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3	Ground-based remote sensing of aerosol optical
4	properties and their radiative impacts
5	in the PRD region of China
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22	Abstract Aerosol direct radiative effects on surface irradiance were examined using seven years
23	of ground-based spectral radiation data collected at Panyu, the main atmospheric composition
24	monitoring station in the Pearl River Delta (PRD) region of China. Aerosol optical properties
25	were derived from sunphotometer measurements, which were then used as input to a radiative
26	transfer model to calculate broadband radiation. In the dry season (October to the following
27	February), the mean aerosol optical depth (AOD) at 550 nm was 0.535. More than 60% of AOD
28	values ranged from 0.2 to 0.6. The mean value of the Angstrom exponent (AE) calculated from
29	AOD retrievals at 440 nm and 870 nm was 1.31, with AE greater than 1.2 in 84.7% of the cases.
30	The mean single-scattering albedo (SSA) at 440 nm was 0.87. About 84.82% of SSA values were
31	between 0.8-0.92. Because few dust events take place in the PRD region, the coarse mode of
32	aerosols was negligible. However, the proportion of fine-mode, weakly absorbing particles was
33	about 9.52%. The magnitudes of AE, SSA, and AOD for this subset of data were 1.30, 0.96, and
34	0.65, respectively. About 90% of aerosols were dominated by fine-mode, strongly absorbing
35	particles with AE, SSA, and AOD equal to 1.35, 0.86, and 0.52, respectively. Because of the
36	strong absorption, the overall aerosol direct radiative effect was to heat the atmosphere and cool
37	the surface. The annual mean shortwave direct radiation forcing at the surface, inside the
38	atmosphere, and at the top of atmosphere was -33.51 \pm 8.41, 27.29 \pm 7.19, and -6.22 \pm 2.22 W·m ⁻² ,
39	respectively. Strongly absorbing aerosols not only changed the amount of global shortwave
40	radiation reaching the surface, but also dramatically changed the proportion of diffuse to direct
41	radiation at the surface. The annual mean reduction in surface global and direct radiation due to
42	aerosols was -37.50 ± 9.43 W·m ⁻² and -81.89 ± 18.42 W·m ⁻² , respectively. The presence of

43	aerosols resulted in 44.38 \pm 9.49 W·m ⁻² more diffuse radiation reaching the earth's surface. Due to
44	the significant radiative impacts of fine-mode, strongly absorbing particles, the properties of
45	carbonaceous aerosols in the PRD region and their impacts on regional climate change should be
46	further studied.
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48	Key Points:
49	• The aerosol properties of the PRD region in China are determined.
50	• The aerosol radiative forcing in the PRD is derived for the first time over a long period of
51	time.
52	• Providing a key constraint for model simulation of regional anthropogenic aerosol effects.
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54	1. Introduction
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56	Aerosols have been recognized as a major factor in determining global and regional climate
57	change [IPCC, 2007]. They play crucial roles in not only solar and thermal radiative transfer in
58	the atmosphere [Kosmopoulos et al., 2008], but also in the hydrologic cycle [Rosenfeld et al.,
59	2008; Clarke and Kapustin, 2010], the carbon cycle [Chameides et al., 1999], and some
60	important environmental issues such as acid rain and tropospheric ozone pollution [Wang, 1999].
61	The spatial and temporal variations in aerosols highlight the need to quantify aerosol radiative
62	properties and forcing on a regional scale. However, limited information on aerosol optical
63	properties and radiative forcing (ARF) are still major uncertainties [Guan et al., 2010] because of

differing aerosol types, compositions, and their generally large spatio-temporal variations. This is
 especially so in the vast territory of China.

Aerosol radiative impacts are strongly dependent on its optical properties. To better 66 understand the direct effect of aerosols on the surface energy budget and their associated impacts 67 on climate, some key parameters such as aerosol optical depth (AOD), the Angstrom exponent 68 (AE), single-scattering albedo (SSA), and the asymmetry factor (ASY) are needed. The latter 69 two parameters are difficult to retrieve from conventional satellite observations [Kaufman et al., 70 2002a, 2002b; Li et al., 2009]. Sunphotometer, lidar, aircraft, and satellite observations have 71 been used to analyze the spatial and temporal characteristics of the above parameters [Xia et al., 72 2007a; Guan et al., 2010; Liu et al., 2012; Che et al., 2014] and to estimate aerosol direct 73 radiative forcing [ADRF; Li et al., 2010; Alam et al., 2011; Choi and Chung, 2014]. The 74 magnitude of the global ADRF (due to absorption and scattering) has been estimated to range 75 from -0.85 W·m⁻² to -0.15 W·m⁻² [*Myhre et al.*, 2013]. Based upon data from 25 observation 76 sites spread across China, the ARF estimate over China shows that aerosols have little impact on 77 the atmosphere-surface system, but substantially warm up the atmosphere at the expense of 78 79 cooling the surface [Li et al., 2010]. Heavy aerosol loading and strong aerosol absorption over 80 northern and southeastern China lead to substantial reductions in direct and global shortwave irradiances at the surface while the instantaneous aerosol direct forcing at the top of the 81 atmosphere (TOA) is close to zero [Xia et al., 2007a; Liu et al., 2012]. 82

Until now, little knowledge about aerosol radiative properties and forcing in the Pearl River
Delta (PRD) region through ground-based remote sensing has been gained. This region is the

largest and most important economic zone in China where the open-door policy and drastic 85 economic reform took place in 1980s. Due to the rapid growth of industrialization and 86 urbanization over the past decades, the amounts and rates of emission of atmospheric pollutants 87 have increased dramatically. As a result, aerosols have become the most serious of atmospheric 88 pollutants in the region [Deng et al., 2006; Wu et al., 2008]. A recent study [Deng et al., 2011] 89 has shown that the probability that AOD at 550 nm is ≥ 0.6 and AOD at 340 nm is ≥ 1.0 is 47% 90 and 55%, respectively, during the dry season (October to January). Under current pollution 91 conditions in the region, more than half of the surface radiation, especially at ultraviolet 92 93 wavelengths, has been reduced by aerosols [Deng et al., 2011]. Part of this reduction is driven by industry that consumes a substantial amount of fossil fuel and coal in this region and releases 94 more absorbing soot and organic aerosols into the atmosphere [Cao, 2003, 2004; Cheng et al., 95 2008; Wu et al., 2009]. Since 2006, extensive measurements of aerosol optical, physical, and 96 97 chemical properties over the PRD region have been made.

In this paper, sunphotometer data from this region are used for the first time to: (1) analyze the frequency and monthly distributions of AOD, SSA, AE, and relative humidity (RH), (2) classify major aerosol types using AE and the scattering co-albedo (ω), and (3) examine ADRF over the region. Section 2 describes the observational site, the instrumentation at the site, and the methodology used in the study. Section 3 presents results and discussion. Conclusions are given in Section 4.

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105 **2. Data and methodology**

107 **2.1 Observational site and instrumentation**

Panyu (113.35°E, 23°N, 141 m above sea level) is located in the central part of the PRD region (Fig. 1). It is the main site of the atmospheric composition network run by the Guangdong Provincial Meteorological Bureau. Since 2011, this site has become part of the China Aerosol Remote Sensing Network (CARSNET). Measurements made at Panyu represent the basic characteristics of atmospheric composition in the PRD region [*Wu et al.*, 2009].

Characterization of atmospheric aerosols is done using a sunphotometer (CIMEL 113 Électronique, Paris, France). This instrument makes direct sun measurements every 15 min with 114 a 1.2° full field-of-view at 1020, 870P1, 670, 440, 870P2, 870, 936, and 870P3 nm (P indicates a 115 polarized filter). The full-width at half-maximum of each interference filter is 10 nm. 116 Measurements at 1020, 870, 670, and 440 nm are used to retrieve AOD and measurements at 936 117 118 nm quantify the total precipitable water vapor. The raw AOD is cloud-screened according to the method of Smirnov et al. [2000] with an uncertainty of $< \pm 0.01$ for wavelengths > 440 nm [Eck 119 et al., 1999]. 120

The CIMEL sunphotometer also measures sky radiances under almucantar and principal plane scenarios at 440, 670, 870 and 1020 nm following the optical air mass protocol described by *Holben et al.* [1998]. Aerosol inversion algorithms and software (SKYRAD.pack version 4.2) are used to retrieve aerosol optical and physical properties from sky radiances. Aerosol particle parameters such as SSA, the refractive index, and the particle volume size distribution are obtained by this technique [*Nakajima et al.*, 1996]. Calibration of the sunphotometer was

127	performed by CARSNET following the protocol developed by the Aerosol Robotic Network
128	(AERONET). Details about the inter-comparison calibration and sphere calibration carried out at
129	the Chinese Academy of Meteorological Sciences are given by Che et al. [2009] and Tao et al.
130	[2014].
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132	2.2 Methodology
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134	2.2.1 Retrievals of AOD, ω, and AE
135	The retrieval of columnar aerosol radiative properties from sky radiances requires an accurate
136	correction for the effects of multiple scattering and for the contribution of light reflected from the
137	earth's surface and scattered downward in the atmosphere [Valenzuela et al., 2012]. The
138	SKYRAD.pack version 4.2 software [Nakajima et al., 1996], which is commonly used to retrieve
<mark>139</mark>	aerosol optical and radiative parameters [e.g., Kim et al., 2005; Liu et al., 2008; Che et al., 2008;
<mark>140</mark>	Khatri et al., 2014; Wang et al., 2014], was used to calculate AOD and SSA by using a radiative
141	transfer code as well as linear and nonlinear inversion schemes. Additional input parameters for
142	the SKYRAD.pack included ozone data from the Atmospheric Infrared Sounder, and surface
143	albedo from the Moderate Resolution Imaging Spectroradiometer (MODIS) Level 2 Collection 5
144	spectral surface reflectance product. The parameter ω describes the loss of photons to absorption,
145	which is useful in identifying particle composition, especially carbonaceous particles [Corrigan
146	et al., 2006]. Here, ω is expressed as

 $\omega(\lambda) = 1 - SSA(\lambda), \qquad (1)$

148 where λ is the wavelength.

149 The AE is a good indicator of the size of particles and is given by the following equation:

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$$AE = -\frac{\ln\left[\frac{AOD(\lambda_1)}{AOD(\lambda_2)}\right]}{\ln\left[\frac{\lambda_1}{\lambda_2}\right]} , \qquad (2)$$

where λ_1 and λ_2 are the two wavelengths chosen to calculate AE. The wavelength pair of 440 nm/870 nm is used in this study. AE can range from negative values to greater than 1 [*Eck et al.*, 153 1999; *Gobbi et al.*, 2007], depending on the particle size.

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155 **2.2.2 Radiative forcing and radiative forcing efficiency**

Radiative fluxes and aerosol direct radiative effects of total, diffuse, and direct shortwave 156 irradiances in the broadband spectral range (0.25-4.00 µm) are calculated using the Santa 157 Barbara DISORT Atmospheric Radiative Transfer (SBDART) tool [Ricchiazzi et al., 1998]. 158 Previous studies have shown that SBDART simulations of downwelling broadband fluxes at the 159 surface agree well with ground-based measurements and that simulated upwelling fluxes at the 160 TOA are compatible with Clouds and Earth's Radiant Energy System satellite retrievals in terms 161 of absolute differences [Xia et al., 2007a; Li et al., 2010]. SBDART simulations and 162 measurements of broadband irradiance agree to within 3% [Halthore et al., 2005]. This model 163 has been used to estimate aerosol radiation forcing in China [Xia et al., 2007c, 2007d; Liu et al., 164 165 2007; Li et al., 2010].

166 The AOD, SSA, AE, and ASY at four AERONET wavelengths (i.e., 440, 675, 870, and 1020

nm) have been used to interpolate and extrapolate into the spectral divisions of the SBDART model [*Xia et al.*, 2007a, 2007b]. The value for the ASY from the AERONET site in Hong Kong was used here and assumed to be constant. Other input parameters for the SBDART model include ozone data from the Atmospheric Infrared Sounder, water vapor from the MODIS atmospheric profiles product, and surface albedo from the MODIS Level 2 Collection 5 spectral surface reflectance product [*Liu et al.*, 2012].

ADRF is commonly used to quantify the direct effect of aerosols on the atmospheric energy budget. The SBDART model was run twice to simulate shortwave irradiances with and without aerosol particles under cloud-free conditions, and then used to determine ADRF at the surface (F_{SFC}) and at the TOA (F_{TOA}). The aerosol radiative forcing within the atmosphere (F_{ATM}) is defined as the difference between F_{TOA} and F_{SFC} . The definition of diurnal mean radiative forcing is often given as

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$$dF = \frac{1}{24} \int F(t) dt$$
, (3)

where F(t) represents instantaneous radiative forcing values. The combined error caused by uncertainties in main input parameters, including AOD, AE, SSA, ASY, surface reflectance, and ozone amounts, is 8.76±3.44 W·m⁻² [*Li et al.*, 2010].

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184 **2.2.3 Data description**

The data derived from the sky radiation algorithm was used to analyze AOD, SSA, and AE at a temporal resolution of 30 min. The temporal resolution of ADRF is the same as that of the aerosol property parameters. The sunphotometer collected 1219 inversion measurements from

188	January 2006 to December 2012. However, observations were not continuous due to instrument
189	calibration and maintenance [Zhu et al., 2014]. In addition, the PRD region is typically cloudy
190	and rainy during the wet seasons (spring and summer), which greatly influences instrument
191	observations and data inversions. For this reason, this study focuses on the dry season (October
192	to the following February). Table 1 summarizes the number of days in each of these months with
193	inversion data available and the total number of data points in each of these months.
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195	3. Results and discussion
196	
197	3.1 Aerosol optical properties
198	Figure 2 shows the frequency distributions of AOD at 550 nm, AE, SSA at 440 nm, and RH
199	over the PRD region. The annual mean RH was 57.53%. The most frequent value of RH ranged
200	from 50-60%, typical of the dry season in the PRD region (Fig. 2d).
201	AOD ranged from $\sim 0.1-1.3$ (Fig. 2a). More than 60% of AOD values were between 0.2 and
202	0.6, with a maximum frequency (19.13%) at AOD equal to 0.3. The proportions of AOD values
203	below and above this AOD range were 23.65% and 16.09%, respectively.
204	The AE is a good indicator of particle size. Values of AE < 0.75 indicate mainly coarse-mode
<mark>205</mark>	particles and values of $AE > 1.2$ indicate mainly fine-mode particles. Values of AE between 0.75
206	and 1.2 suggest complicated aerosol modes [Eck et al., 2005; Yang et al., 2011]. Figure 2b shows
207	that AE varied from 0.7 to 1.6. About 15.4% of the particles had $AE < 1.2$ and up to 84.7% of the
208	particles had $AE > 1.2$, indicating that aerosols in the PRD region were of the fine-mode variety.

Values for SSA ranged from 0.58 to 0.98 with the most frequent value equal to ~0.88 and the mean value equal to 0.81. About 84.8% of SSA values ranged from 0.8 to 0.92, suggesting that the aerosols were strongly absorbing.

Figure 3 shows notched box whisker plots of monthly AOD at 550 nm, AE, SSA at 440 nm, and RH over the PRD region. From October to February, AOD varied from 0.47 to 0.75, with a maximum in February and a minimum in December (Fig. 3a). The annual mean value was 0.535, which is relatively less than values of AOD reported at sites in eastern China like Lin'an [*Che et al.*, 2009] and Taihu [*Liu et al.*, 2012]. This is likely because only the dry season was considered here.

Monthly changes in AE were not apparent (Fig. 3b). The magnitude of AE ranged from 1.31 to 218 1.38 and was relatively higher in February and October and lower in December. The mean value 219 of AE was 1.33, which is higher than the values retrieved at Xinglong [Zhu et al., 2013], and in 220 221 northern [Xia et al., 2007b] and northeastern [Xin et al., 2011] China. Local pollutants and the products of their photochemical reactions are the prime sources of aerosols in the PRD region, 222 while the other sites mentioned are in northern China, which is more susceptible to coarse-mode 223 mineral dust. As a result, the particles have fine-mode properties which reach their maximum 224 225 values in February. The high AOD and AE in February may be related to the high water content in the atmosphere during that month (Fig. 3d) and to pollutants advected in from cities around 226 227 the PRD region.

The mean value of SSA was 0.87 (Fig. 3c), which is lower than the values reported at Xianghe [*Li et al.*, 2007] and Xinglong [*Zhu et al.*, 2014]. This suggests that aerosols are strongly absorbing in the PRD region. There was little month-by-month change in SSA, but relatively higher values are seen in October and February. This could be due to the presence of water-soluble aerosols at higher RH levels, resulting in fine-mode particle growth and an increase in the light scattering coefficient [*Kotchenruther and Hobbs*, 1998]. The RH level was lower in October (Fig. 3d), but AOD, AE, and SSA values were higher. This may be related to the stronger solar radiation present in October which can activate photochemical reactions and yield more secondary organic aerosols [*Liu et al.*, 2009; *Lee et al.*, 2010].

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238 **3.2 Aerosol classification using AE and ω**

The wavelength dependence of ω provides more information about the physico-chemical 239 properties of aerosol types and AE provides information about particle size [Logan et al., 2013]. 240 Therefore, these two parameters were combined to classify aerosol types in the PRD region. 241 242 As done by Logan et al. (2013), $\omega = 0.07$ was chosen as the demarcation line between strongly and weakly absorbing aerosols, and AE = 0.75 was chosen as the demarcation line 243 between fine-mode and coarse-mode particles. Four regions are thus defined: Region I where 244 fine-mode, weakly absorbing particles dominate; Region II where fine-mode, strongly absorbing 245 246 particles dominate; Region III where coarse-mode, strongly absorbing mineral dust particles 247 dominate; and Region IV where coarse-mode, weakly absorbing particles dominate, e.g., desert 248 aerosols.

Figure 4 shows mean classification results for the months of October to February of the years
2006-2012. About 9.5% of all data points fell in Region I, i.e., the region representing fine-mode,

weakly absorbing particles. There was a maximum in October (14.68%), a minimum in January (6.16%), and equal proportions (12%) in November and February. AOD (equal to 0.65) was significantly positively correlated with the aerosol proportions in this region. Up to 90% of particles fell in region II, with a maximum proportion of 93% in January and a minimum proportion of 83.49% in October. In addition, the majority of data points were centered on ω values between 0.07 and 0.17, and AE values between 1.2 and 1.5. As in region I, the proportion in November and February was similar (about 87%). Because few dust events take place in the

PRD region, the number of data points in the coarse-mode regions (III and IV) was negligible.

Overall, fine-mode, strongly absorbing aerosol particles were dominant in the study area with mean AOD, AE, and SSA values equal to 0.52, 1.35, and 0.86. Given the location of Panyu, pollutants at the site were likely a combination of carbon aerosols (e.g., organic and black carbon) or mixtures of sulfate, nitrite, and carbon aerosols generated by vehicles and stoves [*Andreae et al.*, 2008; *Zheng et al.*, 2011; *Wang et al.*, 2012]. Compared to region II, aerosols in region I had a higher AOD (0.65) and larger particle size (AE = 1.30). The AOD was significantly influenced by the proportion of particle amount (r = 0.92).

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267 **3.3 Diurnal radiative effects**

In the PRD region, high aerosol loading prevails all year round with an annual mean AOD equal to 0.54. As a result, aerosols are expected to seriously impact irradiances travelling between the atmosphere and the earth's surface. They are also expected to impact the partitioning between the direct and diffuse components of the surface irradiance. Data from region II are analyzed here because this region contains the largest number of data points.

ADRF varies significantly in different regions and at different time scales [*Yu et al.*, 2006; *Xia et al.*, 2007a; *Li et al.*, 2010]. Global mean values of F_{SFC} , F_{TOA} , and F_{ATM} from observations over land are -11.9, -4.9, and 7.0 W·m⁻², respectively, and from modeling are -7.6, -3.0, and 4.0 W·m⁻², respectively [*Yu et al.*, 2006, 2009]. *Li et al.* [2010] found that F_{SFC} , F_{TOA} , and F_{ATM} was -15.7 ±9.0, 0.3 ± 1.6, and 16.0 ± 9.2 W·m⁻², respectively, over China, and that F_{SFC} could reach -32.8 W·m⁻² in northern China.

Figure 5 shows the monthly mean diurnal ADRF in the PRD region. The annual mean F_{SFC} , 279 F_{ATM} , and F_{TOA} was -33.51±8.41, 27.29±7.19, and -6.22±2.22 W·m⁻², respectively. The large 280 negative F_{SFC} and positive F_{ATM} implies strong cooling at the surface and strong warming in the 281 atmosphere, which were likely induced by the large values of AOD and small values of SSA 282 [Lee et al., 2007; Zhao and Li, 2007]. The largest monthly values are seen in February when F_{SFC} 283 and F_{ATM} reach -42.88 and 36.06 W·m⁻², respectively. Li et al. [2010] reported that the largest 284 negative F_{SFC} (-20 to -32 W·m⁻²) and positive F_{ATM} (20-40 W·m⁻²) were found over east China 285 in 2005, and the second largest negative F_{SFC} (-20 to -30 W·m⁻²) and positive F_{ATM} (20-30 W·m⁻²) 286 were found over the PRD region. The F_{TOA} ranged from 0-10 W·m⁻². Liu et al. [2012] found that 287 at Taihu, annual mean $F_{SFC},$ $F_{TOA},$ and F_{ATM} were -34.8± 9.1, -8.2 ± 4.8, and 26.7 ± 9.4 $W \cdot m^{-2}$, 288 respectively. The magnitude of mean F_{SFC} in this study is similar to that reported in east China 289 and at Taihu. If the same time scale is considered (October to the following February), the F_{SFC} 290 291 in the PRD region was stronger. Given that this study is based on data collected a few years after 292 2005, this suggests that anthropogenic aerosol contamination has become a more serious

problem in the region and has changed the energy balance at the earth's surface there.

The aerosol radiative forcing efficiency (ADRFE), defined as forcing per unit AOD, is an 294 indicator of the radiative forcing potential of a given type of composite aerosol [Pathak et al., 295 2010]. The annual mean ADRFE in this study was -68.25±17.18 Wm⁻² τ^{-1} at the surface, 296 -12.67±4.52 Wm⁻² τ^{-1} at the TOA, and 55.58±14.64 Wm⁻² τ^{-1} within the atmosphere (Fig. 6). 297 These values are significantly higher than those reported by Li et al. [2010] over China (-35.1 298 Wm⁻² τ^{-1} , -0.5 Wm⁻² τ^{-1} , and 34.5 Wm⁻² τ^{-1} at the surface, the TOA, and within the atmosphere, 299 respectively). The ADRFE at the surface was higher than that estimated for Xianghe (-65.4±4.7 300 Wm⁻² τ^{-1}), Taihu (-54.4±5.3 Wm⁻² τ^{-1}), and northern China (-55.2 Wm⁻² τ^{-1}). However, these 301 values were less than estimates for the South Asian region (-89.4 Wm⁻² τ^{-1}) where black carbon 302 and organic carbon aerosols dominate [Wang et al., 2007]. The values found in this study are 303 more in line with those calculated for the Maldives [-72.2 \pm 5.5 Wm⁻² τ^{-1} , Bush and Valero, 2002] 304 and over the Indian Ocean [-75 Wm⁻² τ^{-1} , *Ramanathan et al.*, 2001]. 305

Aerosol loading not only changes radiative forcing in the earth-atmosphere system, but also 306 alters the proportion of direct, global, and diffuse radiation at the surface. These results have 307 implications in the study of the global carbon cycle [Gu et al., 2002]. In eastern China, the mean 308 309 direct shortwave flux reaching the surface in autumn to winter was reduced by about -68.5 $W \cdot m^{-2}$ and diffuse shortwave fluxes were enhanced by 42.5 $W \cdot m^{-2}$ in the presence of aerosols 310 [Liu et al., 2012]. Similar results were obtained in the PRD region, but the radiative forcing was 311 stronger due to the presence of strongly absorbing aerosols (Fig. 7). During the dry season of the 312 313 PRD region, global and direct radiative fluxes at the surface decreased by -37.50±9.43 and -81.89±18.42 W⋅m⁻², respectively, and diffuse shortwave fluxes increased by 44.38±9.49 W⋅m⁻².
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316 4. Conclusions

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Aerosol optical and radiative properties, including frequency, temporal variability, classification and radiative forcing, have been analyzed using seven years of ground-based measurements made at Panyu, an observation site in the PRD region. The major conclusions are summarized as follows:

(1) In the dry season (October to the following February), aerosols in the PRD region were
fine-mode, strongly absorbing particles, with annual mean values of AE and SSA equal to 1.33
and 0.87, respectively. The annual mean AOD at 550 nm was 0.535, which is relatively smaller
in magnitude than AOD at 550 nm retrievals made at sites in eastern China.

(2) To sort out the major aerosol types and their radiative impacts in this region, AE and ω were used. Up to 90% of aerosols were dominated by fine-mode, strongly absorbing particles with mean AE = 1.35, ω = 0.14, and AOD = 0.52. The proportion of fine-mode, weakly absorbing particles was about 9.52%, with AE = 1.30, ω =0.04, and AOD = 0.65. Due to the minimal presence of dust in the PRD region, the aerosol coarse mode was negligible.

(3) Because of the strong absorption, the overall aerosol direct radiative effect is to
significantly heat the atmosphere and cool the surface, and to alter the proportion of direct,
global, and diffuse radiation at the surface. The annual mean shortwave direct radiation forcing
at the surface, inside the atmosphere, and at the top of atmosphere was -33.51±8.41, 27.29±7.19,

and $-6.22\pm2.22 \text{ W}\cdot\text{m}^{-2}$, respectively. The annual mean reduction in global and direct radiation due to aerosols was $-37.50\pm9.43 \text{ W}\cdot\text{m}^{-2}$ and $-81.89\pm18.42 \text{ W}\cdot\text{m}^{-2}$, respectively. The presence of aerosols caused $44.38\pm9.49 \text{ W}\cdot\text{m}^{-2}$ more diffuse radiation to reach the earth's surface.

(4) Given that aerosol radiative properties and forcing have not yet been well studied in the PRD region compared to the rest of China, these results can help constrain uncertainties in estimating regional anthropogenic aerosol radiative forcing. In future work, we will pay more attention on the properties of carbonaceous aerosols in the PRD region and their impacts on regional climate change.

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344 Acknowledgements

This research work is funded by the National Natural Science Foundation of China (41175117, 345 40875090, 41373116), the MOST 973 Program (2013CB955804), the National "973" Program 346 (2011CB403400), the Natural Science Foundation of Guangdong Province (S2012010008749), 347 the National Natural Science Foundation of China and Guangdong Province Joint Fund 348 (U1201232), the Special Research Project of Public Service Sectors 349 (Weather) (GYHY201306042) and the Science and Technology Sponsorship Program of Guangdong 350 351 Province (2010A030200012). The availability of the data used for this study is fully described in section 2 of this paper. The MODIS data were downloaded from the NASA Goddard Space 352 Flight Center. we also thank SKYRAD.pack and SBDART model. The constructive 353 354 suggestions from the anonymous reviewers are greatly appreciated.

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- 546 Tables

Table 1. Number of days in each month with inversion data (AOD, SSA, and AE) available and the total

549 number of data points in each month.

Voor	Number of days				Voor	Number of instantaneous data					
i cai	Oct	Nov	Dec	Jan	Feb	y ear	Oct	Nov	Dec	Jan	Feb
2006	4	12	23	0	0	2006	24	64	102	0	0
2007	18	10	18	15	0	2007	47	57	67	73	0
2008	0	0	0	11	11	2008	0	0	0	54	32
2009	0	0	0	23	8	2009	0	0	0	169	42
2010	10	25	18	0	0	2010	38	119	105	0	0
2011	0	7	20	14	10	2011	0	33	99	51	26
2012	0	0	0	4	2	2012	0	0	0	10	7

- - -



558 Figures



560 Fig. 1. Location of Panyu, the observation site in the PRD region. Mean hourly wind speeds and directions at







for the PRD region. The black curve represents the Gaussian distribution.



Fig. 3. Monthly distributions (green dots) of (a) AOD at 550 nm, (b) the 440-870 nm AE, (c) SSA at 440 nm, and (d) RH over the PRD region. The blue curves through the green dots represent Gaussian distributions. The notches in the notched box whisker plots represent the 95% confidence levels of the median values. Non-overlapping notches indicate that the medians are significantly different from each other.



Fig. 4. Classification of the mean physico-chemical properties of aerosols into four regions using AE and ω at 440 nm. The color scale represents the relative density of points. Panels (a) - (f) correspond to October, November, December, January, February, and total data, respectively, of the years 2006-2012.



Fig. 5. Monthly mean diurnal aerosol direct radiative forcing (ADRF) at the surface (F_{SFC}), at the top of the







Fig. 6. Diurnal ADRF at the surface (SRF, black), top of the atmosphere (TOA, blue), and within the atmosphere (ATM, red) as a function of AOD at 550 nm over the PRD region. Best-fit lines through the points are shown and linear regression functions and statistics (coefficient of determination, R², and standard deviation, SD) are given.





633 Fig. 7. Aerosol-induced monthly variations in surface downwelling global, direct, and diffuse shortwave

634 irradiances.