#### 1 Manuscript No.: acp-2017-165

2 Journal: ACP

The revised manuscript entitled "Measurement of scattering and absorption
properties of dust aerosol in a Gobi farmland region of northwest China—a
potential anthropogenic influence" by Jianrong Bi, et al.

6

#### 7 **Response to Referee#1:**

8 We are grateful to the Editor's and Referee#1's insightful and constructive comments 9 for this manuscript! We have carefully checked and revised the whole manuscript 10 according to Referee#1's comments, which are helpful and valuable for greatly 11 improving our manuscript. Please find a point-by-point reply to the issues as follows 12 (highlighted in the blue font). And we have also uploaded the file of "Response 13 to-Referee#1(acp-2017-165).pdf".

14

### 15 General comments:

Dust aerosol in remote Taklimakan Desert and Gobi deserts of northwest China is 16 thought to be hardly affected by human activities, due to sparse population. The 17 authors conducted a comprehensive field measurement in a Gobi farmland region of 18 northwest China, and demonstrated a potential anthropogenic influence on dust 19 physicochemical properties using multiple ground-based active and passive sensors. 20 The agricultural operations and biomass burning from crop residue prior to growing 21 season were well documented to produce significant impacts on elevated dust 22 loadings and absorption characteristics in Dunhuang farmland during spring of 2012. 23 24 The findings of this study are very interesting and would help to improve our understanding of the interaction among dust aerosol, atmospheric chemistry, and 25 climate change in desert source region. And I suggest that the authors should carry out 26 long-term and continuous measurements of mineral dust at remote Gobi deserts in 27 northwest China, to quantify the potential anthropogenic contributions on regional 28 climatic and environmental changes. I think the English wring is fine, and I 29 recommend this manuscript is appropriate for publishing after minor revision. 30

31	Response: Thank you very much for the Referee's good suggestions and the	
32	acceptance of this work. Indeed, this study only covers several months in spring	
33	during intensive period and it is indispensable to acquire long-term measurements of	
34	mineral dust for fully understanding the potential anthropogenic contributions on	
35	regional environmental and climatic changes. Hence, we have set up two permanent	
36	field observatories (SACOL and Dunhuang) in northwest China to continuously	
37	measure mineral dust since 2013, and will obtain more valuable findings, which will	
38	help quantify the anthropogenic contributions of dust aerosol in remote desert source	
39	region.	
40		
41	Minor comments:	
42	1. Abstract, Page 1, line 27: "In the afternoon (13:00–18:00 LT)"	
43	$\Rightarrow$ Change to "In the afternoon (13:00–18:00 LT, local time)". When an abbreviation	
44	firstly appears in the manuscript, please give the full name.	
45	<b>Response:</b> We have changed "In the afternoon (13:00–18:00 LT)" to "In the afternoon	
46	(13:00–18:00 LT, local time)" in Line 27 and modified the corresponding places in the	
47	entire context.	
48	A	<b>带格式的:</b> 字体颜色:蓝色
49	2. Page 3, line 78: "(i.e., hematite and goethite)"	
50	$\Rightarrow$ Change to "(i.e. hematite and goethite)"	
51	<b>Response:</b> We have changed to "(i.e. hematite and goethite)" in Line 78.	
52	A	<b>带格式的:</b> 字体颜色:蓝色
53	3. Page 4, line 90: "(i.e., Mongolia Gobi desert)"	
54	$\Rightarrow$ Change to "(i.e. Inner Mongolian Gobi desert)"	
55	Response: We have changed to "(i.e. Inner Mongolian Gobi desert)" in Line 90.	
56		
57	4. Page 4, line 111: "close to the east edge of Kumtag Desert"	
58	$\Rightarrow$ Change to "close to the eastern edge of Kumtag Desert"	
59	Response: We have changed to "close to the eastern edge of Kumtag Desert" in Line	
60	111.	
	2	

51		
52	5. Page 5, line 130: "to the southeast"	
53	$\Rightarrow$ Change to "to the southwest"	
54	<b>Response:</b> We have changed to "to the southwest" in Line 130.	
55		
56	6. Page 6, line 154: "High AI values (>0.7) distributions"	
57	$\Rightarrow$ Change to "The distributions of high AI values (>0.7)"	
58	Response: We have changed "High AI values (>0.7) distributions" to "The	
59	distributions of high AI values (>0.7)" in Line 154.	
70	A	带格式的:字体颜色:蓝色
71	7. Page 9, line 265: "(i.e., Mongolia cyclones)"	
72	$\Rightarrow$ Change to "(i.e. Mongolian cyclone)"	
73	Response: We have changed "(i.e., Mongolia cyclones)" to "(i.e. Mongolian	
74	cyclone)" in Line 265.	
75		
76	8. Page 12, line 331: "2 to 4 km"	
77	$\Rightarrow$ Change to "4 km"	
78	<b>Response:</b> We have changed "2 to 4 km" to "4 km" in Line 331.	
79		
30	9. Page 12, line 332: "which was within the planetary boundary layer (PBL)"	
31	$\Rightarrow$ Change to "which was above the planetary boundary layer (PBL)"	
32	Response: We have changed to "which was above the planetary boundary layer	
33	(PBL)" in Line 332.	
34		
35	10. Page 14, line 402: "Likewise"	
36	$\Rightarrow$ Change to "Similarly"	
37	<b>Response:</b> We have changed "Likewise" to "Similarly" in Line 402.	
38		
39	11. Page 19, line 563: "atmospheric boundary layer structure"	
90	$\Rightarrow$ Change to "the structure of atmospheric boundary layer"	
	5	

**Response:** We have changed "atmospheric boundary layer structure" to "the structure

- 92 of atmospheric boundary layer" in Line 563.
- 93

94 12. Page 20, line 575: "lager"

95  $\Rightarrow$  Change to "larger"

- 96 **Response:** We have changed "lager" to "larger" in Line 575.
- 97

98 13. Page 21, line 614: "The findings of this study directly demonstrated mineral dust"

- 99  $\Rightarrow$  Change to "The findings of this study directly demonstrated that mineral dust"
- 100 **Response:** We have changed to "The findings of this study directly demonstrated that

101 mineral dust" in Line 614.

102

#### 103 **Response to Referee#2:**

We appreciate the Editor and Referee#2's valuable and constructive comments for this manuscript, which greatly assist in improving the quality of the original manuscript! We have carefully checked and revised the whole manuscript according to Referee#2's comments. Please find a point-by-point reply to the issues as follows (highlighted in the blue font). And we have also uploaded the file of 'acp-2017-165-supplement.pdf'.

110

## 111 General comments:

This manuscript presents the measurement of scattering and absorption properties of dust aerosol from a comprehensive field campaign in a Gobi farmland region of northwest China during spring 2012. Overall, the manuscript could make a good contribution to the scientific research by providing useful scientific knowledge on the interaction among dust aerosol, atmospheric chemistry, and climate change in desert source region. However, I believe that the manuscript needs the following minor revisions before it is accepted for publication by ACP.

**Response:** Thank you very much for the Referee's insightful suggestions andconstructive comments on this manuscript. We have carefully checked and revised the

**带格式的:**字体:小四,非加粗, 字体颜色:蓝色 whole manuscript according to Referee#2's comments. Please find a point-by-pointreply to the issues as follows.

123

## 124 Minor comments:

Lines 22–24: Please present the more results and discussions on the statement in
 the text about the statement in the abstract that "The anthropogenic dust produced by
 agricultural cultivations (e.g., land planning, plowing, and disking) exerted a
 significant superimposed effect on high dust concentrations in Dunhuang farmland
 prior to the growing season (i.e., from 1 April to May)."

**Response:** We have presented visual photos of a variety of agricultural cultivations in 130 131 Dunhuang farmland (nearby SACOL's Mobile Facility) prior to the growing season (i.e. from 1 April to 10 May, 2012), as shown in Figure S1. Diverse agricultural operations 132 (e.g., land planning, plowing, disking and laying plastic mulch) were carried out in 133 134 loose and bare Dunhuang farmland from 1 April to 10 May, 2012, which produced massive soil dust into the atmosphere, especially under strong surface winds (see 135 Figure S1a-c). Therefore, the mass concentrations of particulate matter  $(PM_{10})$  in the 136 source and adjacent downwind regions (including SACOL's Mobile Facility) were 137 significantly elevated by these human activities. In contrast, the crops in Dunhuang 138 farmland gradually become green since 10 May, 2012, indicating the coming of 139 growth season (Figure S1f). 140

141 We also added more discussions about this in the context (Page 10, Lines 272–278).

- 142 Please check our revised manuscript in detailed.
- 143

144 2. Lines 25–27: It is a misleading conclusion that "Strong south valley wind and 145 vertical mixing in daytime scavenged the pollution and weak northeast mountain wind 146 and stable inversion layer at night favorably accumulated the air pollutants near the 147 surface." Please follow the diurnal changes of winds and PM<sub>10</sub> in Figs. 4 and 6.

148 Response: Thank you very much for your suggestions! The conclusion here

149 corresponds to the diurnal changes of winds and  $PM_{10}$  in Figs. 4 and 6, which can be 150 interpreted by classical mountain-valley wind circulation. Please refer to more

- detailed explanations in Pages 12–13, Lines 336–359.
- 152

153 3. It could be unnecessary to present the wind fields at 500 hPa and 850 hPa levels 154 from the MERRA reanalysis products in Fig. 7, because the dust aerosols in a Gobi 155 farmland region of northwest China are mostly the local emissions and a 156 short-distance transport to the measurement site within the boundary layer.

Response: Thank you very much for your good comments! We mainly intend to 157 elucidate two points using Fig. 7. Firstly, the selected three heavy dust events (i.e. 30 158 April, 1 May, and 10 June) were triggered by different synoptic cyclones. East Asian 159 region was governed by the powerful and stable westerlies at 500 hPa height on 30 160 161 April and 1 May 2012, whereas strong Mongolian cyclones at 500 hPa upper atmosphere hovered about the southern Mongolia on 10 June 2012. Secondly, the 162 ground-based measured strong northeast and east winds (> 10 ms<sup>-1</sup>) under three dust 163 events were completely consistent with the wind fields at 850 hPa levels from the 164 MERRA reanalysis products, which indicated the studied dust events were regional 165 scales instead of local scales. 166

167

168 4. Line 532: "mesoscale cyclones" should be "synoptic cyclones".

169 Response: We have changed "mesoscale cyclones" to "synoptic cyclones" in Line170 532.

171

5. Lines 570-577: It is an interesting result that Figure 10d displays that "the DLW 172 values under dusty cases were always greater than that in clear-sky cases, with the 173 total average differences of +40~+60 Wm<sup>-2</sup>". However, the interpretation is 174 unconvinced. From Fig. 10d, it could be seen that the warming dust layer could 175 enhance that surface DLW with a large (+40~+60 Wm<sup>-2</sup>: not a few percentages!) 176 contribution to the increased DLW. It is unreasonable that the potential greenhouse 177 gases in the atmosphere could substantially affect the DLW differences between dusty 178 and clear-sky cases (Fig. 10d). Also, please present the measured cloud cover or RH 179 on April 9 to support the statement that "it is partly attributable to the higher RH 180

values on 9 June than that in other days."

182 Response: Thank you very much for your insightful and valuable comments! Indeed,
183 the warming dust layer could enhance that surface DLW with a large contribution to
184 the increased DLW (+40~+60 Wm<sup>-2</sup>: not a few percentages!). Hence, we have
185 changed "contribute a few percentages to the increased DLW" to "contribute a large
186 percentages to the increased DLW" in Line 576.

"the potential greenhouse gases in the atmosphere could substantially affect the DLW 187 variations.": "the greenhouse gases" in the manuscript represent the presence of water 188 vapor or clouds in the atmosphere, which causes confusion. Therefore, we have 189 changed "This is because the potential greenhouse gases in the atmosphere could 190 substantially affect the DLW variations." to "This is because the potential water vapor 191 in the atmosphere could substantially affect the DLW variations." in Lines 573-575. 192 Meanwhile, we have presented the diurnal variations of 10-second average relative 193 humidity (RH, %) under completely clear-sky conditions (14 May, 29 May, and 9 194 June) and dust events (30 April and 10 June) in Dunhuang farmland, as shown in 195 Figure S2. We can clearly see that the RH on 9 June keeps the highest value in the 196 whole day, which is greater than that in other cloudless or dusty days. 197

198

6. Please improve the quality of all the Figs., with clarifying the figure captions,
such as horizontal wind vector in Figs. 4, near surface wind in Figs. 6 and 8, and the
same color curves for all the Figs. 10a, 10b, 10c and 10d.

202 **Response:** Thank you very much for your valuable comments for improving the

203 quality of this manuscript! We have improved the quality of all the Figs. in the context

7

and corrected the same color curves for all the Figs. 10a, 10b, 10c, and 10d. Please

205 refer to the revised manuscript in detail.

206

207

209	Measurement of scattering and absorption properties of dust aerosol
210	in a Gobi farmland region of northwest China—a potential
211	anthropogenic influence
212	Jianrong Bi, Jianping Huang, Jinsen Shi, Zhiyuan Hu, Tian Zhou, Guolong Zhang,
213	Zhongwei Huang, Xin Wang, and Hongchun Jin
214	
215	Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of
216	Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China
217	
218	Correspondence to: Jianping Huang (hjp@lzu.edu.cn)
219	
220	Abstract. We conducted a comprehensive field campaign on exploring the optical
221	characteristics of mineral dust in Dunhuang farmland nearby the Gobi deserts of
<b>ว</b> วว	northwest China during spring of 2012. The day-to-day and diurnal variations of dust

northwest China during spring of 2012. The day-to-day and diurnal variations of dust 222 aerosol showed prominent features throughout the experiment, primarily attributable 223 to frequent dust events and local anthropogenic emissions. The overall average mass 224 225 concentration of the particulate matter with an aerodynamic diameter less than 10 µm (PM<sub>10</sub>), light scattering coefficient ( $\sigma_{sp,670}$ ), absorption coefficient ( $\sigma_{ap,670}$ ), and 226 single-scattering albedo (SSA<sub>670</sub>) were 113±169 µgm<sup>-3</sup>, 53.3±74.8 Mm<sup>-1</sup>, 3.2±2.4 227 Mm<sup>-1</sup>, and 0.913±0.05, which were comparable to the background levels in southern 228 United States, but smaller than that in the eastern and other northwestern China. The 229 anthropogenic dust produced by agricultural cultivations (e.g., land planning, plowing, 230 231 and disking) exerted a significant superimposed effect on high dust concentrations in 232 Dunhuang farmland prior to the growing season (i.e., from 1 April to 10 May). Strong 233 south valley wind and vertical mixing in daytime scavenged the pollution and weak northeast mountain wind and stable inversion layer at night favorably accumulated the 234 air pollutants near the surface. In the afternoon (13:00-18:00 LT, local time), mean 235 SSA670 was 0.945±0.04 that was predominant by dust particles, whereas finer 236

particles and lower SSA<sub>670</sub> values (~0.90–0.92) were measured at night, suggesting the potential influence by the mixed dust-pollutants. During a typical biomass burning event on 4 April 2012,  $\sigma_{ap,670}$  changed-increased from ~2.0 Mm<sup>-1</sup> to 4.75 Mm<sup>-1</sup> and SSA<sub>670</sub> changed from ~0.90 to ~0.83, implying remarkable modification of aerosol absorptive properties induced by human activities. The findings of this study would help to advance an in-depth understanding of the interaction among dust aerosol, atmospheric chemistry, and climate change in desert source region.

#### **1. Introduction**

245 Asian mineral dust (also known as dust aerosol) in the atmosphere is deemed to exert a profound impact on air quality and climate change. It can perturb the energy 246 budget of the Earth system directly through scattering and absorption of solar and 247 248 terrestrial radiation (Huang et al., 2009, 2014; Ge et al., 2010; Li et al., 2016) and 249 indirectly by altering cloud microphysical processes and related hydrological cycle (Rosenfeld et al., 2001; J. Huang et al., 2005, 2006, 2010; Yin and Chen, 2007; W. 250 251 Wang et al., 2010; Creamean et al., 2013; Wu et al., 2016), as well as modifying snow and ice surface albedo (Aoki et al., 2006; Huang et al., 2011; Wang et al., 2013; Qian 252 et al., 2014). In addition, alkaline mineral dust carries abundant organic matters and 253 254 iron ions deposited on the surface of earth, and hence affects biomass productivity of in the North Pacific Ocean and relevant atmosphere-ocean carbon exchange, which 255 plays a pivotal role in the global biogeochemical cycle and carbon cycle (Cao et al., 256 257 2005; Jickells et al., 2005; Maher et al., 2010; Shao et al., 2011).

The Taklimakan Desert in northwestern China and Gobi Deserts in southern 258 Mongolia and northern China are widely regarded as two major active centers of dust 259 storms in East Asia (Sun et al., 2001; Zhao et al., 2006; Wang et al., 2008; Ge et al., 260 261 2016). These extensive arid and desert zones frequently generate a great deal of tiny 262 soil particles every spring that are uplifted and entrained into the free atmosphere layer via cold frontal cyclones (Zhang et al., 1997; Aoki et al., 2005; Kai et al., 2008; 263 J. Huang et al., 2009, 2010, 2014). Affected by mid-latitude prevailing westerlies, 264 these dust particles can transport long distances on a subcontinental scale, even sweep 265

266 across the remote Pacific Ocean and occasionally arrive at the west coast of North America during the peak seasons of strong dust storms (Zhao et al., 2006; Uno et al., 267 2009, 2011). They then have a far-reaching influence on climatic and environmental 268 changes both regionally and globally. Until now, there have been a large number of 269 270 intensive field experiments (e.g., ACE-Asia, ADEC, PACDEX, EAST-AIRC) and 271 ground-based aerosol monitoring networks (e.g., AERONET, SKYNET, CARSNET) for probing the Asian mineral dust (Holben et al., 1998; Huebert et al., 2003; 272 Nakajima et al., 2003; Takamura et al., 2004; Eck et al., 2005; Mikami et al., 2006; 273 Huang et al., 2008a; Che et al., 2009, 2015; Li et al., 2011), which is crucial to aid in 274 thoroughly understanding the climatic effects of dust aerosols over East Asian domain. 275 276 Nevertheless, due to poorly sampled over desert source areas of northwest China, the 277 light scattering and absorption properties of mineral dusts in this region are far inadequate and urgently need to be further surveyed. 278

279 The Intergovernmental Panel on Climate Change (IPCC, 2013) reported that the symbol and magnitude of the radiative forcing of mineral dust is greatly reliant on the 280 accurate and reliable knowledge of aerosol total loading, microphysical and chemical 281 characteristics, as well as its spatiotemporal distribution. The current consensus is that 282 283 nearly pure dust aerosol in the globe has relatively low light-absorption, with single-scattering albedo of ~0.96-0.99 (Dubovik et al., 2002; Anderson et al., 2003; 284 285 Uchiyama et al., 2005; Bi et al., 2014, 2016), which principally depends principally on the fraction and mixing ways of ferric iron oxides (i.e., hematite and goethite) in 286 dust (Sokolik and Toon, 1999; Lafon et al., 2004, 2006). However, the coexistences of 287 both mineral dust and other types of aerosols originated from diverse human activities 288 289 (e.g., coal combustion, mobile source emissions, and biomass burning) are ubiquitous 290 in the real atmosphere, which increases the complexity and variability to aerosol key parameters (Arimoto et al., 2004; Xu et al., 2004; Wang et al., 2015). When the lofted 291 dust plumes in desert source areas are traveled eastward across the polluted regions, 292 they commonly mix with anthropogenic pollutants and enhance heterogeneous 293 chemical reactions with other reactive gas species, and then may remarkably alter 294 295 their chemical and microphysical properties (Arimoto et al., 2006; Li and Shao et al., 296 2009; Nie et al., 2014). It is well documented that the mineral dust might have already 297 mixed with polluted aerosols in near dust source regions of northwest China (i.e., Inner Mongolian Gobi desert), besides the mixing processes on the transport pathway 298 (K. Huang et al., 2010). Xu et al. (2004) indicated that both dust aerosol and local 299 300 pollution sources coexisted in Yulin nearby the Mu Us desert of northwest China 301 during April 2001, which produced a significant influence on aerosol properties in the region. Likewise, Li et al. (2010) analyzed trace gases and aerosols observed at 302 Zhangye (39.082°N, 100.276° E, 1460 m above MSL), a rural site within the Hexi 303 Corridor in northwest China during spring 2008, and uncovered that the mixing 304 between mineral dust and anthropogenic air pollutants can be omnipresent in this area, 305 306 including at nighttime or during severe dust events. It implies that prior to moving out 307 from the source region, dust particles were likely in connection with pollutants. For the sparsely populated and lesser anthropogenic affected desert source regions in 308 309 northwest China (e.g., the Taklimakan Desert and its adjacent areas), the interaction between local pollutions and mineral dust is deserved to explore in depth. This is of 310 prime importance to ascertain the relative contributions of two different aerosol 311 sources in atmospheric chemistry and regional climate change. 312

To advance a better understanding of the drought processes and dust-relevant 313 climatic impacts in northwest China (Huang et al., 2008b; Bi et al., 2011; G. Wang et 314 al., 2010), the Semi-Arid Climate and Environment Observatory of Lanzhou 315 University (henceforth referred to as SACOL, http://climate.lzu.edu.cn/english/) 316 carried out a comprehensive field campaign in Dunhuang during spring of 2012. 317 Dunhuang is situated at the westernmost fringe of Hexi Corridor in Gansu province, 318 319 close to the eastern edge of Kumtag Desert and about 450 km in the downwind zone 320 of Taklimakan Desert. It is an important town of the ancient Silk Road and the transportation junction to the ancient western region, central Asia and Europe, which 321 has become a world-famous tourist city with a residential population of 200,000. The 322 agriculture and tourism are the dominant economic industries in Dunhuang. An array 323 of ground-based remote sensing and in situ instruments were set up during the 324 325 intensive period, which sought to investigate the aerosol key properties and its climatic effect on regional scale (Bi et al., 2014). This study especially aims at exploring the light scattering and absorption characteristics of mineral dust and elucidates a potential anthropogenic influence. In the following, we first introduce the site information and integrated measurements in Section 2. The primary results and discussion are described in Sect. 3. The concluding remarks are given in Sect. 4 and followed by the data availability in Sect. 5.

### 332 2. Site and instrumentation

#### 333 2.1. Site information

SACOL's Mobile Facility (SMF) was deployed at Dunhuang farmland (40.492° N, 334 94.955° E, 1061 m above MSL) from 1 April to 12 June 2012. The site is a tiny 335 isolated oasis encompassed by east-west oriented Gobi desert and arid zones in 336 northwest China, with the Mingsha Shan (Echoing-Sand Mountain, elevation: ~1650 337 m) and Sanwei Mountain (elevation: ~1360 m) to the southeast southwest, and the 338 Beishan Mountain (elevation: ~2580 m) to the north (Ma et al., 2013). The underlying 339 340 surface is typically covered with Gobi desert and saline-alkali land, and the principal vegetation types consist of extremely sparse Alhagi. Dunhuang farmland is an 341 important agricultural base in Gobi desert, mainly planting hami melon and cotton. 342 343 There are not any significant manmade pollution sources (e.g., large-scale industries 344 or coal-fired power plants) around the monitoring station. The southwest-northeast oriented National Highway 215 is about 400 m away from the west of the site (Figure 345 346 1a). The nearest Xihu township (with total population of 13,800) is approximately 7 km to the north of Dunhuang farmland, along with some scattered villages stretching 347 from west to east. Meanwhile, the station is located in the northeast of Dunhuang city 348 349 (~45 km), at the west of Guazhou country (~70 km), and at the southwest of Liuyuan 350 town (~80 km). In general, the major anthropogenic emission sources at Dunhuang farmland likely include coal combustion from domestic heating and cooking, mobile 351 sources emissions from vehicle exhaust gas, and biomass burning from crop residue 352 and traditional ritual activities, which are ordinarily considered to be a puny 353 contribution to the mineral dust in present-day climate models. The climate pattern 354

355 here is characterized as extreme drought but with a moderate temperature during the whole sampling period (temperature: 18.3±8.1°C, relative humidity, RH: 21.9±16.5%, 356 mean  $\pm$  standard deviation). Thereby the dust storms frequently take place in this 357 region from spring to early summer. Figure 1(b) shows the overall mean UV aerosol 358 359 index (AI) from 1 April to 12 June 2012 obtained from the Ozone Monitoring 360 Instrument (OMI) absorbing aerosol products (Torres et al., 2007). The AI dataset is a very good indicator for mapping the distribution of absorbing aerosols (mainly black 361 362 carbon and dust). The distributions of High-high AI values (>0.7) distributions are well consistent with the dust-dominated geomorphological features in arid and 363 semiarid regions (i.e., Taklimakan Desert and Gobi deserts). It is very obvious that 364 365 Dunhuang (marked with a pentagram) is also situated at the primary dust belt of 366 northwest China, as presented in Figure 1b.

#### 2.2. Aerosol measurements 367

368 An aerosol integrated observing system is installed in the laboratory of SMF and utilized to continuously measure aerosol optical properties and size distribution in the 369 370 field. Prior to the experiment, the in-situ aerosol instruments and broadband radiometers were newly purchased and calibrated by the manufacturers (Bi et al., 371 2014). Table 1 summarizes the basic specification, measured variables, and accuracy 372 of surface-based instruments deployed at Dunhuang farmland throughout the 373 experiment. We shall describe sequentially as below. 374

An ambient particulate monitor (Model RP1400a, Rupprecht and Patashnick 375 Corp.) can collect the in situ mass concentration of the particulate matter with an 376 aerodynamic diameter less than 10 µm (PM10) based on Tapered Element Oscillating 377 378 Microbalance (TEOM) technique. The measurement range and accuracy of  $PM_{10}$ concentration levels are normally 0-5 gm<sup>-3</sup> and 0.1 µgm<sup>-3</sup>, respectively. The heating 379 temperature ( $\sim 50^{\circ}$ C) of the sampling tube may cause a partial loss of volatile and 380 semivolatile aerosol compounds and hence bring about a negative signal. In this study, 381 we eliminate all the negative values of PM10 concentrations, which account for less 382 than 1% of total data points. 383

An integrating nephelometer (Model 3563, TSI Inc.) is designed to simultaneously 384

measure the total scattering coefficients ( $\sigma_{sp}$ ) and hemispheric backscattering 385 coefficients ( $\sigma_{bsp}$ ) of aerosol particles at three wavelengths of 450, 550, and 700 nm, 386 with the  $\sigma_{sp}$  detection limits of 0.44, 0.17, and 0.26 Mm<sup>-1</sup> (1 Mm<sup>-1</sup>=10<sup>-6</sup> m<sup>-1</sup>), 387 respectively (signal-to-noise ratio of 2) (Anderson et al., 1996). To quantify the 388 389 instrument drift and improve accuracy, we periodically perform the routine calibration 390 using air and high-purity CO<sub>2</sub> gases. Furthermore, the truncation errors of near-forward scattering (i.e., nonideal angular effects) are corrected according to the 391 method of Anderson and Ogren (1998). The observed ambient RH values are mostly 392 393 smaller than 40% throughout the entire period. It is well-documented that RH-induced the variations in aerosol light scattering coefficients are minimized under a low 394 395 sampling stream RH of 10-40% (Covert et al., 1972). In this paper, we computed the 396 scattering Ångström exponent at 450–700 nm (SAE 450/700 nm) from  $\sigma_{sp}$  at 450 nm and  $\sigma_{sp}$  at 700 nm by utilizing a log-linear fitting algorithm. And thus  $\sigma_{sp}$  at 670 nm 397  $(\sigma_{sp,670})$  was logarithmic interpolated between  $\sigma_{sp,450}$  and  $\sigma_{sp,700}$ . 398

A multi-angle absorption photometer (MAAP Model 5012, Thermo Electron 399 400 Corp.) is capable of observing the aerosol light absorption coefficient at 670 nm 401  $(\sigma_{ap,670})$  by filter based methods without requirement of post-measurement data 402 correction or parallel-measured aerosol light-scattering coefficients (Petzold et al., 2002). The instrument detects an emitted light at 670 nm in the forward and back 403 hemisphere of airborne aerosols deposited on a fiber filter, which is used to improve 404 multiple scattering effects in the aerosol optical properties via a radiative transfer 405 scheme (Petzold et al., 2002, 2005). The sample flow rate is 1000 L/h, with flow error 406 of < 1%. We made use of a specific absorption efficiency at 670 nm of  $6.5\pm0.5 \text{ m}^2\text{g}^{-1}$ 407 to estimate black carbon (BC) concentration from  $\sigma_{ap,670}$  as recommended by Petzold 408 et al. (2002). 409

An Aerodynamic Particle Sizer (APS) spectrometer (Model 3321, TSI Inc.) can continuously provide the real-time, high-resolution aerosol size distribution with aerodynamic diameters from 0.5 to 20  $\mu$ m range (52 channels). When extreme dust episodes outbreak, an aerosol diluter (Model 3302A, TSI Inc.) is operated in series with APS to reduce particle concentrations in high-concentration aerosols, which offers a representative sampling that meets the input requirements of the APS
spectrometer. All the mentioned-above aerosol datasets were acquired at 5-minute and
hourly averages, and reported for sampling volumes under standard air conditions (i.e.,
1013.25 hPa and 20 °C).

#### 419 **2.3.** Other ground-based measurements

420 A Micro-Pulse Lidar (Model MPL-4, Sigma Space Corp., U.S.A.) is a compact and unattended apparatus for providing continuous data information of extinction 421 coefficient and depolarization ratio profiles of aerosols and clouds (Welton et al., 422 2000). The MPL-4 emits a laser beam at 527 nm wavelength from a Nd:YLF pulsed 423 laser diode and receives the attenuated backscattering intensity and depolarized 424 425 signals from aerosol particles or cloud droplets with a 30-meter vertical resolution and 426 a 1-minute average interval. And we can acquire the accurate backscattering profile by means of a series of corrections (e.g., dead time, background signal, afterpulse, 427 428 overlap, and range-corrected) according to the standard methods (Campbell et al., 2002). The detailed data acquisition and retrieval algorithms of the lidar system can 429 be referred to the publications of Campbell et al. (2002) and Z. Huang et al. (2010). 430

A weather transmitter (Model WXT–520, Vaisala, Finland) is set up on the top of the SMF trailer and recorded the air temperature (T in  $^{\circ}$ C), relative humidity (RH), ambient pressure (P, unit: hPa), wind speed and wind direction at <u>20–10</u> seconds interval. In this article, we calculated the 5-minute and hourly averages from the raw data.

A dozen of state-of-the-art broadband radiometers were installed in a row on a 436 standard horizontal platform (~4 m above the surface) where the field of view was 437 438 unobstructed in all directions (Bi et al., 2014). The direct normal irradiance and diffuse irradiance were independently measured by an incident pyrheliometer (Model 439 NIP, Eppley Lab.) and by a ventilated and shaded pyranometer (Model PSP, Eppley 440 Lab.), which were mounted on a two-axis automatic sun tracker (Model 2AP, 441 Kipp&Zonen). The global irradiance (0.285-2.8 µm) and downward longwave 442 irradiance (3.5-50 µm) were respectively gathered from a ventilated PSP pyranometer 443 and a ventilated and shaded pyrgeometer (Model PIR, Eppley Lab.). All irradiance 444

quantities were stored in a Campbell data logger with 1-minute resolution.
Additionally, a Total Sky Imager (Model TSI–880, YES Inc.) provides high-resolution
sky pictures every one minute during the daytime, which can detect and identify the
important weather conditions, such as dust storm, smoky pollution, rainy day, cloudy
or cloudless days.

# 450 2.4. MERRA reanalysis products

The MERRA (Modern–Era Retrospective Analysis for Research and Applications) reanalysis assimilates a variety of conventional observations (i.e., temperature, pressure, height, wind components) from surface weather stations, balloons, aircraft, ships, buoys, and satellites from 1980 to the present, which is primarily committed to improve upon the hydrologic cycle and energy budget for the science community (Rienecker et al., 2011). In this paper, we took advantage of the 6-hourly average wind fields at 500 hPa and 850 hPa levels from the MERRA reanalysis products.

## 458 **3. Results and discussion**

#### 459 **3.1 Aerosol optical properties**

The aerosol single-scattering albedo (SSA) at 670 nm is defined as the ratio of the light scattering coefficient ( $\sigma_{sp,670}$ ) to the total extinction coefficient (the sum of  $\sigma_{sp,670}$ and  $\sigma_{ap,670}$ ). The SSA reflects the absorptive ability of aerosol particle and is a key quantity in determining the sign (warming or cooling) of aerosol radiative forcing for a certain underlying surface (Hansen et al., 1997; Ramanathan et al., 2001).

465 Figure 2 delineates the time series of hourly average PM<sub>10</sub> mass concentration, aerosol optical properties and size distribution at Dunhuang farmland during the 466 467 whole period. The overall mean, standard deviation, median, and different percentiles of aerosol optical properties are also tabulated in Table 2. Aerosol optical features 468 469 exhibit dramatic day-to-day variations at Dunhuang. It is apparent that aerosol 470 loadings in April and early May are systematically higher than that in late May and June, which agrees well with the results of columnar aerosol optical depths derived 471 from sky radiometer (Bi et al., 2014). This is chiefly attributed to the invading mineral 472 particulates from the frequent occurrences of intense dust storms in spring season. 473

The highly unstable synoptic cyclones (i.e., Mongolian cyclones) are regularly 474 hovering about the northern China and Mongolia in springtime, which trigger 475 high-frequency strong surface winds (Sun et al., 2001; Shao et al., 2011). The rising 476 temperature in this season leads to the melting of frozen soil and snow cover, leaving 477 478 behind a loose land surface and abundant bare soil sources, therefore affords a 479 favorable condition for dust storms. In addition, the contributions of local dust emissions couldn't be ignored. We have clearly recorded that there were numerous 480 agricultural cultivated operations (e.g., land planning, plowing, and disking) 481 throughout the Dunhuang farmland district from 1 April to 10 May, which produced a 482 great amount of agricultural soil particles under strong winds, and thus had a 483 484 significant superimposed effect on elevated dust loading in the source and downwind regions prior to the growing season. Figure S1 also presents visual photos of a variety 485 of agricultural cultivations in Dunhuang farmland before the planting period, which 486 487 supplies direct and powerful evidences for supporting our results. Those dust aerosols originated from disturbed soils induced by human activities are interpreted as 488 anthropogenic dust (Tegen and Fung, 1995). Recently, some investigators estimated 489 that anthropogenic dust could account for approximately 25% of the global dust load 490 (Ginoux et al., 2012; Huang et al., 2015), and more than 53% of the anthropogenic 491 sources mostly came from semi-arid and semi-wet zones (Huang et al., 2015; Guan et 492 al., 2016). Nonetheless, it still remains a challenging task to distinguish between the 493 natural and anthropogenic fractions of mineral dust by employing a onefold 494 technology, for instance, laboratory analysis, in situ measurements, model simulations, 495 active and passive remote sensing methods (e.g., multichannel lidar, sun/sky 496 497 radiometer), which should be combined together (Bi et al., 2016). The overall mean  $PM_{10}$  concentration was 113±169 µgm<sup>-3</sup> (mean ± standard deviation), which is ~39% 498 lower than the 184.1±212 µgm<sup>-3</sup> average level in Dunhuang (40.1° N, 94.6° E, 1139 499 m) during the spring of 2004 (Yan, 2007), and ~26% smaller than the value of 500 153±230 μgm<sup>-3</sup> measured at Zhangye (39.082° N, 100.276° E, 1460 m) during spring 501 of 2008 (Li et al., 2010). Wang et al. (2015) obtained a total average PM<sub>10</sub> 502 concentration of 172±180 µgm<sup>-3</sup> at SACOL during late spring of 2007 (from 25 April 503

to 25 June). And the mean  $PM_{10}$  levels at Hunshan Dake sandland in northern China during spring of 2001 varied between 226 and 522  $\mu$ gm<sup>-3</sup> (Cheng et al., 2005).

The hourly average aerosol light scattering coefficient at 670 nm ( $\sigma_{sp.670}$ ) was 506 53.3 $\pm$ 74.8 Mm<sup>-1</sup>. The big standard deviations of PM<sub>10</sub> and  $\sigma_{sp}$  are possibly associated 507 508 with the injection of dust particles during the intense dust storms. Our result was about a factor of 3 lower than the  $\sigma_{sp}$  at 500 nm in mentioned-above other sites over 509 northern China (i.e., 126±90 Mm<sup>-1</sup> for Dunhuang, 159±191 Mm<sup>-1</sup> for Zhangye, 510 164±89 Mm<sup>-1</sup> for SACOL). Despite relatively small magnitude, the aerosol light 511 absorption coefficient at 670 nm ( $\sigma_{ap,670}$ ) also presented pronounced variations, with 512 an average value and a maximum of 3.2±2.4 Mm<sup>-1</sup> and 25.0 Mm<sup>-1</sup>, respectively. This 513 result was a factor of 2 smaller than Yulin (6±11 Mm<sup>-1</sup>) nearby Mu Us desert in 514 northwest China (Xu et al., 2004), and a factor of 5~7 far less than that at Shangdianzi 515 rural site (17.5±13.4 Mm<sup>-1</sup>) in northern China (Yan et al., 2008) and Lin'an site (~23 516 Mm<sup>-1</sup>) in southern China (Xu et al., 2002). The mean light scattering and absorption 517 coefficients in this study are comparable to the background levels (~46.9±16.9 and 518 2.5±1.1 Mm<sup>-1</sup>) in Southern Great Plain of U.S.A (Delene and Ogren, 2002). This 519 suggests that extremely low levels of light absorption and scattering substances are 520 widely distributed throughout the Dunhuang region during the spring of 2012. 521 Therefore, a little perturbation stemmed from human activities (e.g., agricultural 522 cultivation, coal combustion from domestic heating and cooking, and biomass burning) 523 would undoubtedly exert a considerable impact on the light absorption property. 524

A few of strong dust episodes (4, 21-22, and 30 April, 1-3, 8-11, and 20 May, 4 525 and 10 June, corresponding to DOY 95, 112-113, 121, 122-125, 129-132, 141, 156, 526 and 162) could remarkably elevate the hourly average values of  $PM_{10}$ ,  $\sigma_{sp}$ ,  $\sigma_{ap}$ , and 527 aerosol size distribution (see Figure 2). During these dust events, the hourly  $PM_{10}$ 528 concentrations generally exceeded 1000 µgm<sup>-3</sup> and even approached 2000 µgm<sup>-3</sup>, 529 which were tenfold greater than the overall mean level. The hourly  $\sigma_{sp}$  were more 530 than 400 Mm<sup>-1</sup> or even close to 800 Mm<sup>-1</sup>, and the corresponding  $\sigma_{ap}$  varied between 531 10 Mm<sup>-1</sup> and 25 Mm<sup>-1</sup>. Moreover, the peak values of aerosol number size distribution 532 concentrated in the particle diameters of 1-3 µm, which was consonant with the result 533

from remote sensing (Bi et al., 2014, 2016).

Figure 3 depicts the time evolutions of MPL normalized relative backscatter and 535 depolarization ratio at Dunhuang farmland from 1 April to 12 June 2012. The 536 537 depolarization ratio ( $\delta$ ) is a useful indication to discriminate between spherical particles ( $\delta$  of ~0–0.1) and nonspherical particles (mainly dust aerosol,  $\delta$  >0.2), since 538 539 it is very sensitive to the nonsphericity of scattering particle (Kobayashi et al., 1985; Murayama et al., 1999; Shimizu et al., 2004; Huang et al., 2015). From Figure 3, we 540 can distinctly see that there was always a dense dust layer appeared at a height below 541 542 2 to 4 km during the whole experiment, with the peak value centered on 1.0–1.5 km, which was within above the planetary boundary layer (PBL). And the  $\delta$  values 543 544 commonly reached above 0.3 (>  $\sim$ 0.3–0.5) during the heavy dust events and varied between 0 and 0.1 under clear-sky conditions (e.g., 6-7 April, 14-15 and 29 May, 9 545 June). 546

#### 547 3.2 Diurnal variations

Figure 4 illustrates the diurnal variations of wind vector (ms<sup>-1</sup>), air temperature (T 548 in °C), RH (%), PM<sub>10</sub> ( $\mu$ gm<sup>-3</sup>),  $\sigma_{sp,670}$  (Mm<sup>-1</sup>),  $\sigma_{ap,670}$  (Mm<sup>-1</sup>), aerosol number size 549 distribution (dN/dlogD in cm<sup>-3</sup>), SAE at 450-700 nm, and SSA at 670 nm in 550 Dunhuang farmland from 1 April to 12 June 2012. Note that the APS spectrometer 551 was operated from 30 May to 12 June. A discernible wind vector was showed shown 552 in the diurnal variation, in other words, strong southwest wind and south wind 553 dominated in the daytime, from 11:00 to 24:00 LT (local time), and transformed into 554 the weak northeast wind prevailed from the midnight to the following morning of 555 10:00 LT. The prominent phenomenon can be roughly interpreted by classical 556 557 mountain-valley wind circulation, which was primarily generated by the diurnal differences of temperature between the mountain slope and the valley floor. During 558 the daytime, the huge Beishan Mountain slope heats up by the solar radiation more 559 rapidly than the valley floor, which causes convection above the mountain slope. The 560 compensating airflow is consequently directed toward the mountain slope, inducing 561 upslope southerly wind, or the valley wind, which usually peaks near midday and 562 gradually disappears after sunset. Conversely, at night, radiative cooling of the 563

564 mountain slope is more quickly than the valley floor, inducing the mountain wind, which generally reaches maximum strength just before sunrise (Arya, 1999). 565 Throughout the experiment, air temperature displayed a large diurnal variation (with 566 the diurnal difference of  $\delta T \sim 26$  °C) and RH always kept below 40% for the whole day. 567 568 It is very clear that the minimal T and maximal RH arose at around 06:00-07:00 LT, 569 and the maximal T and minimal RH occurred at about 16:00 LT, which represented an energetic vertical turbulent motion in daytime and a stable radiative temperature 570 inversion during nighttime. 571

The aerosol optical parameters also exhibited striking diurnal variations, which 572 were closely related to the local meteorological elements. During the daytime 573 (10:00–18:00 LT), the PM<sub>10</sub> concentration remained high level (~57–65  $\mu$ gm<sup>-3</sup>) and 574 increased sharply from 19:00 LT and reached a maximum of 84.2 µgm<sup>-3</sup> at 20:00 LT. 575 The PM<sub>10</sub> began to decrease from 21:00 LT to the next morning. A low level (~40-46 576 µgm<sup>-3</sup>) kept in the midnight (00:00–05:00 LT) and rose gradually from 06:00 LT and 577 attained a secondary peak value of 55.7 µgm<sup>-3</sup> at 07:00 LT. The aerosol light 578 scattering ( $\sigma_{sp.670}$ ) presented a similar pattern with PM<sub>10</sub>, but the maximal value (~42 579 Mm<sup>-1</sup>) appeared at 13:00 LT, with the other two secondary peak values occurred at 580 20:00 (~34.1 Mm<sup>-1</sup>) and 07:00 LT (~27.3 Mm<sup>-1</sup>). The high levels of PM<sub>10</sub> and  $\sigma_{sp}$ 581 during the daytime were primarily attributable to strong south wind from Gobi region 582 and local dust emissions. By contrast, aerosol light absorption coefficient ( $\sigma_{ap,670}$ ) 583 showed a more pronounced diurnal feature, which was well proved to be majorly 584 controlled by anthropogenic emissions (Li et al., 2010). The diurnal  $\sigma_{ap}$  always stayed 585 at a low level (~2.0 Mm<sup>-1</sup>) from 13:00-18:00 LT, and also reached a maximum of 3.3 586 Mm<sup>-1</sup> at 20:00 LT. Subsequently,  $\sigma_{ap}$  dramatically reduced from midnight and 587 preserved at a low value of about 2.2 Mm<sup>-1</sup> from 02:00-04:00 LT, and remained a 588 steadily high level of ~2.7-2.9 Mm<sup>-1</sup> from 05:00-10:00 LT. It was probably explained 589 as follows. The influences of local anthropogenic pollutants were commonly small in 590 the afternoon, because the strong southerly wind from Gobi deserts and powerful 591 daytime vertical convection mixing efficiently dilute local air pollutants. Whereas 592 weak northeast wind and stable temperature inversion at night facilitate the 593

accumulation of pollutants within the PBL, hence nighttime levels were normally 594 larger. Increasing human activities (e.g., domestic cooking, traffic emissions for 595 transportation and agriculture) in the early morning might also be responsible for the 596 597 morning peaks in the aerosol absorption coefficient. The  $\sigma_{ap}$  maximum at 20:00 LT 598 was presumably influenced by the mixture of mineral dust and anthropogenic 599 pollutants. This conclusion could be partly supported by the diurnal variation of SAE at 450-700 nm (Figure 4), which showed high SAE values (~0.5-0.6) appeared at 600 02:00-10:00 LT and low SAE (~0.2-0.3) occurred on 13:00-22:00 LT. Generally, 601 large SAE around 0.6 represents small particles (e.g., urban-polluted aerosol or soot) 602 and low SAE less than 0.3 or negative value corresponds to coarse-dominated large 603 604 size particles (e.g., dust or sea salt) (Anderson et al., 2003).

605 Furthermore, aerosol number size distribution exhibited a noticeable supermicron particles dominated in the entire day, probably linked to the predominant dust aerosol 606 607 in daytime and local anthropogenic emissions at nighttime. In this study, we 608 postulated that the aerosol light extinction at shortwave waveband is completely caused by those particles with aerodynamic diameters of 10 µm or less. And the mass 609 scattering efficiency is designated as the ratio of  $\sigma_{sp}$  to PM<sub>10</sub> concentration. Therefore, 610 the mass scattering efficiency for  $PM_{10}$  aerosols was about 0.67  $m^2g^{\text{-1}}$  in the afternoon 611 and ~ $0.77 \text{ m}^2\text{g}^{-1}$  in the morning (~0.25 for heavy dust events, and ~0.70 for the whole 612 period). Our results were slightly less than ~1.05 m<sup>2</sup>g<sup>-1</sup> in Dunhuang during spring of 613 2004 (Yan, 2007). SimilarlyLikewise, the mass absorption efficiency was ~0.017 614  $m^2g^{-1}$  under heavy dust episodes and ~0.08  $m^2g^{-1}$  in the morning, which was 615 coincident with the laboratory analytical result of natural desert aerosol at 660 nm 616 (~0.01–0.02 m<sup>2</sup>g<sup>-1</sup>) in Ulan Buh desert (39°26'N, 105°40'E) of northern China (Alfaro 617 et al., 2004). These diurnal variations of the mass scattering and absorption 618 efficiencies likely reflect the changes in aerosol chemical composition. The SSA at 619 670 nm displayed distinct differences between daytime and nighttime (Figure 4), and 620 the two minimal values at 07:00 LT (~0.90) and 20:00 LT (~0.921) were consistent 621 with the aforementioned  $\sigma_{ap,670}$  diurnal feature. The peak values of SSA (0.945±0.04) 622 for dominant dust particles in the afternoon agreed well with other field campaigns in 623

Zhangye (0.95±0.02, Li et al., 2010) and Yulin (0.95±0.04, Xu et al., 2004). The daily 624 low SSA (0.90–0.92) or overall mean of 0.913±0.055 at Dunhuang was still bigger 625 than that in both urban (0.81, Bergin et al., 2001) and rural regions (0.81–0.85, Li et 626 al., 2007) adjacent to Beijing, presumably ascribed to dust particles at night. Yan et al. 627 628 (2008) conducted two-year long field measurements at Shangdianzi Global 629 Atmosphere Watch (GAW) rural site in northern China (~150 km from Beijing) and estimated a mean SSA of 0.88±0.05, but their data contained summer when aerosol 630 scattering coefficients may be strengthened by hygroscopic growth and secondary 631 632 chemical process.

The wind rose plots give a further insight into the linkages between the 633 634 meteorological factors and pollutants, as described in Figure 5. In the morning (06:00-09:00 LT), a marked northeast wind was prevalent and wind speed was mostly 635 less than 4 ms<sup>-1</sup>, which revealed that emissions were primarily descended from nearby 636 637 farmlands and rural residences (Figure 5a). Although a prominent northwest wind mainly occurred in the evening hours (19:00-22:00 LT), the east wind and southwest 638 wind also appeared, which indicated that anthropogenic pollutions came from both 639 local sources and a relatively large region along the valley (Figure 5b). And Figure 5c 640 showed the predominant winds were northeast and southwest winds in Dunhuang area, 641 with the maximal hourly-averaged wind speed exceeding 10 ms<sup>-1</sup>. It was very distinct 642 that the southwest and northwest winds created higher levels of PM10 mass 643 concentration (>250  $\mu$ gm<sup>-3</sup>), aerosol light scattering coefficient ( $\sigma_{sp}$  >150 Mm<sup>-1</sup>) and 644 absorption coefficient ( $\sigma_{ap} > 8 \text{ Mm}^{-1}$ ), whereas northeast wind generated slightly 645 smaller concentrations of PM<sub>10</sub> (~50–100  $\mu$ gm<sup>-3</sup>),  $\sigma_{sp}$  (~30–60 Mm<sup>-1</sup>) and  $\sigma_{ap}$  (~2–4 646 Mm<sup>-1</sup>). This was possibly implied that southwest and northwest winds may bring 647 about dust particles and northeast wind may transport the air pollutants. 648

## 649 **3.3 Local anthropogenic emission sources**

As mentioned above, crop residue burning and agricultural cultivated operations before the growing season could produce local emission source proximity to the study area. And sporadic straw burning was indeed to happen throughout the Dunhuang farmland from 1 April to 10 May 2012, which was the major source of black carbon surrounding the site. To clarify the potential anthropogenic influence on aerosoloptical properties in desert region, we investigated a typical biomass burning event.

Figure 6 outlines the time series of 5-minute average wind vector ( $ms^{-1}$ ),  $PM_{10}$ 656 ( $\mu$ gm<sup>-3</sup>),  $\sigma$ <sub>sp</sub> at 450, 550, and 700 nm (Mm<sup>-1</sup>), SAE (450–550, 550–700, and 450–700 657 nm),  $\sigma_{ap,670}$  (Mm<sup>-1</sup>), and SSA at 670 nm during a typical Tomb-sweeping Day on 4 658 April 2012. Tomb-sweeping Day is a Chinese traditional festival for sacrifice rites, in 659 commemoration of the dead ancestors. To pay homage to loved ones, the people 660 burned a lot of joss sticks, candles, and paper offerings, and set off firecrackers in that 661 day throughout the China, which would emit a great amount of air pollutants, such as, 662 biomass burning aerosol, sulfur dioxide, organic matter, and fugitive dust. From 663 Figure 6a, slight and variable winds (with wind speed <4 ms<sup>-1</sup>) mainly came from 664 northeasterly from 00:00 to 12:00 LT, and abruptly changed into weak southeast wind 665 and south wind, finally, gradually intensified southwest wind (>10 ms<sup>-1</sup>) were 666 predominant and triggered a severe dust storm from 15:00 LT to the midnight. Prior to 667 the occurrence of dust episode, the aerosol optical characteristics varied stably, but a 668 moderate increase was evident during 08:00 to 10:00 LT. For instance, PM<sub>10</sub> 669 concentration gradually increased from background level ~30 µgm<sup>-3</sup> to a maximum of 670 62.5  $\mu$ gm<sup>-3</sup> at about 09:00 LT,  $\sigma_{sp,550}$  from ~15 Mm<sup>-1</sup> to 49.6 Mm<sup>-1</sup>, and  $\sigma_{ap,670}$  from 671  $\sim 2.0 \text{ Mm}^{-1}$  to 4.75 Mm<sup>-1</sup>. It is ascribed to the contribution of biomass burning in the 672 process of ritual activities during Tomb-sweeping Day. The SAE value at 450-700 nm 673 remained invariant ( $\sim 0.50$ ) before 08:00 LT and sharply rose to a maximal value of 674 0.87 at 09:00 LT, afterwards gently reduced to around 0.4, which indicated that the 675 fine-mode particles (i.e., black carbon or soot) were dominated from 08:00 to 10:00 676 677 LT. And the SAEs at various wavelengths systematically decreased from 0.4 at 15:00 678 LT to -0.25 at midnight, suggesting the dust-dominant coarse-mode particles were prevailed. Meanwhile, the lidar depolarization ratio ( $\delta$ ) also further verified the 679 existence of small size soot particle. The  $\delta$  value preserved steadily at 0.15–0.20 680 during 08:00 to 10:00 LT, and rapidly attained above 0.3 from 15:00 LT and even 681 approached 0.50 at intense dust storm (see Fig. 3). The diurnal variation of SSA<sub>670</sub> 682 683 showed a more prominent feature, as illustrated in Figure 6f. The SSA<sub>670</sub> values kept 684 between 0.88 and 0.92 during 00:00 to 07:00 LT, and dramatically reduced to a minimum of ~0.83 at 08:30–09:00 LT, then rose to 0.925, confirming the very striking 685 impacts by light absorbing substances. After 15:00 LT, the SSA<sub>670</sub> gradually increased 686 and reached up to about 0.96 during dust storms occurred. Bi et al. (2014) have 687 688 demonstrated that dust aerosols shortwave radiative forcing (ARF) at the top of the atmosphere (TOA) was warming effect when SSA<sub>500</sub> was less than 0.85, but was 689 cooling effect when SSA<sub>500</sub> was greater than 0.85 for Dunhuang Gobi desert area with 690 high surface albedo. Thereby such significant anthropogenic influence would clearly 691 modify the microphysical and chemical properties of dust aerosols and eventually 692 exert remarkable impacts on environmental quality and climatic forcing of dust 693 694 particle on both local and regional scales.

#### 695 **3.4 Dust cases study**

In this section, we particularly explored the absorptive and optical characteristics 696 697 of mineral dust during several typical dust cases and discussed its influence on Earth's radiation balance. Figure 7 provides the wind fields at 500 hPa and 850 hPa levels 698 during three heavy dust events, based on MERRA reanalysis products. Note that 699 Dunhuang farmland is marked with a red pentagram and the white areas at 850 hPa 700 represent the missing values. It is evident that East Asian region was governed by the 701 powerful and stable westerlies at 500 hPa height on 30 April and 1 May 2012, 702 703 whereas two very strong synoptic cyclones at 500 hPa upper atmosphere hovered 704 about the Mongolia and Kazakhstan respectively on 10 June 2012, matching up with corresponding cyclone systems appeared at the 850 hPa level. Although there were 705 missing data in most northwest China, extremely intense northeast wind and east wind 706 (> 10 ms<sup>-1</sup>) at 850 hPa level were prevailed over the northern territory of Xinjiang 707 708 Uygur Autonomous Region during the selected dust storms, where was close to the Dunhuang site. This could be well confirmed by the simultaneous observations of 709 wind speed and wind direction nearby the surface at Dunhuang farmland, as 710 delineated in Figure 8(a). The measured strong northeast and east winds were always 711 dominated in Dunhuang and 5-min average wind speed attained above 10 ms<sup>-1</sup> during 712 intense dust episodes. The selected three dust processes regularly lasted for several 713 24

hours during daytime (e.g., from 10:00 to 18:00 LT) and the dust event on 1 April
could be persistent to the midnight, which contributed massive dust particles into the
atmosphere.

There were no measurements of aerosol scattering coefficient ( $\sigma_{sp}$ ) on 10 June due 717 718 to equipment failure. From Figure 8, we could know that PM<sub>10</sub> concentrations usually exceeded 400 µgm<sup>-3</sup> and even reached up to 1000 µgm<sup>-3</sup> during the heavy dust storms, 719 and corresponding  $\sigma_{sp,550}$  and  $\sigma_{ap,670}$  values were generally more than 100 Mm<sup>-1</sup> and 5 720 Mm<sup>-1</sup>, respectively, or approached 350 Mm<sup>-1</sup> and 15 Mm<sup>-1</sup> in our cases. It is worthy 721 note that even though pure dust aerosol possesses relatively low light-absorption 722 ability (with mass absorption efficiency at 660 nm of  $\sim 0.01-0.02 \text{ m}^2\text{g}^{-1}$ ), the injection 723 of plentiful mineral particles from dust episodes led to considerably high values of 724 725  $\sigma_{ap,670}$ . And the SAEs at diverse wavelengths commonly kept at 0.50 or more during non-dust conditions, while corresponding values dramatically reduced to -0.25~0 726 727 under heavy dust cases, which is taken for granted. The SSA<sub>670</sub> also exhibited apparent diurnal variations in Figure 8(f). The SSA<sub>670</sub> values regularly preserved 728 between 0.88 and 0.92 at nighttime or non-dust weather, and gradually increased to a 729 maximum of ~0.96-0.98 during strong dust processes, which were close to the 730 measured value of ~0.97-0.99 for nearly pure Asian dust particles (Anderson et al., 731 2003; Bi et al., 2016). These abundant mineral particles in desert source regions were 732 very likely mixed with local air pollutants especially at night, when the anthropogenic 733 734 pollutions favorably built up within the PBL. Moreover, airborne dust particles ordinarily traveled long distances to downstream areas via mesoscale synoptic 735 cyclones, which would deteriorate the ambient air quality and affected atmospheric 736 737 chemistry and climate change on regional scale.

Figure 9 describes the column-integrated aerosol optical depth (AOD) at five wavelengths (400, 500, 675, 870, and 1018 nm) versus Ångström exponent ( $\alpha$ ) at 400–870 nm on two completely clear-sky days (14 May and 9 June) and two typical dusty days (30 April and 10 June), which were acquired from sky radiometer (Model POM-01, PREDE Co. Ltd.). The sky radiometer can measure the direct solar irradiances and sky diffuse radiances at narrow spectral wavebands during daytime 744 with 10-minute interval. And the columnar aerosol optical properties under cloudless conditions were retrieved from sophisticated inversion algorithms (Nakajima et al., 745 1996). Note that the cloud contaminated datasets have been eliminated by means of a 746 series of cloud screening procedures developed by Khatri and Takamura (2009). From 747 748 Figure 9, all AOD values under clear-sky days kept very stable variations throughout the day and ranged from 0.02 to 0.12, which were comparable to the clean 749 background levels in the central Tibetan Plateau (Xia et al., 2011) and Badain Jaran 750 Desert (Bi et al., 2013). And the corresponding Ångström exponent  $\alpha$  on 14 May and 751 9 June were greater than 0.6, indicating extremely low aerosol loading. In contrast, 752 the AODs under dust events (30 April and 10 June) displayed pronounced diurnal 753 754 variations and all AOD values were larger than 0.30 (with maximum of 0.60), and  $\alpha$ varied between 0.10 and 0.25, representing high dust concentration levels. These 755 elevated dust particles in the atmosphere would readjust the energy distributions of 756 757 solar radiative fluxes at the surface.

Based on aforementioned measurements of total sky imager, micro-pulse lidar and 758 sky radiometer, we identified three completely clear-sky days (14 May, 29 May, and 9 759 June) and two "clean" dusty days (30 April and 10 June). The "clean" dusty days in 760 this study were denoted as the dust storms weather without the influence of clouds. 761 This afforded us a good opportunity to elucidate the potential impacts of dust events 762 on radiation balance at the ground. Figure 10 draws the 1-minute average solar direct 763 764 normal radiation, sky diffuse radiation, total shortwave radiation, and downward long-wave radiation fluxes under the selected five days, which were derived from the 765 high-precision broadband radiometers as described in section 2.3. All radiative 766 767 quantities presented smooth diurnal variations under clear-sky cases (14 May, 29 May, 768 and 9 June). The airborne dust particles impeded the sunlight to the ground through scattering and absorbing solar radiation, for instance, they could significantly reduced 769 the surface direct radiative fluxes in daytime about 200-350 Wm<sup>-2</sup> (Figure 10a), 770 whereas considerably increased the surface diffuse radiative fluxes up to ~150-300 771 Wm<sup>-2</sup> (Figure 10b). As a result, the overall attenuation effect on total shortwave 772 radiative fluxes varied between -150 and -50 Wm<sup>-2</sup>. The incoming solar energy 773 26

774 absorbed by dust particles would heat the atmospheric dust layer (Bi et al., 2014) that 775 likely played a profound role in the structure of atmospheric boundary layer structure and cloud microphysical process (J. Huang et al., 2006, 2010; Li et al., 2016). 776 The downward longwave radiation (DLW) at the surface was majorly reliant on the 777 778 clouds, water vapor, CO<sub>2</sub>, and other greenhouse gases (Wang and Dickinson, 2013). 779 In general, the presence of clouds in the atmosphere would fluctuate drastically the diurnal variation of DLW. And the smooth changes of DLW under both clear-sky and 780 dusty days in Figure 10d revealed the robustness of the cloud screening method used 781 in this paper. Figure 10d displays that the DLW values under dusty cases were always 782 greater than that in clear-sky cases, with the total average differences of  $+40 \sim +60$ 783 Wm<sup>-2</sup>. The warming dust layer could enhance the surface DLW, hence the dust 784 particles should contribute a few-large percentages to the increased DLW, but not all. 785 This is because the potential greenhouse gaseswater vapor in the atmosphere could 786 787 substantially affect the DLW variations. For instance, the DLW on 9 June were distinctly lager larger than that in other cloudless cases (i.e., 14 and 29 May) and the 788 dusty case of 30 April. It is partly attributable to the higher RH values on 9 June than 789 that in other days, as shown in Figure S2. 790

# 791 **4. Concluding remarks**

792 In this article, we surveyed the optical features and size distribution of dust aerosol in a Gobi farmland region of northwest China from 1 April to 12 June 2012, 793 794 and uncovered a potential anthropogenic influence. The overall average  $PM_{10}$  mass concentration, light scattering coefficient ( $\sigma_{sp,670}$ ), absorption coefficient ( $\sigma_{ap,670}$ ), and 795 single-scattering albedo (SSA<sub>670</sub>) throughout the experiment were  $113\pm169 \ \mu gm^{-3}$ , 796  $53.3\pm74.8$  Mm<sup>-1</sup>,  $3.2\pm2.4$  Mm<sup>-1</sup>, and  $0.913\pm0.05$ , which were comparable to the 797 background levels in southern United States, but lower than that in the eastern and 798 other northwestern China. Frequent dust storms could markedly elevate dust loading 799 and dominated the temporal evolution of airborne aerosol in Dunhuang region. The 800 hourly average PM\_{10},  $\sigma_{sp,670},$  and  $\sigma_{ap,670}$  reached up to 2000  $\mu gm^{\text{-3}},$  800 Mm^{\text{-1}}, and 25 801 Mm<sup>-1</sup> during the severe dust events that were tenfold greater than the total mean 802

values, along with the particle size concentrated in diameters of 1–3  $\mu$ m. Meanwhile, the correspondingly high SSA<sub>670</sub> (~0.96–0.98) and depolarization ratio ( $\delta$  of ~0.3–0.5), and low SAE (-0.25~0) values adequately verified the presence of coarse-mode mineral dust, resulting in significantly reducing the solar direct radiation (~200–350 Wm<sup>-2</sup>) and increasing diffuse radiation (~150–300 Wm<sup>-2</sup>) at the surface, and hence affecting the regional climate.

Owing to relatively low aerosol levels observed in Dunhuang, any slightly 809 anthropogenic perturbation would induce a substantial influence on the aerosol 810 physicochemical property. The so-called anthropogenic dust produced by agricultural 811 cultivating operations (e.g., land planning, plowing, and disking) brought a significant 812 813 superimposed effect on high dust concentrations in Dunhuang farmland prior to the 814 growing season, when the underlying surface was primarily covered with bare soils. This to some extent could be interpreted the drastic changes of aerosol loadings in 815 816 April and early May. In contrast, the local pollutant emissions mainly affected the absorptive characteristics of dust aerosol especially at night, when the anthropogenic 817 pollutants favorably accumulated within the PBL and likely mixed with abundant 818 mineral dust in the atmosphere. Therefore, the diurnal variations of  $\sigma_{ap,670}$  and SSA<sub>670</sub> 819 exhibited prominent features, both of which have got two peak values at night and in 820 the early morning. For instance,  $\sim 3.3 \text{ Mm}^{-1}$  at 20:00 LT and  $\sim 2.9 \text{ Mm}^{-1}$  at 08:00 LT 821 for  $\sigma_{ap.670}$  were much more than the low level of ~2.0 Mm<sup>-1</sup> in the afternoon, which 822 was attributed to the influence of anthropogenic emissions. And the mean SSA<sub>670</sub> of 823 predominant dust particles in the afternoon (13:00-18:00 LT) was 0.945±0.04 that 824 was evidently greater than the mixed dust-pollutants dominated SSA<sub>670</sub> of ~0.90 at 825 826 07:00 LT and ~0.92 at 20:00 LT.

The findings of this study directly demonstrated <u>that</u> mineral dust in Dunhuang farmland was substantially affected by anthropogenic pollutants, which would help to promote a further insight into the interaction among dust aerosol, atmospheric chemistry, and regional climate in desert source region. However, the potentially anthropogenic influences on dust aerosol in Dunhuang showed far smaller than that measured in eastern China, which was expected for the remote desert areas with sparsely population and lesser human activities. Recently, Huang et al. (2016) indicated that most of the drylands in the world were fragile and susceptible to climate change and human activities and would be subject to the acceleration of drought expansion by the end of twenty-first century. Under the possible scenario, it is very critical to make clear the relative contributions of natural and anthropogenic forcing factors on global climate change, such as, natural dust and anthropogenic dust, which calls for further investigating through a lot more observations and technologies.

#### **5. Data availability**

All ground-based aerosol datasets used in this paper are available via contactingJianrong Bi (bijr@lzu.edu.cn).

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1171	Table 1. The main	n aerosol observing and other ground-based instruments deployed	ed at Dunhuang	
1172	farmland during sp	pring of 2012.		
Measured variables		Model, Manufacturer	Accuracy	
PM <sub>10</sub> concentration		Ambient particulate monitor (RP1400a), R&P Corp.	0.1 μgm <sup>-3</sup>	
Aerosol scattering coefficient		Integrating nephelometer (TSI 3563), TSI Inc. 450, 550, and 700 nm	0.44, 0.17, and 0.26 Mm <sup>-1</sup>	
Aerosol absorption coefficient		Multi-angle absorption photometer (MAAP 5012), Thermo	0.66 Mm <sup>-1</sup>	
Aerosol size distribution		Aerodynamic particle sizer (APS 3321), TSI Inc., 0.5~20 µm	0.001 cm <sup>-3</sup>	
Aerosol-attenuated backscatter profile		Micro-pulse lidar (MPL-4), Sigma Space Corp.	Spatial resolution: ~30 m	

A m Ta: ±0.3°C, RH: 0.1%, P: Weather transmitter (WXT-520), Vaisala, Ta, RH, P, u, WD Meteorological elements 0.1 hPa, u: 0.1 ms<sup>-1</sup>, WD:1° Global: 8.46, diffuse: 8.48 Pyranometer (PSP<sup>a,b</sup>), Eppley Lab., 0.285~2.8 μm Global and diffuse radiation  $\mu VW^{-1}m^{-2}$ Pyrheliometer (NIP<sup>b</sup>), Eppley Lab., 0.285~2.8 μm  $8.38 \ \mu V W^{-1} m^{-2}$ Direct radiation Downward long wave radiation Pyrgeometer (PIR<sup>a,b</sup>), Eppley Lab., 3.5~50 μm  $2.98 \ \mu V W^{-1} m^{-2}$ Total Sky Imager (TSI880), YES Inc., 352×288 pixel 24-bit color JPEG image Sampling rate: 1 minute 1173 <sup>a</sup>The instrument is equipped with the Eppley ventilation system (VEN).

<sup>b</sup>The instrument is mounted on a two-axis automatic sun tracker (Model 2AP, Kipp&Zonen).
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**Table 2.** Statistical summary of hourly average aerosol optical properties measured during
 intensive observation period<sup>a</sup>

Variable	Mean	Std <sup>b</sup>	Median	10 <sup>th</sup> percentile	25 <sup>th</sup> percentile	75 <sup>th</sup> percentile	90 <sup>th</sup> percentile
PM <sub>10</sub> (µgm <sup>-3</sup> )	113	169	54	17	29	111	300
$\sigma_{sp} (Mm^{-1})$	53.3	74.8	28.3	11.2	16.0	55.8	123.5
$\sigma_{ap} (Mm^{-1})$	3.20	2.40	2.50	1.27	1.69	3.90	5.94
SSA (670 nm)	0.913	0.055	0.923	0.850	0.892	0.949	0.967
SAE (450/700 nm)	0.45	0.45	0.42	-0.1	0.1	0.73	0.99

<sup>1176</sup> 



 $^aAll$  aerosol data reported for volumes under 1013.25 hPa and 20  $^\circ\!\!\mathbb{C}.$ 



(b)

45N

35N

25N

- 1199 1200
- 1201
- 1202
- 1203



**Figure 2.** Time series of hourly average (a)  $PM_{10}$  mass concentration in  $\mu gm^{-3}$ , (b) aerosol absorption coefficient at 670 nm, (c) aerosol scattering coefficient at 670 nm, and (d) aerosol size distribution in cm<sup>-3</sup> at Dunhuang farmland during the whole sampling period.



MPL Normalized Relative Backscatter



Figure 4. The diurnal variations of (first row, left to right) wind vector (ms<sup>-1</sup>), air temperature (T 1232 in °C), relative humidity (RH in %), (second row, left to right) PM<sub>10</sub> concentration (µgm<sup>-3</sup>), 1233 aerosol scattering coefficient at 670 nm ( $\sigma_{sp,670}$  in Mm<sup>-1</sup>), aerosol absorption coefficient at 670 nm 1234  $(\sigma_{ap,670} \text{ in Mm}^{-1})$ , (third row, left to right) scattering Ångström exponent at 450–700 nm (SAE 1235



450/700 nm), aerosol single-scattering albedo at 670 nm (SSA<sub>670</sub>), and aerosol size distribution



**Figure 5.** Wind rose plots for (a) morning hour (06:00–09:00 LT), (b) evening hour (19:00–22:00 LT), and (c) all hours; shade represents wind speed (ms<sup>-1</sup>). Wind roses for all hours, with shade representing levels of (d) PM<sub>10</sub> concentration ( $\mu$ gm<sup>-3</sup>), (e) aerosol scattering coefficient at 670 nm ( $\sigma_{sp}$  in Mm<sup>-1</sup>), and (f) aerosol absorption coefficient at 670 nm ( $\sigma_{ap}$  in Mm<sup>-1</sup>).



**Figure 6.** Time series of (a) wind vector (ms<sup>-1</sup>), (b)  $PM_{10}$  concentration ( $\mu$ gm<sup>-3</sup>), (c) aerosol scattering coefficient ( $\sigma_{sp}$  in Mm<sup>-1</sup>) at 450 nm (red), 550 nm (green), and 700 nm (blue), (d) scattering Ångström exponent (SAE) at 450–550 nm (red), 550–700 nm (green), and 450–700 nm (blue), (e) aerosol absorption coefficient at 670 nm ( $\sigma_{ap}$  in Mm<sup>-1</sup>), and (f) single-albedo albedo at 670 nm (SSA<sub>670</sub>) during a typical Tomb-sweeping Day on 4 April 2012, which implies a potential anthropogenic influence on aerosol optical properties. All data points are obtained from 5-minute average values.



Figure 7. The wind fields (black arrows) at 500 hPa (left panel) and 850 hPa (right panel) levels
during three heavy dust events on 30 April (top), 1 May (middle), and 10 June (bottom) 2012,
based on MERRA reanalysis data. Note that the Dunhuang farmland is marked with a red
pentagram and the white regions at 850 hPa are on behalf of the missing values.





**Figure 8.** The same as Figure 6, except for (a) wind speed (ms<sup>-1</sup>) and wind direction (°) during three heavy dust events on 30 April, 1 May, and 10 June 2012. There were no measurements of aerosol scattering coefficient ( $\sigma_{sp}$  in Mm<sup>-1</sup>) on 10 June due to equipment failure.





1320

**Figure 9.** Time evolutions of aerosol optical depth (AOD) at five wavelengths (400, 500, 675, 870, and 1018 nm) versus Ångström exponent ( $\alpha$ ) at 400–870 nm on (a) 14 May, (b) 9 June, (c) 30 April, and (d) 10 June 2012. Note that Figures 9(a)–9(b) are adopted from *Bi et al.* (2014) with an addition of the Ångström exponent plot in the original publication.



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Figure 10. Diurnal variations of ground-based measured-measurements of 1-minute average (a)
direct, (b) diffuse, and (c) global irradiances, and (d) downward long wave irradiance under
completely clear-sky conditions (14 May, 29 May, and 9 June) and dust events (30 April and 10
June).