosol volume and the wavelength and size dependence of total scattering coefficients over variety of locations and seasons.

Similar to Kassianov et al. (2012), we calculate CMF of aerosol volume as a ratio of the coarse-mode volume (PM_{10,v} – PM_{4.5,v}) over the total volume (PM_{10,v}), where PM_{4.5,v} is either PM_{1,v} or PM_{2.5,v} (aerodynamic particle diameter below 2.5 \ \mu m). We define the corresponding fractions as the CMF_PM_{10,1} (aerodynamic particle diameter range 1–10 \ \mu m) and CMF_PM_{10,2.5} (aerodynamic particle diameter range 2.5–10 \ \mu m), respectively. These two fractions illustrate relative contributions of supermicron particles with different size ranges to the total volume. The selection of the upper size cutoff (10 \ \mu m) for the aerosol volume is made to match the upper size cutoff (10 \ \mu m) for the aerosol total scattering coefficient. Also, we calculate the corresponding CMFs of the aerosol total scattering coefficient at three wavelengths (0.45, 0.55, and 0.7 \ \mu m) and denote them as CMF_{SCA_{0.45}}, CMF_{SCA_{0.55}}, and CMF_{SCA_{0.7}}, respectively. Finally, we assess the contributions of coarse particles to aerosol volume and total scattering following a probabilistic description (Kassianov et al., 2012). Our description involves cumulative percentiles, which describe the percentage of time (or frequency) when the CMF of a given aerosol property exceeds a specified threshold (or magnitude).

Both CMF_PM_{10,1} and CMF_PM_{10,2.5} depend strongly on the location and season (Figure 1). To illustrate, we consider two extreme examples with the smallest and largest values of the CMF_PM_{10,1}. Its smallest
values take place during summer at the continental location (the SGP site): The \( \text{CMF}_\text{PM10-1} \) exceeds about 15% for 50% of time (Figure 1a). In contrast, its largest values occur during winter at the marine location (the ENA site): The \( \text{CMF}_\text{PM10-1} \) exceeds 87% for 50% of time (Figure 1b). In other words, the coarse-mode contribution to the aerosol volume has strong spatial and seasonal changes, and its largest value (87%) is about 6 times larger than the corresponding smallest (15%) value at the selected frequency threshold (50% of time). Relative to the \( \text{CMF}_\text{PM10-1} \), the \( \text{PM\_ratio} \) exhibits even larger changes (more than 1 order of magnitude) with season and location (section 4). For this reason, we use the \( \text{PM\_ratio} \) for the \( \text{PM}_{10,v} \) prediction (section 4).

In contrast to the \( \text{CMF}_\text{PM10-1} \) and \( \text{CMF}_\text{PM10-2.5} \), the coarse-mode fractions of the total scattering coefficient show weaker spatial and seasonal changes. For example, their largest values occur during winter at the marine location (the ENA site): The \( \text{CMF}_\text{SCA0.45} \) and \( \text{CMF}_\text{SCA0.7} \) exceed 67% and 75% for 50% of time, respectively (Figure 1b). These largest values (67% and 75%) are about 4 and 2 times higher than the corresponding values (18% and 31%; Figure 1a) observed during summer at the typical continental location (the SGP site). Since the spatial and seasonal changes of the \( \text{CMF}_\text{SCA0.45} \) (67% vs. 18%) in comparison with those of the \( \text{CMF}_\text{SCA0.7} \) (75% vs. 31%) are stronger, we use the \( \text{CMF}_\text{SCA0.45} \) in our consequent analysis.

### 4. Results

Here we demonstrate that the joint application of two parameters (\( \text{CMF}_\text{SCA0.45} \) and \( \text{SCA\_ratio} \)) allows reasonable prediction of the \( \text{PM}_{10,v} \) values. A scatterplot, which pairs up values of the \( \text{PM\_ratio} \) and \( \text{SCA\_ratio} \) (Figure 2a), illustrates three important points. First, the upper limit of the \( \text{PM\_ratio} \) (~40) exceeds that of the \( \text{SCA\_ratio} \) (~3.8) by about an order of magnitude. Second, the \( \text{PM\_ratio} \) is small to moderate (up to 10) for all selected locations, except the marine location (the ENA site) where the \( \text{PM\_ratio} \) has the largest values (up to 40). Third, the complex relationship between the \( \text{PM\_ratio} \) and \( \text{SCA\_ratio} \) has a “hockey stick” shape (Figure 2a), which is mainly attributed to different types of aerosol. Note that different types of aerosol are also responsible for an intricate association between fine-mode fraction of aerosol optical depth and its Ångström exponent (0.44–0.87 μm) (e.g., Eck et al., 2010). Thus, it is unlikely that the complex relationship between the \( \text{PM\_ratio} \) and \( \text{SCA\_ratio} \) can be approximated by a simple function. Note that the “hockey stick” shape represents all possible cases with small, moderate, and large values of the \( \text{CMF}_\text{SCA0.45} \). The
Table 1
Aerosol Properties Measured for Five Locations and Two Seasons (at the TCAP, SGP, and ENA Sites Only), Which Are Relevant to This Study

| Location                          | CARES<sup>a</sup> | STORMVEX<sup>b</sup> | TCAP<sup>c</sup> | SGP<sup>d</sup> | ACE-ENA<sup>e</sup> |
|-----------------------------------|-------------------|-----------------------|------------------|----------------|---------------------|
| Location                          | Sacramento, CA (about 90 miles east of the Pacific Ocean) | Storm Peak, CO (high elevation, mountain top) | Cape Cod, MA | Lamont, OK | Graciosa Island (Azores, Portugal) Eastern North Atlantic |
| Locations info (aerosol type)     | T<sub>0</sub> (urban), T<sub>1</sub> (continental) | Storm Peak Laboratory (SPL) (alpine) | Near the coast of North America (coastal) | Cattle pasture and wheat fields (continental) | Azores island chain (maritime) |
| Periods                           | June 2010         | March 2011            | July–August 2012 | January 2013 | June–July 2017 |
| Sites info (aerosol type)         | T<sub>0</sub> (urban), T<sub>1</sub> (continental) | Storm Peak Laboratory (SPL) (alpine) | Near the coast of North America (coastal) | Cattle pasture and wheat fields (continental) | Azores island chain (maritime) |
| Aerosol size distribution (cm<sup>-3</sup>) | SMPS<sup>f</sup>, APS<sup>g</sup> | SMPS<sup>f</sup>, APS<sup>g</sup> | SMPS<sup>f</sup>, APS<sup>g</sup> | SMPS<sup>f</sup>, APS<sup>g</sup> | UHSAS<sup>h</sup>, APS<sup>g</sup> |
| Aerosol total scattering coefficient (Mm<sup>-1</sup>) | Nephelometer<sup>i</sup> | Nephelometer<sup>i</sup> | Nephelometer<sup>i</sup> | Nephelometer<sup>i</sup> | Nephelometer<sup>i</sup> |

Note: General information regarding the selected sites (second, third, and fourth rows) and measurements of aerosol properties (fifth and sixth rows) is included. Important details and the corresponding references are provided in the footnotes. Figure 1 contains maps with schematic locations of the selected sites.

<sup>a</sup>The Carbonaceous Aerosols and Radiative Effects Study (CARES) (Kassianov et al., 2012; Setyan et al., 2014; Zaveri et al., 2012).  
<sup>b</sup>The Cloud Property Validation Experiment (STORMVEX) (Marchand et al., 2013); the major dust episode (Kassianov et al., 2017).  
<sup>c</sup>The Two-Column Aerosol Project (TCAP) (Berg et al., 2016).  
<sup>d</sup>The Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) Central Facility (McComiskey & Ferrare, 2016).  
<sup>e</sup>The Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) (Wu et al., 2018).  
<sup>f</sup>Scanning Mobility Particle Sizer (SMPS): the mobility diameter range 0.012–0.737 and 0.008–0.858 μm at the T<sub>0</sub> and T<sub>1</sub> sites, respectively. The SMPS-measured aerosol mobility size distributions are converted into their aerodynamic size counterparts at the STORMVEX, TCAP, and SGP sites using an assumed density (1.8 g/cm<sup>3</sup>) similar to Kassianov et al. (2012). The density (1.4 g/cm<sup>3</sup>) estimated by Setyan et al. (2012) is used for the conversion of the SMPS-measured size distributions collected at the CARES T<sub>0</sub> and T<sub>1</sub> sites. Note that the SMPS size spectra are not available at the TCAP site during winter (Figure 1b).  
<sup>g</sup>Aerodynamic Particle Sizer (APS): aerodynamic diameter range 0.5–20.0 μm. Figure S1 shows an example of the APS aerodynamic size distribution.  
<sup>h</sup>Ultra-high-sensitivity aerosol spectrometer (UHSAS): optical size range 0.06–1 μm. The UHSAS-measured aerosol optical size distributions are converted into their aerodynamic size counterparts at the ENA site using the complex RI and density estimated from the complementary chemical composition measurements. Note that information on sea salt and dust is not provided by these measurements. Figure S1 shows an example of the UHSAS-based aerodynamic size distribution.  
<sup>i</sup>TSI integrating nephelometer (TSI Inc., Model 3563): total scattering coefficient at three wavelengths (0.45, 0.55, and 0.7 μm).
segregation of these cases by site- and season-dependent CMF_SCA$_{0.45}$ gives an opportunity to approximate this complex relationship by a group of linear regressions for a given site or season of interest (Figure 2b).

However, it is unclear whether such simple approximation can be used to predict the PM$_{10,v}$ reasonably well from the measurement-based variables (CMF_SCA$_{0.45}$ and SCA$_{ratio}$) using the introduced approach (section 2). To illustrate a benefit of this approximation in terms of the PM$_{10,v}$ prediction, we consider two scenarios. The first scenario is “site-specific,” which characterizes the situation when the site- and season-dependent data are available. Here, the existing seven data sets with the required size spectra and light scattering data (section 3), describing the site- and season-dependent changes of the aerosol properties, are used to generate seven groups of linear regressions. We use these seven groups and the introduced approach (section 2) to predict the PM$_{10,v}$ for the corresponding locations (Figure 3, left column). The second scenario is “generic,” which characterizes situation when the site- and season-dependent data are not available. To demonstrate such situation, we combine the existing seven data sets into a single data set. Then, we generate single group of linear regressions from the combined data set assuming that the site and season-dependent data are not available. We use this single group and the introduced approach (section 2) to predict the PM$_{10,v}$ at all the sites and seasons considered here (Figure 3, right column).

We begin with considering the “site-specific” scenario (Figure 3, left column). The time series of the measured PM$_{10,v}$ exhibit strong temporal and spatial changes. They are more pronounced during summer for the coastal location (Figure 3c; the TCAP site), which defines a crossroad of distinct air-mass flow patterns (Berg et al., 2016). The opposite is true for the typical continental location during summer (Figure 3e; the SGP site) where the temporal variability of the measured PM$_{10,v}$ is fairly small. In general, there are very similar patterns of the predicted and measured PM$_{10,v}$ for the selected sites and seasons. As a result, statistics describing the predicted and measured PM$_{10,v}$ are comparable and the corresponding values of the correlation coefficients are quite high (about 0.66–0.97) (Table 2). However, there are a few exceptions, which define occasional substantial differences between the predicted and measured PM$_{10,v}$ especially at the urban (Figure 3a; the CARES T0 site) and marine (Figure 3f; the ENA site) locations. Likely, these exceptions represent events with the substantial contributions of refractory species, such as black carbon, sea salt, and dust, to the measurement-based PM$_{10,v}$ (supporting information Figure S2). The current version of our approach does not take into account these contributions due to lack of information related to the refractory species. It should be emphasized that a simple screening of these events (when information about chemical composition is available) can improve substantially agreement between the predicted and measured PM$_{10,v}$. For example, the correlation coefficient increases from 0.66 (all data) to 0.83 (screened data) at the ENA site (Figure S2). The “site-specific” scenario with the existing site- and season-dependent data (Table 1) is presumed to be applicable to locations and seasons where these data likely define a dominant type of local
aerosol. For example, marine aerosol is mostly observed at the remote ENA site, which is located well away from the major continental sources (Zheng et al., 2018), and thus, the “site-specific” scenario could be relevant for the multiyear aerosol data collected at the ENA site. Additional integrated measurements of the required aerosol properties are needed to better characterize the spatial and temporal changes of the atmospheric aerosol and thus to improve the \( PM_{10,v} \) predictions for locations and seasons where such measurements are not available.

We continue with considering the “generic” scenario (Figure 3, right column). Mainly, there are coherent patterns of the predicted and measured \( PM_{10,v} \) values for the selected sites and seasons. The “generic” patterns have several noticeable differences in comparison with those obtained for the “site-specific” scenario. These differences include rougher and smoother variations in the predicted \( PM_{10,v} \) for the urban (Figure 3g; the CARES T0 site) and marine (Figure 3l; the ENA site) location, respectively. Thus, the corresponding values of root-mean-square error (RMSE) are larger (the CARES T0 site) and smaller (the ENA site), while

**Table 2**

| Site       | OBS       | “Site-specific” | “Generic”  |
|------------|-----------|-----------------|------------|
|            | \( PM_{1,v} \) | \( PM_{10,v} \) | \( r \) | RMSE         | \( PM_{10,v} \) | \( r \) | RMSE         |
| CARES T0   | 4.18      | 14.65           | 17.21      | 0.87 | 6.74         | 24.99 | 0.83 | 22.09       |
| CARES T1   | 2.78      | 10.56           | 11.13      | 0.87 | 3.61         | 10.15 | 0.84 | 5.03        |
| SPL        | 1.19      | 3.03            | 3.40       | 0.96 | 1.17         | 4.33  | 0.90 | 1.97        |
| TCAP       | 8.92      | 21.23           | 23.02      | 0.80 | 8.14         | 16.72 | 0.79 | 9.35        |
| SGP winter | 3.06      | 4.59            | 5.26       | 0.93 | 1.35         | 6.08  | 0.89 | 2.49        |
| SGP summer | 4.76      | 5.80            | 5.84       | 0.97 | 0.52         | 10.22 | 0.93 | 4.92        |
| ENA        | 0.90      | 8.54            | 11.10      | 0.66 | 7.43         | 9.41  | 0.65 | 6.39        |

Note. The corresponding statistics describe relationship between the measured and predicted \( PM_{10,v} \) values for the selected sites. These statistics include the correlation coefficient (\( r \)) and root-mean-square error (RMSE). The mean values and RMSE have the same dimension (\( \mu m^3/cm^3 \)), while the correlation coefficient is dimensionless.
the corresponding values of correlation coefficient are almost scenario invariant (Table 2). The differences (Figure 3 and Table 2) suggest that the “generic” scenario with the existing aerosol data (Table 1) can offer overall information on the temporal changes of the predicted PM$_{10,v}$ for a given location and season, such as its general pattern and periods with large and small values, but the range of these changes can be underestimated or overestimated substantially especially at fine scales (e.g., 1-hr averages). Likely, the “generic” scenario with the existing aerosol data (Table 1) would be more beneficial for predicting the PM$_{10,v}$ changes at coarser temporal resolution (e.g., daily averages) rather than those at finer scales (e.g., 1-hr averages). Note that daily averages typically describe the temporal changes of pollutants (e.g., Vahlsing & Smith, 2012).

5. Conclusions

We introduce a new method for predicting total aerosol volume PM$_{10,v}$ (particulate matter with aerodynamic particle diameter below 10 μm). The PM$_{10,v}$ prediction involves both (1) fine aerosol volume PM$_1$, (particulate matter with aerodynamic particle diameter below 1 μm) calculated from measured size spectra of sub-micron particles and (2) measured wavelength and size dependence of aerosol total scattering coefficient. The wavelength dependence is described by a ratio of total scattering coefficients measured at two wavelengths (0.45 and 0.7 μm) and for 10-μm size cutoff (aerodynamic particle diameter). The size dependence is described by coarse-mode fraction of the aerosol total scattering coefficient calculated at single wavelength (0.45 μm) using two scattering coefficients measured with 1- and 10-μm size cutoffs (aerodynamic particle diameters).

To illustrate the performance of this method, we use aerosol size distributions and total scattering coefficients measured at five sites with continental, alpine, coastal, and marine environments. Distinct environmental conditions and aerosol types observed at these sites are responsible for strong temporal and spatial changes of the total aerosol volume. Despite of these strong changes, we demonstrate that the introduced method is able to predict PM$_{10,v}$ reasonably well. For example, the correlation coefficient between the measured and predicted PM$_{10,v}$ volumes is relatively high (0.65–0.97) and the RMSEs are typically smaller than the corresponding mean values of the measured PM$_{10,v}$. The ARM user facility (e.g., Mather & Voyles, 2013; Miller et al., 2016) and Global Atmosphere Watch (GAW) stations (e.g., Andrews et al., 2019) offer important measurements of the aerosol properties required for potential PM$_{10,v}$ prediction around the world. The introduced approach, if proven accurate, could be applied to the existing and future ARM and GAW data with worldwide coverage, and the expected PM$_{10,v}$ predictions would provide useful, but previously missed, pieces of the climate puzzle.

Data Availability Statement

Here is a list of websites with description of the selected field campaigns as well as the SGP site (Table 1) and the corresponding data archives: CARES (https://www.arm.gov/research/campaigns/aaf2009carbonaerosol); STORMVEX (https://www.arm.gov/research/campaigns/amf2010valclradar); TCAP (https://www.arm.gov/research/campaigns/amf2012tcap; https://www.arm.gov/research/campaigns/amf2012aps); SGP (https://www.arm.gov/capabilities/observatories/sgp; https://adc.arm.gov/discovery/); and ACE-ENA (https://www.arm.gov/research/campaigns/aaf2017ace-ena; https://www.arm.gov/research/campaigns/ena2017aceenawinterapasi).

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