

Sources and Properties of Atmospheric Aerosol in Texas: DISCOVER-
AQ Measurements and Validation

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1. Executive Summary

During this project (January-September, 2015) we have been working on the data collected by the Cloud and Aerosol Spectrometer with Polarization (CASPOL) during its first deployment in the field campaign— the National Aeronautics and Space Administration (NASA) Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) campaign. As an instrument that measures the particle scattering properties, CASPOL shows the potential to assess the quality of satellite retrievals of aerosol features, which have been widely used to study the sources and properties of atmospheric aerosols.

During the fall of 2013, the Houston area was the site of the 2nd field intensive DISCOVER-AQ campaign. During the field campaign, we operated CASPOL on top of the 70 m tall Moody Tower (MT) on the University of Houston campus, a central urban location and site of many complementary measurements of air pollutants and meteorological parameters. After a theoretical analysis, we found that the statistics of CASPOL backward polarized scattering measurements of individual particles can be made to assess the aerosol typing criteria of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), though the wavelengths of two instruments are not the same. In addition, the aerosol type assumption in the Moderate-resolution Imaging Spectroradiometer (MODIS) aerosol optical depth (AOD) retrievals can be assessed using CASPOL in combination with in situ sun photometer measurements in the same location. However, after checking the available CALIOP depolarization ratio profiles and MODIS AOD retrievals during the period when CASPOL data were of high quality, too few comparison cases could be found to make such an assessment of any significance. Thus, we focused our comparisons on the MODIS fine mode fraction (FMF) retrievals for which more comparison cases were found.

The high quality CASPOL measurements coincided with MODIS overpasses under cloud free conditions on 13 days. The forward scattering measurements from CASPOL were used to derive aerosol size distributions, which were then compared with the MODIS 3 km FMF retrievals. The sub-micrometer fraction (SMF) of aerosol particles sampled by CASPOL and Aqua MODIS FMF retrieval show close variations. However, the monthly mean MODIS FMF data from Terra differs from the MODIS on Aqua by a factor of ~3. Both MODIS FMF retrievals and CASPOL measurements show a significant

decrease of the proportion of coarse particles from noon to the early afternoon during the campaign. The decreased amount is dependent upon the particle size cutoff that discriminates fine and coarse particles. The comparison cases are not enough to make the results statistically significant, and hence long-term CASPOL field measurements are needed to assess the satellite aerosol-property retrievals in the future.

2. Introduction

Atmospheric aerosols are significant components in the Earth-atmosphere system that influence weather and climate through modifying the radiation budget and interacting with clouds (e.g., Rosenfeld et al., 2014; Tao et al., 2012). Moreover, parts of aerosol particles are harmful to human health. Exposure to high concentrations of aerosols with diameters less than or equal $2.5\ \mu\text{m}$ ($\text{PM}_{2.5}$) and $10\ \mu\text{m}$ (PM_{10}) can cause a higher risk of respiratory and cardiovascular diseases (Dominici et al., 2006) as well as low birth weight and preterm birth (Sapkota et al., 2012). Hence, global observations of aerosols are needed for studying aerosol effects on climate and environment.

Satellite remote sensing provides the global distribution of aerosols and their properties, which have been widely used in aerosol studies. A number of space-borne sensors—such as the Advanced Very High Resolution Radiometer (AVHRR) (Ignatov et al., 2004), the Total Ozone Mapping Spectroradiometer (TOMS) (Torres et al., 1998), the MODerate Resolution Imaging Spectroradiometer (MODIS) (King et al., 1992), the Advanced Along-Track Scanning Radiometer (AATSR) (Sayer, 2008), the Multiangle Imaging Spectroradiometer (MISR) (Martonchik and Diner, 1992), and the Polarization and Directionality of the Earth's Reflectances (POLDER) (Herman et al., 2005; Tanré et al., 2011)—have been designed to retrieve aerosol properties in the past decades. Based on the observations of these sensors, aerosol features can be retrieved through specific algorithms in which certain assumptions have to be made. The retrieved features include aerosol optical depth (AOD or τ) and aerosol size parameters, such as Ångström exponent and fine mode fraction (FMF).

The Cloud and Aerosol Spectrometer with Polarization (CASPOL) is a new instrument that has the potential to be used to evaluate satellite aerosol retrievals. The CASPOL measures the forward (4° to 12°) and backward (168° to 176°) scatter radiation for individual particles at 680 nm. The parallel and perpendicularly polarized components of the backward scatter radiation are measured separately, and hence the backward depolarization ratio can be derived. The diameters of the particles that can be sampled by CASPOL range from $0.6\ \mu\text{m}$ to $30\ \mu\text{m}$. The particle-by-particle backscatter features of different particle groups have been studied using CASPOL. Glen and Brooks (2013) successfully categorized thirteen dust events into three groups based on their plots of depolarization ratio vs. total back scatter intensity. Ice crystals and dust particles may also be differentiated from CASPOL measurements, as suggested by the laboratory experiment for single dust and ice particles (Glen and Brooks, 2014). In addition, aerosol size distributions can be inferred from the forward scattering measurements, and hence it provides a chance to assess the qualities of satellite aerosol size retrievals, such as MODIS FMF retrievals.

The MODIS instrument is a radiometer that measures reflected solar and emitted terrestrial radiation in 36 bands between 0.4 and $14.4\ \mu\text{m}$. Because MODIS aerosol retrievals are based on reflected solar radiation, they are only made during the daytime.

MODIS retrieves aerosol properties by comparing measured reflectances to modeled reflectances from a wide range of aerosol simulations (Levy et al., 2007). The modeled reflectances depend on a number of assumptions about aerosol composition, size distribution, and absorption and scattering properties, as well as environmental conditions including surface properties, atmospheric profile, and viewing geometry. MODIS aerosol retrievals have been extensively used in different research areas, including environment and health (e.g., van Donkelaar et al., 2010), and weather and climate (e.g., Niu and Li, 2012).

MODIS aerosol retrievals of aerosol optical depth and fine mode fraction have been used to estimate aerosol number concentrations and anthropogenic aerosols. Anthropogenic aerosols are mostly dominated by fine particles whereas natural aerosols are dominated by coarse particles (e.g., Deuzé et al., 2001; Kaufman et al., 2001; Tanré et al., 2001). Thus, FMF retrievals have been widely used to characterize the anthropogenic component (e.g., Bellouin et al., 2005; Christopher et al., 2006; Kaufman et al., 2005; Ramachandran, 2007; Yu et al., 2009).

Uncertainties of MODIS aerosol property retrievals stem from the applicability of those retrieval assumptions in different environments, including assumed spectral dependence of surface reflectivity and aerosol models. The previous quality assessments of MODIS optical depth and fine mode fraction retrievals have been made through comparisons with ground-based or airborne sun photometer retrievals (e.g., Anderson et al., 2005a; Chu et al., 2005; Kleidman et al., 2005; Remer et al., 2005). Because both the MODIS and sun photometer make retrievals of total column features of aerosols, they are directly comparable. The results show a poor correlation between MODIS and sun-photometer FMF retrievals over land. Over oceans, MODIS appears to overestimate low values of FMF and underestimate high values of FMF. Based on these comparison studies, Levy et al. (2010) concluded that the MODIS Collection 5 aerosol size retrievals show noteworthy uncertainties and hence have little physical validity in a quantitative sense. They also recommended that FMF be kept for the purpose of examining the algorithm performance only.

One of the new products of the MODIS Collection 6 is the new aerosol retrieval whose spatial resolution of aerosol retrievals increases from the previous 10 km to the current 3 km (R C Levy et al., 2013). MODIS onboard Terra and Aqua satellites have been collecting data for more than ten years. However, to the best of our knowledge, no study has compared MODIS FMF retrievals with aerosol size distributions measured in the field. In our study, we compare the MODIS FMF retrievals with CASPOL aerosol size distribution measurements in the Houston urban area during the 2nd intensive Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) campaign in 2013.

3. Data and methodology

The Cloud and Aerosol Spectrometer with Polarization (CASPOL) is a new optical particle counter developed by the Droplet Measurement Technology, Inc. (DMT) and calibrated in the laboratory (Glen and Brooks, 2013; 2014). This instrument measures the particle-by-particle optical properties of aerosols. Glen and Brooks (2013) showed a schematic of CASPOL. Two detectors in the backwards direction used to measure the total and perpendicularly polarized backscatter radiation; but now they measure the parallel and perpendicularly polarized backscatter radiation. The forward scatter

intensity, measured by the detector in the forward direction, is used to derive the size distribution of particles within 28 bins from 0.68 μm to 50 μm in diameter based on the Mie scattering theory. The size range covers the cut-off point of about 1 μm that is often adopted to discriminate fine and coarse aerosol particles (Anderson et al., 2005b). Thus, the ratio of the concentration of particles smaller than the cut-off to the total concentration can characterize the proportion of fine particles. For the purposes of this study, we define this as the CASPOL particle submicrometer fraction (SMF), following the similar naming by Anderson et al. (2005a). The CASPOL bin boundaries closest to 1 μm are 0.96 μm and 1.03 μm , and we therefore take the two values as possible cut-offs to calculate SMFs.

The 2nd intensive DISCOVER-AQ campaign took place over the Houston from August 15th to October 2nd in 2013. One of the objectives of the campaign was improved interpretation of satellite observations with regard to air pollutants. During the field campaign, CASPOL was put on the Moody Tower (29.7176° N, 95.3414° W) for measurements from 15 August to 2 October in 2013, providing a chance to compare the CASPOL in-situ aerosol measurements with satellite aerosol retrievals. The Moody tower has been the location of a number of previous field campaigns (e.g., Brooks et al., 2010; Lefer et al., 2010; Rappenglück et al., 2010; Wong et al., 2011). The height of the Moody Tower is about 70 meters. It is low enough so that the aerosols being sampled are representative of the aerosols at the surface, but tall enough so that any intermittent point sources will not interfere with the measurements.

Fig. 1 shows a schematic of the experimental setup for CASPOL measurements during the campaign. The CASPOL inlet was specially designed to rotate freely in order to keep pointing to the upstream wind. The inflow air was first run through a heated stainless-steel pipe (1.5 m in length) wrapped with a non-conductive tubing, so the relative humidity was kept constant to avoid condensation (Quinn et al., 1998). The sample flow beyond the heated pipe was then split into CASPOL at a rate of 1.2 L min⁻¹ and a dump line at a rate of 10 L min⁻¹, controlled by a pair of mass flow controllers and pumps. Behind the CASPOL were two groups of thermocouples (TC1 and TC2) and relative humidity meters (RH1 and RH2) with a high-efficiency particulate arresting (HEPA) filter in between. At least twice a week, the inlet line was changed, and the tubing was changed or dried. Data collected during rainy periods was removed.

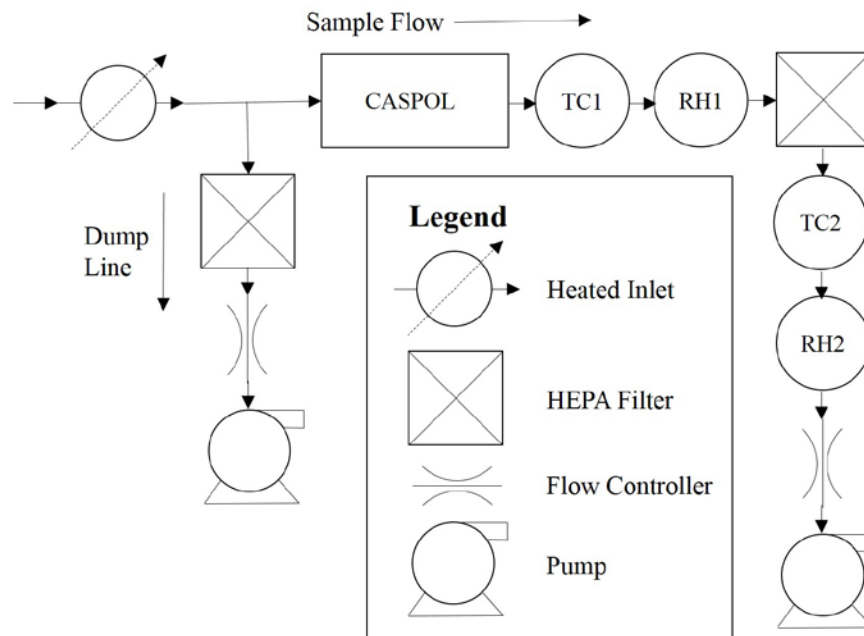


Figure 1. Experimental setup for CASPOL measurements.

DISCOVER-AQ was the first field campaign in which CASPOL was deployed. Following a period of successful data collection, the CASPOL experienced a serious drop in observed concentration on 18 September, 2013. A plausible cause was an (unspecified) instrument sampling in the CASPOL inlet line at that time. Hence, we also removed the data later than 17 September. Recent laboratory calibration indicates that the CASPOL is once again working well and accurately counting and sizing aerosols. Hence, we tentatively conclude that the problem experienced in the field was related to a blockage in the CASPOL inlet tubing, rather than an issue in the instrument itself. As a consequence, only the CASPOL measurements during 5-17 September were kept to ensure the validity of the data.

Based on observations of bimodal aerosol size distributions, the MODIS aerosol algorithm over dark continental surfaces is modeled as a combination of fine-dominated and coarse-dominated aerosol models. Each of the aerosol models consists of two modes—accumulated and coarse modes, and the lognormal size distribution is assumed for each mode. The lognormal size distribution is assumed by MoDIS's Algorithm Theoretical Basis Document (ATBD). MODIS aerosol algorithm. While the MODIS team does not provide the specific reason for not choosing other size distributions in the ATBD, a lognormal distribution is a very common assumption in aerosol studies based on the fact that measured size distributions are commonly lognormal in shape. During DISCOVER-AQ, the coarse mode is always assumed to be nonspherical dust. The modeled fine mode includes three spherical aerosol models whose absorbance levels are different, representing weakly, moderately, and strongly absorptive aerosols. The selection of one of the three types is a function of geography and season. This project took place over the Houston area in September 2013. Based on this location and season, the weakly absorptive aerosols were selected as the fine-dominated model in the MODIS algorithm. Table 1 documents the size parameters of the weakly absorptive fine model and the coarse model in the MODIS aerosol algorithm, where r_v , σ , and V_0 are median radius, standard deviation, and volume concentration, respectively.

Table 1. Size parameters of the weakly absorptive fine model and the coarse model in the MODIS aerosol algorithm (Levy et al., 2009).

	Fine model (weakly absorptive)		Coarse model	
	Accumulated	Coarse	Accumulated	Coarse
Median radius, r_v (μm)	$0.0434\tau+0.1604$	$0.1411\tau+3.3252$	$0.1416 \tau^{-0.0519}$	2.2
Standard deviation, σ (μm)	$0.1529\tau+0.3642$	$0.1638\tau+0.7595$	$0.7561\tau^{0.148}$	$0.554 \tau^{-0.0519}$
Volume concentration (V_0)	$0.1718 \tau^{0.8213}$	$0.0934 \tau^{0.6394}$	$0.0871 \tau^{1.026}$	$0.6786 \tau^{1.0569}$

Fig. 2 shows the volume size distributions of the fine (weakly absorptive) and coarse models in the conditions of optical depth, τ , = 0.2, 0.8, 1.4, and 2.0, where R and V are radius and volume of particles, respectively. The vertical dashed black line corresponds to $R = 0.5 \mu\text{m}$. The peaks of the coarse model are at $R = 2.2 \mu\text{m}$. The peaks of the fine model vary between $R = 0.1 \mu\text{m}$ and $R = 0.5 \mu\text{m}$. It appears that about $0.5 \mu\text{m}$ in radius (i.e., $1 \mu\text{m}$ in diameter) is also a division between the assumed fine and coarse particles in the MODIS aerosol algorithm, in agreement with the cut-off point from observations. Next we compare MODIS FMF retrievals with a particle submicron fraction calculated from the CASPOL measurements during the campaign.

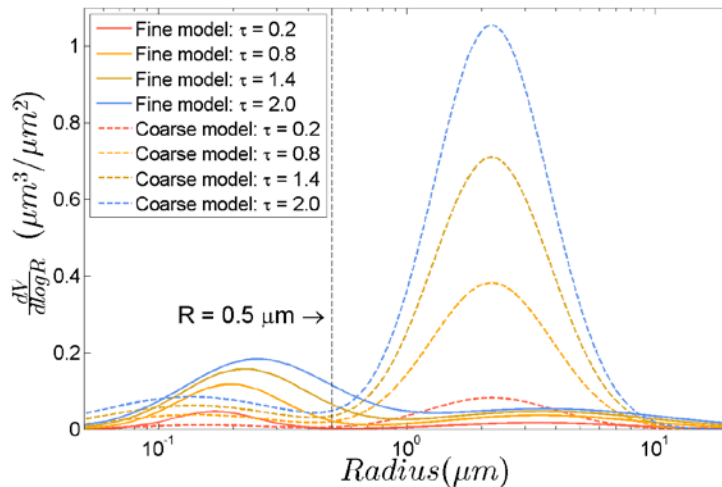


Figure 2. The dynamic volume size distributions of the fine (weakly absorptive) and coarse (dust) models in the conditions of different AODs in the MODIS aerosol algorithm.

4. Audits of data quality

The MODIS fine mode fraction, FMF, is retrieved in a diagnostic way and can take only 11 discrete values. In the retrieval algorithm, 13 values of FMF (from -0.1 to 1.1 with an increment of 0.1) are tried by turns, and then the one kept at last minimizes the difference in the apparent reflectance at the $0.66 \mu\text{m}$ band between the look up table and the MODIS. The two non-physical values of FMF (-0.1 and 1.1) are tried for bearing

possibly inappropriate assumptions in the algorithm to some extent. MODIS aerosol retrievals are assigned a quality assurance confidence (QAC) flag, the value of which ranges from 0 to 3. Larger the value is, better the quality is. The QAC value is determined by the behavior of the algorithm, such as the number of dark pixels (Levy et al., 2009). The quality assurance on FMF retrievals is stricter than those of other aerosol-property retrievals. The values of FMF over land are reported only when retrieved AOD > 0.2 and QAC \geq 1. Hence, MODIS FMFs are of high quality as diagnostic retrievals.

The CASPOL was operated one and a half months during the field campaign, but only 13 day (5-17 September) high quality data were kept. Before this period, the condensation building up in the lines leading to the CASPOL was not correctly set up. Right after this period, an unusual drop in the CASPOL observed particle concentration was present. The high quality was ensured by filtering the CASPOL data that are in good agreement with trends in the other particle concentration measurements (Rebecca Sheesley, personal communication) collected at the Moody Tower. Hence, the quality of the CASPOL measurements used in the study is good.

5. Results

Terra and Aqua, two polar-orbiting satellites, each carry a MODIS instrument. In the daytime, Terra passes the Houston area around noon and Aqua passes the same area in the early afternoon (near 2 pm locally). Fig. 3 shows the monthly mean distributions of FMF retrievals from Terra and Aqua over southeast Texas in September 2013. In each panel, the red circle is centered at the Moody Tower with a radius of 50 km. We averaged the available FMF retrievals within the circle for the comparison with the CASPOL particle submicron fraction. Overall, the monthly mean Aqua FMF is 3 times higher than Terra.

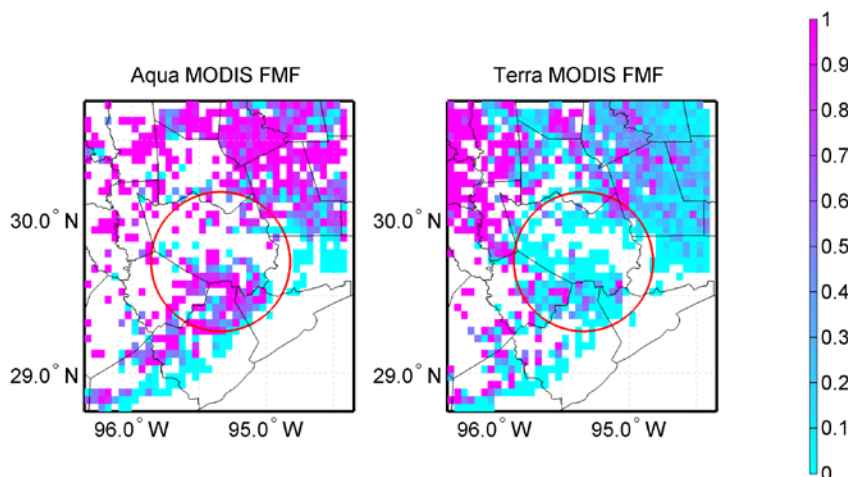


Figure 3. The monthly mean Aqua (left panel) and Terra (right panel) MODIS FMF distributions in September 2013 over an area from 28.75° N to 30.75° N and from 96.35°

W to 94.35° W. The red circle in each panel is centered at the Moody Tower with a radius of 50 km.

Because the CASPOL measures aerosol particles at the surface, but MODIS aerosol retrievals are column-averaged, it was not certain that the results would be comparable. One question is whether column based measurements can provide information about linear-surface air quality conditions, and if so, with what level of certainty. In a previous study, based on the aircraft measurements in Beijing, China, 102 out of 129 (79.1%) profiles show vertically homogenous shapes of number size distributions, and 27 out of 129 (20.9%) profiles show increased coarse aerosols above the planetary boundary layer, which is presumably due to long-distance transportation of dust aerosols (Liu et al., 2009). The major dust source nearest to Houston is the western Texas, a part of the Chihuahuan Desert. In general, the lower troposphere prevailing wind changes from southwesterly in April to southeasterly in September, which is a result of the geostrophic balance between the North Atlantic subtropical high (NASH) and the broad continental low over the Rockies (Cook et al., 2008). As a consequence, the Houston area is generally unlikely to be downwind of the major dust source in September. Therefore, the vertical distributions of SMF were more likely to be homogenous during the DISCOVER-AQ campaign, though variations were possible. One limitation is that any vertical layers of transported aerosol arising from biomass burning north of Houston would not be detected by the CASPOL.

Based on position in the center of Houston, air masses over the Moody Tower are positioned such that it is possible to be influenced by one of four major aerosol sources: 1) the Ship Channel source, which is a heavily industrialized area on the east side of Houston; 2) an Urban source, which consists of the densely populated, urban center of Houston; 3) a marine source, which consists of transported aerosols from the Gulf of Mexico and potentially further (Goudie and Middleton, 2001), and 4) the Semi-Urban/Rural source, which consists of transported aerosols from the west and passes over the less densely populated zones of the greater Houston area. Conveniently, these sources come from four different wind directions relative to the Moody Tower. Time periods of when these sources potentially occurred were determined using the NOAA, Atmospheric Resources Laboratories Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1997, 1998; Draxler et al., 1999) to create five day back trajectories with one hour intervals using Global Data Assimilation (GDAS) model data with 0.5 degree resolution. Ten cases were found in the data when HYSPLIT back trajectories were consistent, indicating the wind direction was from one of the four sources. These cases rang from six to thirty hours in length. The Ship Channel case was sampled when the HYSPLIT showed the wind was from 45° to 135° , the Ocean case from 135° - 225° , the Semi-Urban/Rural case from 225° - 315° , and the Urban case from 315° to 45° (Fig. 4). In total, five Ship Channel cases, three Urban cases, and two Ocean cases were identified for further analysis of the scattering properties. During the CASPOL sampling period, no Semi-Urban/Rural cases were identified during the time period of the campaign.

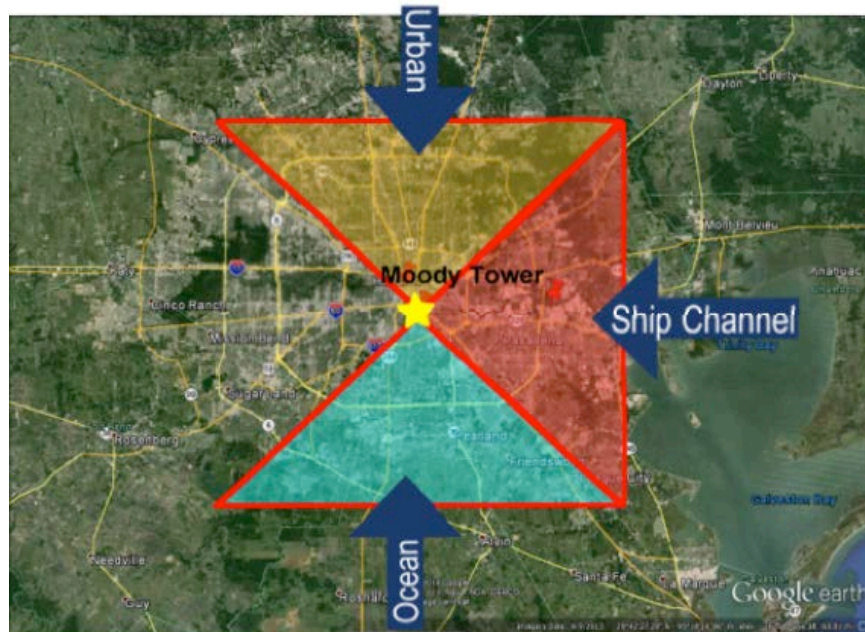


Figure 4. A map of the Houston Metropolitan area showing the three sources, Ocean, Ship Channel, and Urban, and the direction they come from (map courtesy of Google earth). We note that a fourth type of air mass, Semi-Urban/Rural source, is possible any time flow is from the west, but the westerly flow never occurred during this sampling campaign.

During the sampling period, 5-17 September, 2013, we found 7 Aqua and 6 Terra MODIS FMF retrieval cases. For each case, we counted the particles sampled by CASPOL as to their sizes 30 minutes before and after the satellite overpassing time. Fig. 5 shows normalized aerosol number size distributions measured by CASPOL for all 13 cases. The vertical dashed black line marks $R = 0.5 \mu\text{m}$. As shown in Fig. 5, the normalized concentrations of coarse particles for the Terra cases are higher than those for the Aqua cases. This diurnal variation was also found in previous studies. Previous field measurements of aerosol size distributions at the same place from 30 April to 7 May 2009 showed the same phenomenon—a decrease of the average aerosol diameter from noon to the early afternoon (Levy et al., 2013). Moreover, an analogous variation was found in the urban atmosphere of Beijing in China as shown by an observational study (Shi et al., 2007). They attributed the afternoon dispersion of accumulation mode particles to the enhanced vertical mixing and wind speed. In addition, this CASPOL observed diurnal feature is consistent with that observed by the Terra and Aqua MODIS as shown in Fig. 5, however, the MODIS observed diurnal feature may result from either instrumental issues or uncertainties in the FMF retrievals.

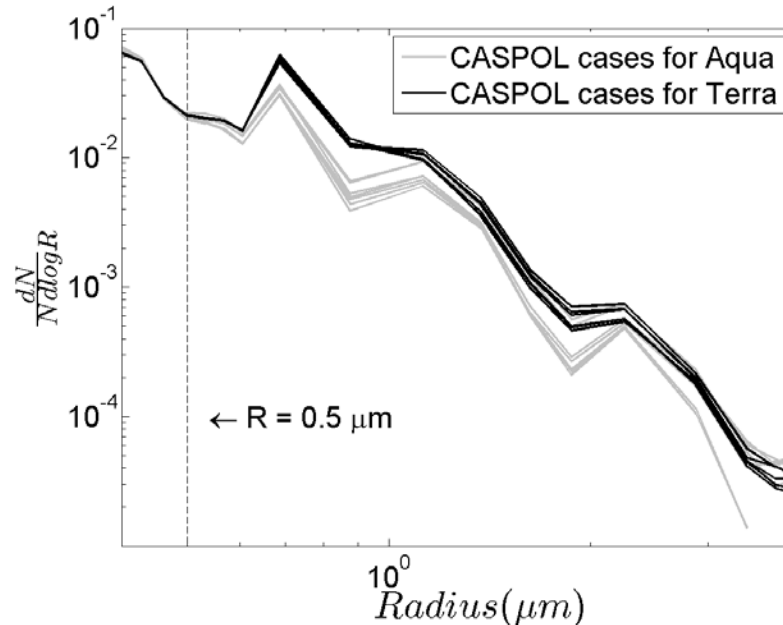


Figure 5. The normalized number size distributions from CASPOL measurements for the 7 Aqua and 6 Terra comparison cases.

The CASPOL data includes 2 bins with cut-offs very close to the 1 micrometer cut-off employed in MODIS analysis. We chose the cut-offs of 0.96 and 1.03 μm in diameter to calculate the CASPOL SMFs from the number size distribution for each case. Fig. 6 shows the scatterplots of CASPOL SMF vs. MODIS FMF for the two cut-offs. It appears that CASPOL SMF measurements and Aqua MODIS FMF retrievals are correlated. They show a correlation coefficient (ρ) of 0.652 for the 0.96 μm cut-off and 0.632 for the 1.03 μm cutoff. However, Terra MODIS FMF retrievals are not consistent with CASPOL. Previous studies have shown that the transition of electronics of the Terra MODIS might degrade the quality of its FMF retrievals (Chu et al., 2005; Remer et al., 2005). Overall, these results suggest that Terra MODIS FMF retrievals are erroneous. Determining the cause of the problems with the Terra retrievals is beyond the scope of this study. The averaged SMF of Terra cases are lower than that of Aqua cases. They are 0.747 and 0.805 for the cut-off of 0.96 μm ; and they are 0.790 and 0.846 for the cut-off of 1.03 μm . Both differences pass the unpaired one-tail t -tests at a significance level of 0.01, suggesting a significant decrease in the proportion of the coarse particle concentration from noon to the early afternoon.

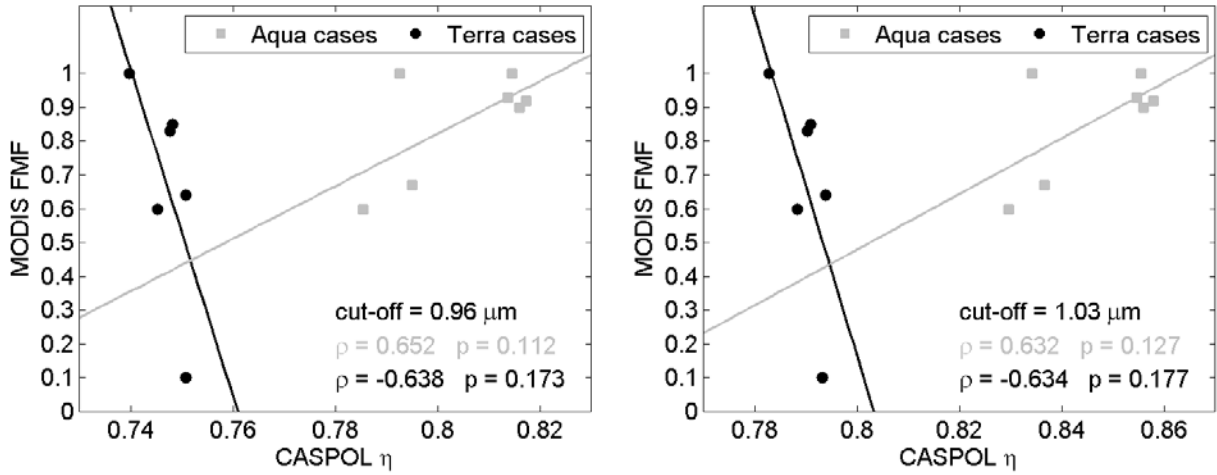


Figure 6. Scatterplots of MODIS fine mode fraction vs. FMF CASPOL submicron fraction. SMF. The lines are the linear regressions. ρ and p are the correlation coefficient and p-value, respectively. SMF is calculated using a cut-off of $0.96 \mu\text{m}$ on the left panel, and it is calculated using a cut-off of $1.03 \mu\text{m}$ on the right panel.

The value of SMF is dependent on the choice of the cut-off point that separates the fine and coarse particles. Fig. 7. shows the sensitivity of SMF to small deviations of the cut-off around $1 \mu\text{m}$ in diameter. As the cut-off increases from $0.75 \mu\text{m}$ to $1.25 \mu\text{m}$, SMF increases from around 0.4 to above 0.9 for the Aqua cases and from just below 0.4 to just below 0.9 for the Terra cases. The shapes of the curves are generally consistent for the Aqua and Terra cases, respectively, as shown in the left and right panels in Fig. 7. The coherent variations of SMF as such suggest that a small difference in the chosen cut-off does not lead to inconsistent characterizations of the fine-particle fraction. The values of SMFs defined by different cut-offs and associated MODIS FMF retrievals for all the comparison cases are documented in Table 2.

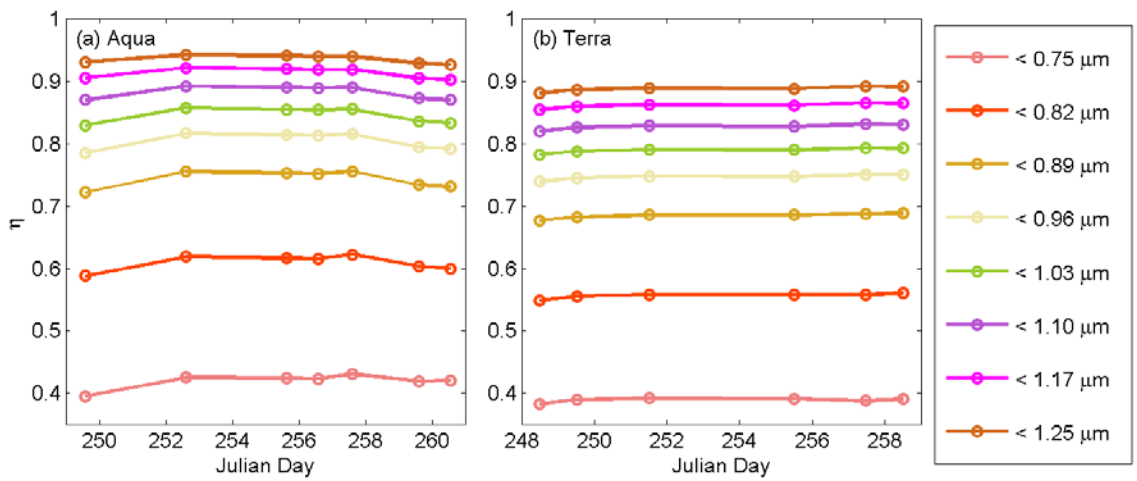


Figure 7. Time series of CASPOL SMF measurements defined by different cut-off points. The left panel shows the Aqua cases, and the right panel shows the Terra cases.

Table 2. MODIS FMF retrievals and CASPOL SMF measurements for all comparison cases.

Day	Time (CDT)	Sensor	FMF	CASPOL Submicron Fraction (< cut-off μm)							
				<0.75	<0.82	<0.89	<0.96	<1.03	<1.10	<1.17	<1.25
6	14:04	Aqua	0.60	0.395	0.588	0.722	0.785	0.830	0.871	0.906	0.931
9	14:35	Aqua	0.92	0.425	0.619	0.756	0.817	0.858	0.893	0.922	0.943
12	15:05	Aqua	1.00	0.424	0.617	0.753	0.815	0.855	0.891	0.920	0.941
13	14:10	Aqua	0.93	0.423	0.616	0.752	0.814	0.855	0.890	0.919	0.940
14	14:55	Aqua	0.90	0.431	0.622	0.756	0.816	0.856	0.890	0.919	0.940
16	14:40	Aqua	0.67	0.419	0.604	0.734	0.795	0.837	0.873	0.905	0.929
17	13:45	Aqua	1.00	0.420	0.600	0.732	0.793	0.834	0.871	0.903	0.928
5	11:49	Terra	1.00	0.382	0.549	0.677	0.740	0.783	0.821	0.854	0.881
6	12:30	Terra	0.60	0.390	0.556	0.683	0.745	0.788	0.826	0.860	0.887
8	12:20	Terra	0.85	0.392	0.558	0.686	0.748	0.791	0.829	0.863	0.889
12	11:55	Terra	0.83	0.391	0.558	0.686	0.748	0.790	0.828	0.862	0.889
14	11:40	Terra	0.64	0.388	0.558	0.688	0.751	0.794	0.832	0.866	0.893
15	12:25	Terra	0.10	0.391	0.560	0.689	0.751	0.793	0.831	0.865	0.892

6. Summary

The CASPOL was used for the first time to make in-situ aerosol optical property measurements in the field as part of NASA's DISCOVER-AQ field campaign in Houston, Texas. Four potential large scale aerosol source locations that could be experienced at the Moody Tower were determined. Five day HYSPLIT back trajectories were used to identify periods of time when each aerosol source was dominant. Using the scattering signature technique, the three cases (Ship Channel, Ocean, Urban) could be distinguished from each other. Thus, the CASPOL can distinguish aerosol sources. However, these signatures were created using large sets of data ranging from 31 to 121 hours in length. If the CASPOL is to be used as an air quality monitoring tool it must be able to distinguish aerosol source in a shorter time period in order to provide source information in a timely manner.

To examine if the CASPOL can distinguish aerosol source in a short time period, the average number of particles potentially sampled by the CASPOL in one and eight hours was calculated, and used to create scattering signatures. One hour of data was not enough to distinguish the cases; however, with eight hours of data, the cases are distinguishable. This shows that the CASPOL can be successfully used to monitor air quality over a short period of time. Next, short sets of data were pulled by examining the backscatter intensity plotted over time. Peaks were identified and examined, and were approximately one half to three hours in length. The signatures created with these peaks were classified into two categories; local and transported, with local containing the Ship Channel and Urban sources, and transported being the Ocean source. It was found that if the number concentration is above approximately 100 – 200 L⁻¹, than the CASPOL can differentiate aerosol source with only a few hours worth of data. Thus, number concentration is of greater importance to how well the CASPOL can recreate

scattering signatures, and not the length of time the sample spans. Finally, an estimation of aerosol age was made for each case using O₃ concentrations and OC/EC ratios. The CASPOL has successfully been used to determine aerosol source even with a few hours of data. It can differentiate between aerosol sources by using the scattering signature technique developed by Glen and Brooks (2013). The information gathered by the CASPOL could be used to provide deterministic actions to the public to take actions to protect health, such as staying indoors if the CASPOL shows that particulate matter is from a certain source area. These results show that the CASPOL can be used to type aerosols in near real time and that it can have an important impact as an air quality monitoring and diagnostic tool.

CASPOL shows the potential for assessing the quality of satellite aerosol retrievals based on its measurements during the DISCOVER-AQ campaign. The aerosol size distribution derived from the CASPOL forward scattering measurements can be compared with MODIS FMF retrievals. The results show that Aqua MODIS FMF retrievals are consistent with CASPOL SMF measurements; the correlation coefficient between them is greater than 0.6. However, a large discrepancy is present between the variations of Terra MODIS FMF retrievals and CASPOL SMF measurements, the correlation coefficient of which is negative. The results are not sensitive to small perturbations of the particle size cutoff that makes a distinction between fine and coarse particles. In addition, both CASPOL and MODIS capture a decrease of the proportion of the coarse particle concentration from noon to the early afternoon. However, this MODIS observed variation might not be realistic, because of the potential uncertainties in the Terra MODIS FMF retrievals. The CASPOL data are of high quality only during 5-17 September, and hence the comparison cases are too few to make the results statistically significant. Longer CASPOL field measurements are needed to sufficiently evaluate the satellite aerosol-property retrievals in the future.

The backward polarized scattering measurements from CASPOL also have the potential to be compared with satellite aerosol observations. The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), the lidar instrument on the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite (Winker et al., 2009; Winker et al., 2010), provides a vertical profile of attenuated backscattered radiation at two wavelengths, 532 and 1064 nm. Since only 13-days of CASPOL data was of high quality during the DISCOVER-AQ campaign, too few CALIOP tracks adequately close to the Moody Tower were found during the short period for comparisons. Thus, long-term CASPOL measurements will be needed for successful comparisons with CALIOP products in the future. The depolarization ratio, a ratio of the perpendicular and parallel polarization components of backscatter coefficient, is profiled at 532 nm. Six pre-defined aerosol types are identified based mainly on CALIOP volume depolarization ratio measurements and associated extinction-to-backscatter ratios (lidar ratios) (Omar et al., 2009). In future studies, CALIOP volume depolarization ratio and attenuated backscatter intensity may be compared with those measured by CASPOL for examining the corresponding CALIOP aerosol typing criteria.

7. Recommendations for future work

We have the following recommendations:

- Make longer CASPOL field measurements so that statistically significant comparisons with satellite retrievals can be made.
- Terra MODIS FMF retrievals should be used in aerosol studies with great caution.
- Based on long-term CASPOL field measurements, assess the qualities of the CALIOP volume depolarization ratio and attenuated backscatter intensity.
- Conduct CASPOL and particle absorbance (such as aethalometer) measurements simultaneously, and then the statistics of scattering properties and the absorbance of particles can be theoretically linked. The linkage can help assess MODIS aerosol type assumptions using CASPOL measurements.

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