Source Sector and Region Contributions to BC and PM$_{2.5}$ in Central Asia

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Abstract

Particulate matter (PM) mass concentrations, seasonal cycles, source sector and source region contributions in Central Asia (CA) are analyzed for the period April 2008-July 2009 using the Sulfur Transport and dEposition Model (STEM) chemical transport model and modeled meteorology from the Weather Research and Forecasting (WRF) model. Predicted Aerosol Optical Depth (AOD) values (annual mean value ~0.2) in CA vary seasonally with lowest values in the winter. Surface $PM_{2.5}$ concentrations (annual mean value ~10 µg/m^3) also exhibit a seasonal cycle, with peak values and largest variability in the spring/summer, and lowest values and variability in the winter (hourly values from 2 – 90 µg/m^3). Surface concentrations of black carbon (BC) (mean value ~0.1 µg/m^3) show peak values in the winter. The simulated values are compared to surface measurements of AOD, and $PM_{2.5}$, PM_{10}, BC, organic carbon (OC) mass concentrations at two regional sites in the Kyrgyz Republic (Lidar Station Teplokluchenka (LST) and Bishkek). The predicted values of AOD and PM mass concentrations and their seasonal cycles are fairly well captured. The carbonaceous aerosols are underpredicted in winter, and analysis suggests that the winter heating emissions are underestimated in the current inventory.

Dust, from sources within and outside CA, is a significant component of the PM mass and drives the seasonal cycles of PM and AOD. On an annual basis, the power and industrial sectors are found to be the most important contributors to the anthropogenic portion of $PM_{2.5}$. Residential combustion and transportation are shown to be the most important sectors for BC. Biomass burning within and outside the region also contributes to elevated PM and BC concentrations. The analysis of the transport pathways and the variations in particulate matter mass and

54   composition in CA demonstrate that this region is strategically located to characterize regional

55   and intercontinental transport of pollutants. Aerosols at these sites are shown to reflect dust,

56   biomass burning and anthropogenic sources from Europe, South, East and CA, and Russia

57   depending on the time period.

58   Simulations for a reference 2030 emission scenario based on pollution abatement measures

59   already committed to in current legislation show that $PM_{2.5}$ and BC concentrations in the region

60   increase, with BC growing more than $PM_{2.5}$ on a relative basis. This suggests that both the health

61   impacts and the climate warming associated with these particles may increase over the next

62   decades unless additional control measures are taken. The importance of observations in CA to

63   help characterize the changes that are rapidly taking place in the region are discussed.

1. Introduction

Central Asia (CA), a region of republics located between Europe and Asia, faces severe environmental problems, with origins dating back to the 1960s and best symbolized by the Aral Sea catastrophe (Whish-Wilson, 2002). The Aral Sea has shrunk to only about 30 percent of its 1960 volume and roughly half its geographical size due to diversion of water for crop cultivation and other purposes. The resulting desertification of the lake-bed has resulted in extensive dust storms from the region, which have impacted the surrounding agriculture, ecosystem, and the population's health.

The 2012 Environmental Performance Index (EPI), which tracks performance of 132 countries across a variety of environmental and ecosystem vitality indicators, ranked CA countries among the weakest performers (Kazakhstan 129, Uzbekistan 130, Turkmenistan 131, Tajikistan 121, and Kyrgyzstan 101) (Emerson et al., 2012). CA and the surrounding areas are developing quickly and air pollution emissions are projected to increase significantly for the next several decades (Shindell et al., 2012). In recognition of the need to improve the environmental conditions in the region five CA countries have formulated the Framework Convention on Preservation of Environment for Sustainable Development of CA (UNEP, 2006).

Despite the awareness of the environmental conditions, it remains an understudied region and there is a general lack of air pollution observations within CA. Furthermore the recent assessment of the intercontinental transport of pollution (HTAP, 2010) has indicated that the major transport pathway of pollution from Europe to Asia is via low altitude flows passing through CA. The magnitude of the pollution transport from Europe to Asia is highly uncertain in large part due to the lack of observations of pollutants along this pathway. To help better characterize the air pollution levels and the transport pathways in the region a study was

87   undertaken between Russia, Kyrgyz Republic, and USA scientists to observe and model aerosols

88   in the region. Measurements of particulate matter (PM) mass and composition were taken at two

89   locations in the Kyrgyz Republic (Lidar Station Teplokluchenka (LST) and Bishkek) and

90   modeling analysis was performed to assess the contributions of local, regional and distant

91   sources to the PM concentrations in the region (Miller-Schulze et al., 2012, Chen et al., 2012,

92   2013).

93   In this paper we present a modeling analysis of $PM_{2.5}$, PM_{10}, ($PM_{2.5}$ refers to particles in

94   the size range of less than 2.5 µm aerodynamic diameter (AD) and PM_{10} refers to particles in the

95   size range of less than 10 µm AD), black carbon (BC) and organic carbon (OC) mass

96   concentrations and aerosol optical depth (AOD) over the time period of April 2008 to July 2009.

97   The Sulfur Transport and dEpostion Model (STEM), a hemispheric chemical transport model

98   (D'Allura et al., 2011), is used to estimate spatial and temporal variations in PM in CA, and to

99   assess the contributions to PM from wind-blown dust, open biomass burning, and anthropogenic

100   sources, and different geographical source regions and source sectors (transportation, power,

101   industry and residential). The simulated values are compared to surface measurements of AOD,

102    $PM_{2.5}$, PM_{10}, BC, OC mass concentrations at the two regional sites in CA. The transport of

103   aerosols into CA is also explored through three dimensional backward trajectory analysis.

104   Transport from CA and their impacts on downwind areas are also analyzed via forward trajectory

105   analysis. Finally we present results of how the PM concentrations may change in the future using

106   emission scenarios for 2030 that reflect possible air quality and climate policies.

107

108   **2. Data and Methods**

2.1. Observations

Surface observations from two sites established in the Kyrgyz Republic to measure PM concentrations and AOD in CA are used in the analysis. The locations of the Bishkek (42°40'46.65"N, 74°41'38.13"E, elevation 1743 above sea level (ASL)) and LST (42°27'49.38"N, 78°31'44.17"E, elevation 1921 m ASL) sites are denoted by circle and triangle markers, respectively, in Fig. 1. Both sampling sites are in mountain ranges with valleys to the north, with mountains that reach elevations greater than 3500 m ASL south of the Bishkek site and 4600 m ASL south of the LST site, and essentially no population to the south. At each site, $PM_{2.5}$ mass was measured continuously with tapered element oscillating microbalance (TEOM) instruments and $PM_{2.5}$, PM_{10}, BC, and OC were obtained using filter-based sampling with samples collected for 24 h every other day. AOD was measured every day at 10:30 am local time (LT) using Microtops-II sun-photometers (SP). A stationary three wavelength aerosol Lidar measured vertical profiles of extinction and depolarization on an event basis at the LST site. The Lidar vertical profiles provide information on vertical distribution of the particles, and were also used to calculate AOD from the Lidar Extinction (LE) profiles and to estimate the height of the planetary boundary layer (PBL) as described in Chen et al., (2013). These observations sites are now part of the UNEP project ABC measurement network (http://www.rrcap.ait.asia/abc/index.cfm). Further details of the study can be found in Miller-Schulze et al., (2011). Observations from these sites were obtained for the period April 2008 to July 2009 (the TEOM measurements were available from April 2008 and filter measurements began from July 1, 2008).

130   The Moderate Resolution Imaging Spectroradiometer (MODIS) collection 5.1 Level 2

131 AOD products (~ 10 km horizontal resolution) at 550 nm wavelength from Terra and Aqua

132 satellites were used to compare the observed and simulated AOD. The MODIS Level 2 data were

133 used and included land and ocean AOD retrieved via the dark target algorithm (Remer et al.,

134 2005, Levy et al., 2007), and the Deep Blue AOD over land (Hsu et al., 2004, Hsu et al., 2006),

135 which can retrieve AOD over bright and desert surfaces. This is particularly relevant for the CA

136 region, which contains major dust sources in western China, South Asia, the Middle East, and

137 Africa (Ginoux et al., 2001). The MODIS Level 2 to Level 3 averaging procedure outlined in

138 Hubanks et al., (2008) was employed to interpolate the 10 km Level 2 AOD products to the 60

139 km horizontal model resolution on a daily basis. Level 2 QA flag weightings were used to reduce

140 the uncertainty associated with the MODIS retrievals. In grid cells where both the MODIS dark

141 target and Deep Blue AOD were retrieved, the mean value of the two was used.

142 **2.2. Modeling System**

143 2.2.1 Meteorological Model

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145   The Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) version

146 3.2 was used to generate the meteorological fields needed for simulating the transport patterns in

147 the STEM model (D'Allura et al., 2011). The WRF simulations for each day were initialized

148 using the meteorological boundary conditions obtained from National Centers for Environmental

149 Prediction (NCEP) Final Analysis (FNL, http://rda.ucar.edu/datasets/ds083.2/). A daily 24 hour

150 spin up time for WRF was used (i.e. WRF was run for 48 hours each day and the first 24 hours

151 were treated as spin up and were discarded). The STEM model simulation time period was from

152 April 2008 to July 2009.

153   2.2.2 Chemical Transport Model

154   The STEM model (Carmichael et al., 2009) was used to simulate the mass of sulfate, BC,

155   OC, other primary emitted $PM_{2.5}$, and other primary emitted PM_{10} (i.e., non-carbonaceous PM

156   such as fly ash, road dust, and cement), which were simulated as a single mass component with

157   aerodynamic diameters less than or equal 2.5 µm, and between 2.5 and 10 µm (denoted as coarse

158   fraction), respectively and referred to in this paper as $OPM_{2.5}$ and OPM_{10}, dust (fine and coarse)

159   and sea salt (fine and coarse). Nitrate and secondary organic aerosols (SOA) were not included in

160   the model for this application. The nitrate aerosol is estimated to be a minor component of the

161   PM mass in CA (Baurer et al., 2007). The importance of SOA will be discussed later in the

162   paper. The dry deposition of aerosols was modeled using the "Resistance in Series

163   Parameterization" (Wesely and Hicks, 2000) and wet deposition was calculated as a loss rate

164   based on the hourly precipitation calculated from the WRF model. Further details of the wet

165   scavenging can be found in Adhikary et al., (2007). The modeled AOD at 550 nm wavelength

166   was calculated using the simulated three dimensional aerosol distributions and species specific

167   extinction coefficients as described in Chung et al., (2010).

168   2.2.3 Modeling Domain

169   The STEM and WRF computation domains were identical, with a 60 ×60 km horizontal

170   resolution (249 ×249 horizontal grid cells) and 22 vertical layers up to 10 hPa. The domain (Fig.

171   1) covered much of the northern hemisphere in a polar stereographic projection, centered over

172   the Arctic region and extended to 35° N to include the major emission regions of North America,

173   Europe, and Asia. This modeling system has been applied to simulate aerosol distributions in

174   several field campaigns as described in D'Allura et al., (2011) and further details describing the

175   model can be found there. STEM was initialized with a one month spin up using March 2008.

176   Much of the analysis for this paper is focused on the domain denoted by the rectangle centered

177   over CA shown in Fig. 1. This domain has large gradients in topography (insert Fig. 1), which

178   significantly impact the transport patterns in the region.

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180   2.2.4 Air Mass Trajectories

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182   The CA observation sites are impacted by dust, anthropogenic pollution, and biomass

183   burning emissions from various source regions. To further understand the transport pathways and

184   source region influences on the PM distributions at these sites, three dimensional ten day air

185   mass trajectories (both forward and backward in time) from each site were calculated for the

186   entire time period (April 2008 – July 2009). In this trajectory analysis, we utilized the three

187   dimensional wind fields (including u, v and w components) along with the above ground level

188   (AGL) altitude simulated by the WRF meteorological model consistent with Dallura et al., 2011

189   study. These trajectories describe the general flow patterns based on wind fields alone and

190   provide useful information about the history of air mass particularly the influence of source

191   regions over which the air mass had resided before arriving at the site of interest. Note that these

192   trajectories do not account for any other atmospheric processes such as diffusion or chemical

193   evolution along its path (Kurata et al, 2004 and Guttikunda et al., 2005).

194   To understand the differences in transport patterns at the surface and aloft, and to study

195   the impact of topographic gradients in the vicinity of the sites, trajectories were initialized at

196   different altitudes (0.1 (100m), 0.3 (300m) 0.5 (500m), 1, 2, 3, and 5 km) a.g.l at the site

197   locations (i.e. latitude and longitude) daily every 3 hours for a ten day period both backward and

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198   forward in time. The trajectories were terminated when they touched the ground, or went out of

199   the model domain or exceeded the ten day calculation period. The trajectories (at or below 1 km)

200   were used to characterize transport pathways impacting the surface concentrations at these sites,

201   which are discussed later in section 3.5.

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203   **2.3. Emissions**
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205   **2.3.1 Base emissions**
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207   Anthropogenic emissions of BC, OC, $PM_{2.5}$, PM_{10}, and SO_2 were based on the ARCTAS

208   emissions described in D'Allura et al., (2011), but updated with newly available information. For

209   India and China BC, OC, and SO_2 emissions from Lu et al., (2011) were utilized, and INTEX-B

210   emissions were used for the rest of Asia (Zhang et al., 2009). For Europe, the EMEP 2008

211   (http://www.ceip.at/webdab-emission-database/officially-reported-emission-data) emissions were

212   used for SO_2, $PM_{2.5}$, and PM_{10}, and the EUCAARI 2005 inventory was used for the

213   carbonaceous particles (BC and OC) (Visschedijk et al. 2009 and Denier van der Gon et al.

214   2009). The shipping emissions came from the IIASA base year 2005 inventory (UNEP and

215   WMO, 2011). Mass conservative regrinding tools including MTXCALC and MTXCPLE from

216   the IOAPI m3tools suite (http://www.baronams.com/products/ioapi/AA.html#tools) were used to

217   interpolate the input raw emissions described above on to the model grid.

218   Anthropogenic emissions for SO_2, BC and OC were available by major economic sectors;

219   i.e., transportation, residential, industry, and power. The industry and power sectors were treated

220   as small and large point sources, respectively, and emitted into the first 6 model levels (lowest 2

221   km). The residential and transportation emissions were treated as area sources and partitioned

222   into the first two model levels with a 90 – 10 percent split. Monthly emission allocation factors

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223 were applied over India and China for the economic sectors from Lu et al., (2011). The rest of

224 the domain (i.e. excluding India and China) used same emission rates for all months due to

225 unavailability of monthly emission allocation factors.

226 The Fire Inventory from NCAR (FINN v1) was used for BC, OC, CO, SO_2, $PM_{2.5}$ and

227 PM_{10} biomass burning emissions from forest, grassland and crop residual fires. The FINN

228 database, which is based on MODIS fire detection as thermal anomalies, provides global

229 coverage of fire emissions at a spatial resolution of ~ 1km on a daily timescale (Wiedinmyer et

230 al., 2011). The WRF-Chem fire utility (http://bai.acd.ucar.edu/Data/fire/) was employed to

231 interpolate the speciated FINN emissions to the WRF model grid. The gridded two-dimensional

232 FINN emissions were used as input to the WRF-Chem (Grell et al., 2005) plume rise model

233 (Grell et al., 2011), which implements the Freitas et al., (2007) and Freitas et al., (2010)

234 algorithm to compute injection heights and to calculate the vertical distribution of fire emissions

235 at an hourly time step, which were further utilized as input to STEM model simulations.

236 Sea salt and dust emissions were calculated using the WRF meteorological fields based

237 on the methods described in Gong, (2003) and Uno et al., (2004), respectively. The dust

238 emissions were further constrained with snow cover (SNOWC variable from WRF output) and

239 only grid cells with snow cover < 1% were used for dust emission calculations.

240 Fig. 2 shows the annual gridded anthropogenic SO_2 and BC, dust, and biomass burning

241 $PM_{2.5}$ emissions in Gg per grid in and around CA. Large BC emission hotspots can be seen over

242 the Indo-Gangetic plain and eastern China. Significant BC emissions are also seen over Europe,

243 but are relatively lower in intensity than the Asian sources. The SO_2 emissions show Eastern

244 China as the largest source region followed by regions of South Asia, Europe, and Russia. The

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245　　major natural dust emission sources (Fig. 2c) include Africa, the Middle East, CA, Western India

246　　boundaries, and Western China. The major sources of biomass burning are Eastern Europe,

247　　portions of Siberian (between 40 – 60° N), Southeast Asia, Southern China and India (Fig. 2d).

248　　　　Dust emissions have a strong seasonal cycle. The major dust sources in the region (Fig.

249　　1) are located to the east, west and south of the observation sites and include the cool winter

250　　deserts around the Aral and Caspian seas and those in western China and northern Pakistan as

251　　well as the sub-tropical deserts in western India, around the Persian Gulf and northern Africa.

252　　The emissions from the cool winter deserts occur when the surfaces are free of snow cover (from

253　　March through October). Emissions from the sub-tropical deserts can occur throughout the year.

254　　　　The open biomass burning emissions that impact CA also have a strong seasonality with

255　　minimum impact in winter (Supplemental Materials Fig. S1). Fires typically begin in the spring

256　　in Siberia along 50° N latitude and in northern India and South East Asia and in summer the high

257　　latitude burning shifts to the west. In October the fire activity decreases and remains low until

258　　spring, with the most active fire regions associated with agricultural burning in northern India

259　　and southeast China.

260　　**2.3.2 Future Emissions Scenarios**

261　　　　In addition to the base emissions, a series of simulations were analyzed using emission

262　　scenarios for 2030. These scenarios were developed for the WMO/UNEP report that looked at

263　　short lived climate pollutants as described in Shindell et al., (2012) and Anenberg et al., (2012).

264　　The reference scenario for 2030 was based on the implementation of control measures currently

265　　approved in the various regions and assumed their perfect implementation. The 2030 reference

266 scenarios were developed from a reference global emissions inventory with a 2005 reference

267 year, and assumed significant growth in fossil fuel use relative to 2005, leading to increases in

268 estimated CO_2 emissions (45%). Abatement measures prescribed in current legislation were

269 projected to lead to reductions in air pollutant emissions, which varied by pollutant and region.

270 In the 2030 reference scenario, total primary $PM_{2.5}$ emissions remain approximately constant,

271 while BC and OC decline by a few percent. However, in the study domain emission changes

272 varied widely. BC emissions increased by 10 – 100% in CA, South and Southeast Asia and in

273 western China, and decreased in East Asia and Europe. The $PM_{2.5}$ emissions showed similar

274 regional changes but grew at smaller rates (10 – 40%). SO_2 emissions generally increased

275 throughout the region by 10 – 20%. Spatial maps of emission changes for the 2030 reference

276 scenario are presented in Figs. S2b, S3b, and S4b.

277   A series of emission control scenarios for 2030 were developed to evaluate the impact of

278 additional abatement measures designed to reduce the levels of short lived climate pollutants

279 (e.g., BC). The BC measures in the scenarios included two different sets of assumptions (low and

280 lowest). The first focused on reductions from incomplete combustion sources. These included

281 implementation of Euro 6 equivalent vehicle emission standards (requiring installation of diesel

282 particulate filters) and improving traditional biomass cook stoves in developing countries

283 (assuming 25% decrease in BC and 80 – 90% decreases in OC, CO, non-methane volatile

284 organic compounds(NMVOC), methane, and direct $PM_{2.5}$, relative to emissions from traditional

285 stoves). Under this scenario BC and $PM_{2.5}$ emissions in the study region are projected to decrease

286 throughout most of the domain, with SO_2 emissions showing almost no change (see Figs. S2c,

287 S3c, and S4c).

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288   The lowest option assumed the additional elimination of high-emitting vehicles, biomass

289   cook stoves (in developing countries), and agricultural waste burning. These BC measures in the

290   lowest scenario reduced global anthropogenic BC emissions by 75%. Measures targeting BC

291   also substantially reduced total primary $PM_{2.5}$ (-50%), OC (-79%), NO_x (-27%), and CO (-44%).

292   These BC measures have little impact on SO_2 emissions. Projected emissions of BC and OC

293   under these scenarios are reduced most in Asia, followed by Africa. North America, and Europe.

294   These measures were also combined with a scenario designed to stabilize greenhouse

295   gases at 450 ppm of CO_2 equivalent (lowest+450ppm scenario), consistent with a global average

296   temperature increase of ~2° C. These CO_2 measures reduced SO_2 (-30%) (Fig. S4d) and NO_x (-

297   20%), but had little further impact on BC (~5% decline, Fig. S2d) since the major sources of CO_2

298   differ from those of BC. $PM_{2.5}$ emissions were substantially further reduced under this scenario

299   (Fig. S3d).

300   **2.4. Simulations Analyzed**

301   Several simulations were analyzed for this paper. The *base* simulation included all

302   sources and used the meteorology from the WRF model for the period April 2008 – July 2009.

303   To investigate the contributions from specific source sectors, additional simulations were

304   performed where emissions from one sector were set to zero everywhere. The contribution from

305   each sector was calculated as the difference between the base simulation and the simulation with

306   emissions from that particular sector set to zero. This was repeated for each sector and for

307   biomass burning. Additional simulations were performed to assess the source contribution from

308   specific regions to the particle levels in CA. The specific regions used are shown in Fig.1. In

309   these simulations all anthropogenic emissions were set to zero in that region. In a similar manner

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310   regional dust and fire sources were also studied and the source regions are also shown in Fig.1.

311   Finally, simulations were performed using the *reference* 2005 and 2030 emissions and the *low*

312   and *lowest + 450 CO$_2$* 2030 scenarios (described above). For these runs, the meteorology, open

313   biomass burning, and dust emissions were the same as in the base run (i.e., 2008/2009).

3. Results and Discussion

315
3.1. Regional Perspective

317   CA is a region with high aerosol loadings as shown in the mean MODIS retrieved AOD

318   at 550 nm for the time period of study (April 2008 – July 2009) (Fig. 3).AOD (period mean)

319   throughout CA (~ 45-90° E, 35-50° N) are greater than 0.25, with the highest regional values

320   around the desert areas near the Caspian and the Aral seas. There are also high values (>0.6)

321   along CA's eastern border, which reflect the deserts and rapidly developing cities in western

322   China, and to the south over Pakistan and northern India.

323   The predicted period mean AOD spatial distribution shown in Fig. 4d captures the main

324   observed features. The period-mean predicted surface concentrations of PM$_{2.5}$, BC, and total dust

325   (fine and coarse) are also plotted. The period mean PM$_{2.5}$ concentrations in CA (10 to 35μg/m^3)

326   have a similar geographical distribution as AOD. Dust is the major component of predicted PM$_{10}$

327   in CA and concentrations are high (25-100 μg/m^3). The BC levels in CA are typically less than

328   0.3 μg/m^3 and its spatial pattern reflects contributions from both anthropogenic and biomass

329   burning sources.

3.2 Comparison with surface observations in CA

331   The surface observations at the two CA sites provide the opportunity for the first time to

332   evaluate the performance of chemical transport models in estimating the distribution of aerosols

333   in CA and to assess the emission estimates in the region. A comparison of the predicted and

334   observed meteorology is presented in Fig. 5, where the distributions of key meteorological

335   parameters for the entire measurement period are shown as box-plots. The model accurately

336   predicted the magnitude and variability in temperature and relative humidity. For example the

337   model mean value of temperature and relative humidity are 279.3K and 61.6% in comparison to

338   the observed values of 280.3K and 59.2% at the LST site. The model wind speeds were biased

339   high by about 30% (mostly in winter, see section 3.4) and the direction had a southwest bias.

340   These biases are due in part to the 60 km model grid size, which is too coarse to capture the steep

341   gradients in topography in and around the observation sites, and to the site placement in small N-

342   S oriented valleys. More details regarding the evaluation of the modeled meteorology can be

343   found in the Supplemental Materials, Table S1.

344   The observed and modeled distributions of AOD and PM are compared in Fig. 6. The

345   AOD observations based on the LE on average are ~50% larger than those from the SP. Modeled

346   AOD on average are ~20-30% higher when compared to SP at the Bishkek and LST sites and ~1

347   % lower when compared to the LE values. The variability in the predictions is slightly under-

348   estimated. $PM_{2.5}$ is over predicted (~50%) and the spread is accurately captured, while PM_{10} is

349   over predicted by ~70%. This leads to an underestimation of the $PM_{2.5}/PM_{10}$ (0.4 predicted

350   versus 0.5 observed) and also helps account for the overestimation in modeled AOD (by ~20-

351   30%).

352   Chemical analysis of the filter and soil samples in the CA dust regions have been used to

353   estimate the dust contribution to measured PM at the two sites and to help identify source regions

354   of importance (Park et al., 2014). The emission regions within CA, including around the Aral

355   Sea, and western China were identified as the most important dust sources, which is consistent

356   with the regions identified in the simulations. Dust was estimated to comprise between 5-40% of

357      $PM_{2.5}$ mass at the LST site and to vary by season (minimum values in winter). The observation-

358   based estimates of dust percentage contribution suggest that modeled dust is over predicted by

359   ~2 times. Thus it appears that dust is a main reason for the over prediction of $PM_{2.5}$ and PM_{10},

360   and that dust emission models need to be refined for CA applications.

361 **The overestimation in PM mass at the surface could also be impacted by**
362 **errors in the modeled PBL heights. The PBL height as determined by the**
363 **Lidar aerosol profiles varies seasonally and is highest in the summer**
364 **(from 2-4 km AGL) and lowest in the winter (Nov-Feb, 0.5-1.5 km AGL)**
365 **(Fig. S5). The predicted PBL heights show a similar seasonal cycle with a**
366 **tendency to under-predict the heights in all seasons as indicated by the**
367 **comparison of the distributions of the observed and predicted values**
368 **(Fig. 5), and this occurs in all seasons (Fig S5). The lower PBL height in**
369 **the model contributes a systematic high bias in surface concentrations**
370 **driven by near surface emissions.**
371   Further statistical details of the model-observation comparison can be found in the

372   supplemental materials (Tables SM2 & 3). The seasonal variability in the observations is

373   discussed in further detail later (section 3.4).

374   We have evaluated this model framework in other regions outside CA including arctic

375   region and continental US, which are summarized below. Dallura et. al., 2011 evaluated the

376   performance of the WRF-STEM modeling framework (used in this study) through the case study

377 of the ARCTAS mission DC-8 flight conducted on July 9 2008. The WRF model compared well

378 with observed meteorological variables (including temperature, pressure, wind speed and wind

379 direction) along the flight and correctly simulated the various meteorological processes (i.e.

380 cyclonic circulation, low pressure system, anticyclones and geostrophic wind flows) that

381 facilitate transport of pollutants into the Arctic region. This study also compared CO along the

382 flight and found that the simulated values were able to capture the magnitude and temporal

383 variability seen in observed values. The study also found that the fires from North Asia and

384 anthropogenic CO from China were the major sources contributing to observed CO levels along

385 the flight suggesting that the midlatitude pollution and summertime forest fires/biomass burning

386 transport events were well represented in the STEM model.

387 The study by Huang et al., 2012 used the STEM model to simulate summertime sectoral

388 and regional contributions to BC over continental US and evaluated the model performance aloft

389 and at surface using measurements from ARCTAS mission DC8 flights conducted on June 20

390 and 22 and 24 that sampled over California and Interagency Monitoring of Protected Visual

391 Environments (IMPROVE) surface site network. Huang et al., 2012 found that the predicted BC

392 values captured many of the vertical features seen in the flight observations with a tendency to

393 over predict surface BC and > 4km.   The comparison of predicted BC with observed values

394 from IMPROVE network showed that the model was able to capture the gradients in BC values

395 with high concentrations in western and eastern NA regions with slight positive and negative

396 biases over the mountain regions and the eastern US and CA.

397 Since the Huang et al., 2012 study was focused on summer, we evaluated the simulated

398 BC values with IMPROVE BC observations for the simulation time period (i.e. April 2008 –

399   July 2009) used in this study. The IMPROVE BC analysis is limited to 27 sites on the US west

400   coast (i.e. west of 120 W) to be consistent with the focus of this paper (i.e., long range transport

401   of pollutants on a hemispheric scale). The mean observed and modeled BC for the West Coast

402   sites are ~0.16 and 0.19 µg m−3 (n = 3759 points) with mean bias and RMSE of 0.03 µg m−3and

403   ~ 32 % respectively. The mean simulated/observed BC at surface sites is ~ 1.2, which compares

404   well with the corresponding ratio of 1.6 reported in the global model performance over North

405   America region by Koch et al., 2009. The detailed analysis of the IMPROVE BC sites and

406   associated seasonality will be the addressed in a future paper.

407   These studies indicate that the simulated transport patterns are able to capture the main

408   important features of the hemispheric flows at the scale discussed here for improving the current

409   understanding of the source receptor relationships of aerosols at intercontinental scales. The

410   prediction of aerosol mass and composition at the hemispheric scales is also quite good, but has

411   considerably higher uncertainties associated with emission estimates (in particular forest

412   fires/biomass burning and natural dust emissions) and wet removal processes (Bates et al., 2006).

413   **3.3 Source Contributions to PM$_{2.5}$**

414   Model simulations were performed to identify the component, source region and

415   emission sector contributions to PM$_{2.5}$ mass. Period means for the spatial average over the entire

416   CA region (see Fig. 1) and for the grid cells for the Bishkek and LST observation locations are

417   presented in Fig.7, and their comparison provides insights into the spatial variability of PM and

418   its sources within CA, and how representative the observation sites are at characterizing CA PM.

419   The component contribution to AOD at the sites and for the CA average are similar, with the

420   major contributions coming from fine dust, sulfate, and OC. Spatial maps of mean percent

421 contributions of the various components (i.e., BC, OC, sulfate, OPM, dust and sea salt) to AOD

422 and $PM_{2.5}$ mass are presented in Figs. S6 & S7, respectively. Coarse particles contribute ~10% to

423 mean AOD. Dust accounts for >60% of the calculated $PM_{2.5}$ mass at the observation sites and for

424 the CA region. The dust source regions (see Fig. 1) contributing to $PM_{2.5}$ vary within CA. Dust

425 from the CA source regions has the largest influence on the region mean dust- $PM_{2.5}$ mass. At the

426 LST site, which is located in the far east of CA, western China dust sources have their largest

427 influence (~40%). African and Middle East source regions have their largest influence on the

428 Bishkek site (20 and 15%, respectively), and collectively contribute ~25% to regional CA dust

429 $PM_{2.5}$.

430   The source region contributions to the non-dust $PM_{2.5}$ are very similar for the Bishkek

431 and LST sites, with CA sources making the largest contribution (~50%) followed by Europe

432 (~20%), the Middle East (~15%), and biomass burning (~15% from all sources). For the entire

433 CA region the European source contribution is as large as the CA sources (~30% each), with

434 larger contributions from biomass burning and Russia sources and smaller contributions from

435 Middle East emissions than at the observation sites. The significant contribution from Europe

436 sources reflects their large anthropogenic emissions and the general westerly transport patterns.

437 On average the impact from South Asia sources are small. North America sources contribute

438 between 3 to 7% to $PM_{2.5}$ in CA. Of the biomass burning contribution to $PM_{2.5}$, the Siberian and

439 European fires (see Fig. 1 for fire regions) contribute 63 and 25%, respectively, with

440 contributions from South/Southeast Asia and North America fires each contributing ~5%. The

441 power and industrial sectors are identified by the simulations as the largest contributors (~40%

442 each) to non-dust $PM_{2.5}$ mass in CA.

443

3.4. Seasonal Variations in AOD and PM at the Observation Sites

AOD has a seasonal cycle with the lowest values in winter and highest values in spring and summer as shown in Fig. 8 by the AOD observations at the LST site from both the SP and the LE profiles. A similar seasonal cycle was observed at the Bishkek site (not shown) and in the MODIS AOD retrievals (Fig. S8). The modeled AOD captures this seasonal variation, with a tendency to over predict the values during the periods with the lowest observed AOD.

The seasonal cycles in $PM_{2.5}$ (Fig. 9) and PM_{10} are similar to those for AOD, with minimum values in October – February. At the LST site $PM_{2.5}$ from both the filter based and the non-volatile TEOM measurements are plotted. Both the observations and the model find a strong similarity in the time series at the two sites.

There is also a clear seasonality in the surface meteorology in the region as shown by the time series in surface temperature and relative humidity at the two sites (Fig. 10). There are distinct temperature minima in the winter and relative humidity minima in the summer. However there is not a clear seasonality in wind speed and direction, and the winds are generally from the south and less than 4m/s throughout the year at the LST site (not shown).

The source region and component contributions exhibit seasonal variability as shown by the modeled contributions to $PM_{2.5}$ mass in Fig. 11. Dust is found to be the main driver of the seasonal cycle of $PM_{2.5}$. The dust contribution to $PM_{2.5}$ is peak in spring and minimum in winter (<20%). During this time period the transport of air masses to the sites are from the west and the southwest. When the transport is from the east then dust sources from western China can impact

22

464   the stations. This transport pattern occurs episodically throughout the year, with contributions

465   from western China sources as large as 20 to 50%. The dust seasonal cycle is in turn influenced

466   by the seasonal variations in meteorology that drives the dust emissions and transport. The

467   seasonal changes in the dust source regions can be seen in the seasonal spatial maps of AOD

468   (Fig. S8). Throughout the domain, AOD in the dust regions are highest in March – October and

469   lowest in winter (Fig. S8) as the nearby dessert regions are snow covered.

470   Biomass burning also adds to the seasonal cycle, and its contribution is minimum in the

471   winter. South Asia sources can impact the sites in the winter time. The periods when North

472   America sources impact the site are associated with strong transport events across the Atlantic

473   and subsequent subsidence towards the surface associated with high pressure systems as they

474   move towards CA. The transport pathways are discussed in more detail in Sect. 3.7.

475   **3.5 Source Contributions to BC**

476   Because of its dual role as an air pollutant and as a climate warming agent there is special

477   interest in understanding the regional and sector contributions to BC (Ramanathan and

478   Carmichael, et al., 2008). BC comprises on average only about 1-2% of $PM_{2.5}$ mass in CA. The

479   period mean predicted BC surface concentrations are ~0.1 $\mu g/m^3$ at the two observation sites and

480   0.15 $\mu g/m^3$ for the CA regional average. As shown in Fig. 7, European emissions contribute

481   ~50% to the mean BC concentrations in CA, while Middle-eastern and biomass burning sources

482   each contribute ~15-20%. Residential and transport are the most important sectors each

483   contributing ~30% to BC in CA, followed by industry (~20%), and with power the least

484   important. This is in contrast to the sector contributions to non-dust $PM_{2.5}$ mass, where power

485   and industry are the most important sectors. On average biomass burning contributes ~10% to

486   BC mass, with Siberian and European fires accounting for 61% and 33%, respectively. The

487   source contributions to OC are shown in Fig. S9.

488   There is also large seasonal variability in BC concentrations and source sector/region

489   contributions (Fig. 12). BC surface concentrations show the highest values in fall/winter (as do

490   the observations), when there is maximum contribution from the residential sector, reflecting the

491   wide-spread use of biofuels and coal for heating in the region. The source region contributions

492   vary by season, with maximum contributions from Europe and China. South Asia sources

493   contribute in the winter. Biomass burning also is an important source of BC and plays an

494   important role in influencing daily and seasonal variability in BC concentrations.

495   Predicted BC captures the seasonality and the magnitude of the spring and summer

496   values as observed, but concentrations are biased low in the fall/winter. Median BC

497   concentrations (and variability) are underestimated by a factor of 2 at both observation sites (Fig.

498   6 and Tables S2&3)). The high wind speed bias in winter (~ factor of 2), should result in too

499   rapid dispersion and could contribute to the negative bias, but the negative bias in the PBL

500   heights should lead to higher predicted concentrations. Thus this negative bias is likely related to

501   emissions (an indication of an underestimation of the heating fuel use).

502   The OC concentrations follow a similar seasonal cycle as BC and are also under

503   predicted (Fig. 6 and Tables S2 & 3)). Furthermore the OC/BC ratio is under predicted by a

504   factor of ~3 (Fig. 6). The observed OC/BC ratio follows a seasonal cycle with values >15 in

505   summer and ~5 in September through April. Part of this under prediction in OC and the OC/BC

506   ratio is due to the fact that SOA is not estimated in the model. However a source contribution of

507   OC using the filter data and chemical mass balance (CMB) approach found that SOA sources

24

508 were very low in winter and only ~ 20% in summer (Miller-Schulze et al., 2011). Thus SOA

509 cannot account for the model under prediction of winter values. There appears to be an

510 underestimation of regional OC primary emissions. SOA can however help account for the large

511 values of OC/BC observed in the summer and not predicted.

512   Biomass burning emissions cannot account for the underestimation in winter BC and

513 OC. The largest impact of fires at the observation sites is in the late summer, when the fires are

514 concentrated in western Russia and the wind direction is such that the smoke is transported into

515 CA. Fires from South Asia can impact the sites associated with the fires and high pollution levels

516 in northern India and with winds from the south, which can occur in late fall, but not frequently.

517   The fact that BC and OC are systematically under predicted in the winter suggests that

518 local/regional emissions during the heating season may be underestimated. This is supported by

519 the results of the CMB analysis of OC discussed above that found the contribution from biofuel

520 combustion increased 2-3 times in the fall and winter periods. The uncertainty in emissions can

521 also be partly caused by the lack of seasonal emissions over this region as described earlier in

522 section 2.

523

524 **3.6 Vertical Distributions**

525   Figure 13 shows the predicted weekly averaged vertical distributions of $PM_{2.5}$, dust, and

526 BC for the entire simulation period at the LST site. These plots show more clearly that much of

527 the variability in the PM loadings is associated with dust and biomass burning episodes (as

528 represented by the enhancements in BC). Typically the high PM episodes show elevated PM

25

529 mass that extend from the surface to 2 to 4 km. The vertical extents show a seasonality

530 associated with seasonal variations in the PBL heights. These vertical distributions indicate that

531 much of the transport of aerosols in CA occurs via low altitude pathways. In some cases there are

532 large amounts of dust and biomass burning aerosol in the 3-6 km altitude range that are

533 decoupled from the surface (e.g., dust in early May 2009), reflecting that some aerosols are lifted

534 out of the boundary layer and are transported at high altitude over CA, enhancing AOD but not

535 contributing to ground-level mass concentrations at the observation sites. These vertical features

536 are confirmed by the aerosol extinction profiles observed at the LST site as discussed in Chen et

537 al., (2012 b). The variation in weekly averaged AOD can be significant (Fig. 13 bottom panel)

538 and is driven by variations in dust and biomass burning emissions.

539

540 **3.7 Transport Pathways**

541 The three dimensional ten day air mass trajectories (described in section 2.2) were utilized to

542 further understand the transport pathways of air masses entering into and exiting out of the CA

543 region and its subsequent impact of source regions on the aerosol distributions at the CA sites.

544

545 3.7.1 Transport into CA

546 The air mass transport into CA is discussed through back trajectories associated with the

547 five events labeled on Fig. 13. These five events represent transport episodes with elevated

548 surface $PM_{2.5}$ (averaged over the three hour time window consistent with trajectory time step)

549 with varying contributions from biomass burning, anthropogenic pollution, and dust sources. In

26

550 each trajectory figure (Figs. 14 and 15), the regions with active dust (blue diamond hatches) and

551 biomass burning emissions (green square hatches) for the event time period and prior ten days

552 are identified and MODIS AOD (values printed in black) overlaid. The trajectories are color

553 coded by region to distinguish the source impacts. The regions include: Africa (blue), Middle

554 East (green), CA (yellow), North Asia biomass (> 50° N, black), Europe (brown), China (red)

555 and South Asia (orange).

556   Figure 14 shows the first two events for August 2008. The first event in early August

557 (Fig. 13, event labeled 1) is a high dust event associated with trajectories from the west passing

558 over the dust regions of the Aral Sea (~45N, 55E) and eastern trajectories passing over the

559 Taklimakan region of western China (90-100E, 40N). These were both regions with active dust

560 emissions as indicated by the elevated MODIS AOD values. Thus the PM increases during this

561 event were due largely to dust emitted from CA and western China deserts. The second event in

562 the latter half of August is characterized by high levels of BC without dust. During this episode

563 the transport to the site was under the influence of a high pressure system located to the

564 northwest and air masses were transported over the active fire region in western Russia.

565   Figure 15 shows winter and spring events. The November episode (event 3) is a period

566 with elevated BC and $PM_{2.5}$ from pollution sources from South Asia (including some fires) and

567 western China and low fire and dust emission activity. The January episode (event 4) is a period

568 of elevated BC with air masses coming from Europe, indicating the influence of anthropogenic

569 pollution coming from this industrialized region, and from CA sources. Dust emissions from CA

570 and Africa were low during this period. The final illustrative episode is for April 2009 (event 5),

571   a period with both elevated levels of dust from western China, CA and Africa and BC from both

572   fire and anthropogenic pollution from Europe, CA and Russia sources.

573   These examples provide insights into the source region contributions to PM mass in CA

574   as presented in Figs. 11 & 12). CA is an ideal location to observe a variety of source regions as it

575   is at the crossroad of transport patterns with air masses impacted from dust, anthropogenic

576   activity and biomass burning from different geographical regions.

577   3.7.2 Long range transport of CA sources

578   The transport pathways out of CA were also evaluated by calculating forward trajectories

579   from the observation sites. Selected forward trajectories initialized at or below 1 km are used to

580   represent the transport of boundary layer PM from CA and these are shown for summer, winter

581   and spring periods in Fig. 16. In these plots the MODIS AOD, dust, and fire emissions plotted

582   for each event represent values averaged over the subsequent ten days and trajectories were

583   stopped if they impacted the surface. During the summer, outflow from CA is towards the north

584   in association with the summer monsoon system. Figure 16a shows the subset of forward

585   trajectories that reside for at least 3 days over the region 48 - 65° N during June 2008.

586   Trajectories typically pass over Russia and reach into the Arctic and also can be caught in

587   westerly storm tracks and reach the higher latitudes of the northern hemisphere. During the

588   winter, air mass transport out of CA is associated with the winter monsoon system and the

589   transport pathway is over Mongolia and then over eastern China as seen in Fig. 16b. In this

590   figure, forward trajectories that pass over the region west of 130° E during late December 2008

591   are shown. Figure 16c illustrates transpacific transport of CA air masses. Plotted are the forward

592   trajectories that stay within the 30 – 50° N region for at least 3 days during the month of April

593   2009. During the spring transport from CA is dominated by strong westerly flows and air masses

594   are transported over China, Korea, and Japan and then across the Pacific, reaching North

595   America in 7 – 10 days.

596   PM arising from dust and anthropogenic emissions from CA impact the entire northern

597   hemisphere as illustrated in the hemispheric maps of seasonal average surface dust

598   concentrations due to CA emissions only (Fig. 17). The predominant westerly flows in all

599   seasons in the northern hemisphere result in the bulk of the CA emissions being transported to

600   the west. The contribution of CA sources to surface PM concentrations of 0.35 $\mu g/m^3$ or greater

601   covers large portions of the northern hemisphere, including the Arctic, all of Asia, much of

602   Europe, and portions of the continental US.

603   There are episodic and seasonal components to the intercontinental transport as shown in

604   the time series of the vertical profiles of $PM_{2.5}$ at Mt. Bachelor, Oregon (43.97N, 121.69W,

605   2700m agl) (Fig. 18). The bulk of the CA particle transport takes place in the free troposphere

606   and impacts surface concentrations in the US as the boundary layer grows and entrains "plumes"

607   aloft. This occurs most frequently in spring, summer and fall. The episodic contributions of CA

608   sources to surface concentrations can exceed $1.5\mu g/m^3$. In the fall, there is also strong transport

609   of dust from CA across Europe and out into the Atlantic.

610

611   **3.8 Future Scenarios**

612   How might PM levels change in CA over the next few decades? To address this question,

613   simulations were repeated for various emission scenarios developed and used in the

29

614   WMO/UNEP assessment on short-lived climate pollutants (Shindell et al., 2012) as described in

615   Sect. 2. Dust and biomass burning emissions and meteorology were the same as those used in the

616   2008/2009 simulations. The period mean changes in surface BC and $PM_{2.5}$ concentrations in

617   2030 for the reference scenario are shown in Fig. 19a, d, respectively. This scenario reflects all

618   present agreed policies affecting emissions and assumes that they are fully implemented. Under

619   this scenario $PM_{2.5}$ increases significantly in South Asia and western China (>50%) and in parts

620   of CA, including the area where the sampling sites are located. $PM_{2.5}$ decreases in Western

621   Europe and Eastern China (< 10%). BC surface concentrations show a similar pattern to $PM_{2.5}$,

622   although covering larger portions of CA with relatively larger increases in BC than in $PM_{2.5}$.

623   These results suggest that health impacts and climate warming due to BC and $PM_{2.5}$ may increase

624   in coming decades unless additional emission control measures are implemented.

625   Results for two other scenarios are also presented in Fig. 19. One scenario specifically

626   targets BC emission reductions in recognition that BC is also a major contributor to atmospheric

627   warming (Ramanathan and Carmichael, 2008). These additional measures significantly reduce

628   2030 BC concentrations by greater than 35% throughout most of the domain, with only a few

629   regions (e.g., Myanmar and eastern Afghanistan) showing increases in BC relative to 2005

630   levels. This scenario assumes that all BC emission reduction measures are perfectly implemented

631   and 100% effective. BC measures also impact emissions of co-emitted pollutants (e.g., OC and

632   SO_2). $PM_{2.5}$ concentrations under this scenario (Fig. 19e) are reduced, but by much smaller

633   amounts, and concentrations still increase relative to 2005 over large regions of South Asia and

634   western China, and parts of CA. These results suggest that health impacts in these regions may

635   increase due to the $PM_{2.5}$ increases whereas positive radiative forcing and health effects due to

636   BC may decrease. When the BC measures are used along with greenhouse gas measures aimed at

30

637   keeping CO_2 levels below 450 ppm, the $PM_{2.5}$ levels in South Asia are lower than 2005 levels

638   (Fig. 19f), with few exceptions (one being Myanmar region). This is due to the large decreases in

639   SO_2 and NO_x emissions under this scenario, resulting in significant reductions in particulate

640   nitrate and sulfate (e.g., a ~30% increase in particulate sulfate in CA).

641   To get a regional perspective of how the future emission changes would specifically

642   impact the CA region (See Fig. 1 for the CA region definition used in this study), we have

643   summarized the changes in emissions and the corresponding concentrations in Table 1.The

644   reference 2030 emissions scenario reports an increase in emissions of BC, SO_2 and $PM_{2.5}$ over

645   Central Asia by ~ 22, 17 and 14 % respectively w.r.t to base 2005 levels. However, the

646   corresponding BC, SO_4 and anthropogenic portion of $PM_{2.5}$ decrease by ~ 5, 12 and 5 %

647   respectively. This suggest that on average the concentration levels go down even though CA

648   emissions increase due to long range transport of pollutants into CA region from the surrounding

649   regions, but locations within CA still increase when dominated by local sources. The Low GWP

650   emission scenario predicts decrease in BC and $PM_{2.5}$ emissions by 31% and 10% respectively

651   with the corresponding decrease in concentrations by ~ 37% and 10 % respectively, while SO_2

652   emissions remained same as the reference 2030 scenario. The lowest + 450 ppm scenario shows

653   a consistent decreasing trend in emissions and concentration of all the species. This analysis

654   suggests that the impact of the changes in major source region emissions and their subsequent

655   transport to Central Asia will play a major role in determining the future aerosol levels in CA.

656   **4. Summary**

657   AOD in CA during the period April 2008 through July 2009 averaged ~0.3 and displayed

658   a seasonal cycle with the lowest values in the winter and highest values in spring to mid-summer

659 with observed values of AOD > 0.6 and even > 0.8. Surface $PM_{2.5}$ measured at two sites in

660 eastern CA averaged ~10 $\mu g/m^3$ but with large variability (hourly values from 2 - 90 $\mu g/m^3$).

661 Surface concentrations of PM also showed a seasonal cycle with peak values and largest

662 variability in the spring/summer, and lowest values and variability in the winter. BC at these sites

663 averaged ~0.1 $\mu g/m^3$ with peak values (~ 1 $\mu g/m^3$) in the winter.

664   The seasonal cycles and source sector and source region contributions to PM in CA were

665 analyzed using the STEM chemical transport model. Dust was the largest component of the

666 $PM_{2.5}$ and PM_{10} mass in the region in all seasons except winter, whereas sulfate was the largest

667 anthropogenic component of the $PM_{2.5}$ mass. Dust was also found to be the major driver of the

668 seasonal cycles of AOD and PM concentrations. On an annual basis the power and industrial

669 sectors were the most important contributors to $PM_{2.5}$, while residential and transportation were

670 the most important sectors for BC. Open biomass burning within and outside the region also

671 contributed to elevated PM and BC concentrations and to the temporal variability.

672   The model simulations showed a systematic over prediction of PM mass. This is most

673 likely due in large part to the over prediction in dust. Carbonaceous PM was underpredicted and

674 it is speculated that the winter emissions associated with residential heating may be

675 underestimated in the current emissions inventory. The predicted wind speeds were biased high

676 (by ~30%) and the direction had a southwest bias. The high bias in wind speeds may also

677 contribute to the over-prediction in PM_{10}, as dust emissions depend strongly on wind speed.

678 Efforts to improve the dust emissions and to improve the wind speed and direction predictions

679 using a finer model resolution are planned. Additional efforts are needed to improve the

680 anthropogenic emissions estimates for CA.

681   Currently there are few measurements in CA that can be used to quantify the

682   intercontinental transport of pollution from Europe to Asia. The analysis of the transport

683   pathways and variations in PM mass and composition observed at the two sites in CA

684   demonstrate that this region is strategically located to characterize regional and intercontinental

685   transport of pollutants. Aerosols at these sites were shown to reflect dust, biomass burning, and

686   anthropogenic sources from South, East, and CA, Europe, and Russia depending on the time of

687   year. For example, during the spring fine particles from Europe and Africa were transported to

688   CA, on to eastern Asia, and then across the Pacific to North America.

689   Observations of PM and its composition in this region are of growing importance as it is

690   estimated that $PM_{2.5}$ levels are likely to increase significantly in Central and South Asia and

691   western China over the next few decades. Simulations for a reference 2030 emission scenario

692   showed that BC concentrations had a larger relative increase than $PM_{2.5}$ concentrations. This

693   suggests that health impacts and climate warming associated with these pollutants may increase

694   over the next decades unless additional control measures are implemented. Continued pollutant

695   observations in CA will help to characterize the changes that are rapidly taking place in the

696   region.

6. References

Adhikary, B., Carmichael, G. R., Tang, Y., Leung, L. R., Qian, Y., Schauer, J. J., Stone, E. a., Ramanathan, V. and Ramana, M. V.: Characterization of the seasonal cycle of south Asian aerosols: A regional-scale modeling analysis, J Geophys Res, 112(D22), D22S22, doi:10.1029/2006JD008143 , 2007.

 Anenberg, S. C., Schwartz, J., Shindell, D., Amann, M., Faluvegi, G., Klimont, Z., Maenhout, G., Pozzoli, L., van Dingenen, R., Vignati, E., Emberson, L., Muller, N. Z., West, J. J., Williams, M., Demkine, V., Hicks, W. K., Kuylenstierna, J., Raes, F., and Ramanathan, V.: Global air quality and health co-benefits of mitigating near-term climate change through methane and black carbon emission controls, Environ. Health Perspect., 120, 831–839, 2012.

Bates, T. S., Anderson, T. L., Baynard, T., Bond, T., Boucher, O., Carmichael, G., Clarke, A., Erlick, C., Guo, H., Horowitz, L., Howell, S., Kulkarni, S., Maring, H., McComiskey, A., Middlebrook, A., Noone, K., O'Dowd, C. D., Ogren, J., Penner, J., Quinn, P. K., Ravishankara, A. R., Savoie, D. L., Schwartz, S. E., Shinozuka, Y., Tang, Y., Weber, R. J., and Wu, Y.: Aerosol direct radiative effects over the northwest Atlantic, northwest Pacific, and North Indian Oceans: estimates based on in-situ chemical and optical measurements and chemical transport modeling, Atmos. Chem. Phys., 6, 1657-1732, doi:10.5194/acp-6-1657-2006, 2006.

Bauer, S., D. Koch, N. Unger, S. M. Metzger, D. T. Shindell, and D. G. Streets: Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone, Atmos. Chem. Phys., 7, 5043–5059, 2007

Carmichael, G. R., Adhikary, B., Kulkarni, S., D'Allura, A., Tang, Y., Streets, D., Zhang, Q., Bond, T. C., Ramanathan, V., Jamroensan, A., and Marrapu, P.: Asian aerosols: current and year 2030 distributions and implications to human health and regional climate change, Environ. Sci. Technol., 43, 5811–5817, 2009.

Chen, B., Sverdlik, L. G., Imashev, S. A., Solomon, P. A., Lantz, J., Schauer, J. J., Shafer, M. M., Artamonova, M. S., and Carmichael, G.: Empirical relationship between particulate matter

735 and aerosol optical depth over Northern Tien-Shan, Central Asia, Air Quality, Atmosphere &
736 Health, 6.2, 358-396, doi: 10.1007/s11869-012-0192-5, 2012.
737

738 Chen, B. B., Imashev, S. A., Sverdlik, L. G., Solomon, P. A., Lantz, J., Schauer, J. J., Shafer, M.
739 M., Artamonova, M. S. and Carmichael, G. R.: Ozone Variations over Central Tien-Shan in
740 Central Asia and Implications for Regional Emissions Reduction Strategies, Aerosol Air Qual
741 Res, 13, 555–562 Available from: 10.4209/aaqr.2012.06.156, 2013.

742 Chung, C. E., Ramanathan, V., Carmichael, G., Kulkarni, S., Tang, Y., Adhikary, B., Leung, L.
743 R. and Qian, Y.: Anthropogenic aerosol radiative forcing in Asia derived from regional models
744 with atmospheric and aerosol data assimilation, Atmos Chem Phys, 10(13), 6007–6024,
745 doi:10.5194/acp-10-6007-2010, 2010.

746 D'Allura, A., Kulkarni, S., 5 Carmichael, G. R., Finardi, S., Adhikary, B., Wei, C., Streets, D.,
747 Zhang, Q., Pierce, R. B., Al-Saadi, J. A., Diskin, G., Wennberg, P.: Meteorological and air
748 quality forecasting using the WRF–STEM model during the 2008 ARCTAS field campaign,
749 Atmos. Environ., 45, 6901–6910, doi:10.1016/j.atmosenv.2011.02.073, 2011.

750 Denier van der Gon, H., Visschedijk, A., Droge, R., Mulder, M., Johansson, C., and Klimont, Z.:
751 A high resolution emission inventory of particulate elemental carbon and organic carbon for
752 Europe in 2005, 7th International Conference on Air Quality – Science and Application (Air
753 Quality 2009), Istanbul, 2009.

754 Emerson, J. W., Hsu, A., Levy, M. A., de Sherbinin, A., Mara, V., Esty, D. C., and Jaiteh, M.:
755 Environmental Performance Index and Pilot Trend Environmental Performance Index, Yale
756 Center for Environmental Law and Policy, New Haven, 2012

757 Freitas, S. R., Longo, K. M., Chatfield, R., Latham, D., Silva Dias, M. A. F., Andreae, M. O.,
758 Prins, E., Santos, J. C., Gielow, R. and Carvalho Jr., J. A.: Including the sub-grid scale plume
759 rise of vegetation fires in low resolution atmospheric transport models, Atmos Chem Phys ,
760 7(13), 3385–3398, doi:10.5194/acp-7-3385-2007 , 2007.

761 Freitas, S. R., Longo, K. M., Trentmann, J. and Latham, D.: Technical Note: Sensitivity of 1-D
762 smoke plume rise models to the inclusion of environmental wind drag, Atmos Chem Phys, 10(2),
763 585–594, doi:10.5194/acp-10-585-2010, 2010.

764 Ginoux, P., Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O. and Lin, S.-J.: Sources
765 and distributions of dust aerosols simulated with the GOCART model, J Geophys Res-Atmos,
766 106(D17), 20255–20273, doi:10.1029/2000JD000053, 2001.

767 Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron
768 particles, Global Biogeochem. Cy. , 17(4), 1097, doi:10.1029/2003GB002079 , 2003.

769 Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and
770 Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmos. Environ. 39, 6957–
771 6975, 2005.

772 Grell, G., Freitas, S. R., Stuefer, M., and Fast, J.: Inclusion of biomass burning in WRF-Chem:
773 impact of wildfires on weather forecasts, Atmos. Chem. Phys., 11, 5289–5303, doi:10.5194/acp-
774 11-5289-2011, 2011.

775 Guttikunda, S., Tang, Y., Carmichael, G., Kurata, G., Pan, L., Streets, D., Woo, J.-H.,
776 Thongboonchoo, N., and Fried, A.: Impacts of Asian megacity emissions on regional air quality
777 during spring 2001, J. Geophys. Res., 110, D20301, doi:10.1029/2004JD004921, 2005.

778 Hsu, N. C., Tsay, S.-C., King, M. D. and Herman, J. R.: Aerosol properties over bright-reflecting
779 source regions, IEEE T Geosci Remote , 42(3), 557–569, doi:10.1109/TGRS.2004.824067,
780 2004.

781 Hsu, N. C., Tsay, S.-C., King, M. D. and Herman, J. R.: Deep Blue Retrievals of Asian Aerosol
782 Properties During ACE-Asia, IEEE T Geosci Remote , 44(11), 3180–3195,
783 doi:10.1109/TGRS.2006.879540, 2006.

784 Huang, M., G. Carmichael, S. Kulkarni, D. Streets, Z. Lu, Q. Zhang, B. Pierce, Y. Kondo, J.
785 Jimenez-Palacios, M. J. Cubison, B. E. Anderson, and A. Wisthaler: Sectoral and geographical
786 contributions to summertime continental United States (CONUS) black carbon spatial
787 distributions, *Atmos. Environ., 51*, 165-174, doi:10.1016/j.atmosenv.2012.01.021,2012

788 HTAP: Hemispheric Transport of Air Pollution 2010 (Executive Summary). Available online
789 from: http://www.htap.org/publications/2010_report/2010_Final_Report/EBMeeting2010.pdf
790 (Accessed 26 February 2013), 2010.

791 Hubanks, P. A., King, M. A., Platnick, S., and Pincus, R.: MODIS atmosphere L3 gridded
792 product algorithm theoretical basis document, MODIS Algorithm Teoretical Basis Document
793 No. ATBD-MOD-30 ,available at: http://modis-
794 atmos.gsfc.nasa.gov/_docs/L3_ATBD_2008_12_04.pdf,2008.

795 Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S.,
796 Berntsen, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F.,
797 Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S.,
798 Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M.,
799 Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G.,
800 Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø., Stier, P.,
801 Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. A., and Zhao, Y.: Evaluation of black
802 carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001-9026,
803 doi:10.5194/acp-9-9001-2009, 2009

804

805   Kurata, G., Carmichael, G. R., Streets, D. G., Kitada, T., Tang,Y., Woo, J. H., and
806   Thongboonchoo, N.: Relationships between emission sources and air mass characteristics in East
807   Asia during the TRACE-P period, Atmos. Environ., 38(40), 6977–6987, 2004.

808   Levy, R. C., Remer, L. A., Mattoo, S., Vermote, E. F. and Kaufman, Y. J.: Second-generation
809   operational algorithm: Retrieval of aerosol properties over land from inversion of Moderate
810   Resolution Imaging Spectroradiometer spectral reflectance, J Geophys Res-Atmos , 112(D13),
811   D13211, doi:10.1029/2006JD007811, 2007.

812   Lu, Z., Zhang, Q. and Streets, D. G.: Sulfur dioxide and primary carbonaceous aerosol emissions
813   in China and India, 1996–2010, Atmos Chem Phys, 11(18), 9839–9864, doi:10.5194/acp-11-
814   9839-2011 Available online from: http://www.atmos-chem-phys.net/11/9839/2011/), 2011.

815   Miller-Schulze, J. P., Shafer, M. M., Schauer, J. J., Solomon, P. a., Lantz, J., Artamonova, M.,
816   Chen, B., Imashev, S., Sverdlik, L., Carmichael, G. R. and Deminter, J. T.: Characteristics of
817   fine particle carbonaceous aerosol at two remote sites in Central Asia, Atmos Environ, 45(38),
818   6955–6964, doi:10.1016/j.atmosenv.2011.09.026, 2011.

819

820   Park, S., Martin M. Shafer, James J. Schauer, Paul A. Solomon, Jeffrey Lantz, Maria
821   Artamonova, Boris Chen, Sanjar Imashev, Leonid Sverdlik, Greg R. Carmichael: Chemical
822   characteristics, dynamics and provenance of mineral dust observed at two sites in Central Asia,
823   Atmos. Environ., in preparation,2014.

824   Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon,
825   Nat Geosci, 1(4), 221–227 Available online from:
826   http://www.nature.com/ngeo/journal/vaop/ncurrent/full/ngeo156.html (Accessed 18 February
827   2013), 2008.

828   Remer, L. A., Kaufman, Y. J., Tanré, D., Mattoo, S., Chu, D. A., Martins, J. V, Li, R.-R., Ichoku,
829   C., Levy, R. C., Kleidman, R. G., Eck, T. F., Vermote, E., and Holben, B.N.: The MODIS
830   aerosol algorithm products and validation, J. Atmos. Sci., 62, 947–973, doi:10.1175/JAS3385.1,
831   2005.

832   Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang,
833   X.- Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF, Version 3.
834   Technical Report NCAR/TN475+ST R. National Center for Atmospheric Research Technical
835   Note, Boulder, Colorado, 2008.

836   Shindell, D., Kuylenstierna, J. C. I., Vignati, E., van Dingenen,R., Amann, M., Klimont, Z.,
837   Anenberg, S. C., Muller, N.,Janssens-Maenhout,G., Raes, F., Schwartz, J., Faluvegi, G.,Pozzoli,
838   L, Kupiainen, K., Hoglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Hicks, K.,
839   Kim Oanh, N. T., Milly, G., Williams, M., Demkine, V., and Fowler, D: Simultaneously
840   mitigating near-term climate change and improving human health and food security, Science,
841   335, 183–189, doi:10.1126/science.1210026 , 2012.

37

842   UNEP: Framework Convention on Environmental Protection for Sustainable Development in
843   Central Asia, Regional Resource Centre for Asia and the Pacific (PRC.AP), available at:
844   http://www.rrcap.ait.asia/library/ca-report/Brief-Convention.doc, (last access: 25 April 2014),
845   2006.

846   UNEP and WMO: Integrated Assessment of Black Carbon and Tropospheric Ozone: Summary
847   for Decision Makers. available at:
848   http://www.unep.org/dewa/Portals/67/pdf/BlackCarbon_SDM.pdf (last access: 26 February
849   2013), 2011.

850   Uno, I., Satake, S., Carmichael, G. R., Tang, Y., Wang, Z., Takemura, T., Sugimoto, N.,
851   Shimizu, A., Murayama, T., Cahill, T.A., Cliff, S., Uematsu, M., Ohta, S., Quinn, P. K., and
852   Bates, T.S.: Numerical study of Asian dust transport during the springtime of 2001 simulated
853   with the Chemical Weather Forecasting System (CFORS) model, J. Geophys. Res.,
854   109(D19S24), doi:10.1029/2003JD004222, 2004.

855   V Visschedijk, A., Denier van der Gon, H., Droge, R.,van der Brugh, H.: A European high
856   resolution and size-differentiated emission inventory for elemental and organic carbon for the
857   year 2005, TNO-034-UT-2009-00688_RPT-ML, TNO, Utrecht, 2009

858   Wang, W., Bruyere, C., Duda, M., Dudhia, J., Gill, D., Lin, H.-C., Michalakes, J., Rizvi, S. and
859   Zhang, X.: Version 3 Modeling System User ' s Guide January 2011, National Center for
860   Atmospheric Research (NCAR), available at:
861   http://www.mmm.ucar.edu/wrf/users/docs/user_guide_V3.2/ARWUsersGuideV3.pdf (last
862   access: 25 April 2014), 2011.

863   Wesely, M. L. and Hicks, B. B.: A review of the current status of knowledge on dry deposition,
864   Atmos Environ, 34(12–14), 2261–2282, doi:http://dx.doi.org/10.1016/S1352-2310(99)00467-7,
865   2000.

866   Whish-Wilson, P.: The Aral Sea environmental health crisis the desiccation of the Aral Sea
867   health in the Aral Sea region, Journal of Rural and Remote Environmental Health,1(2), 29–34,
868   available at: http://www.jcu.edu.au/jrtph/vol/v01whish.pdf (last access: 25 April 2014), 2002.

869   Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. a., Orlando, J. J.
870   and Soja, a. J.: The Fire INventory from NCAR (FINN): a high resolution global model to
871   estimate the emissions from open burning, Geoscientific Model Development, 4(3), 625–641,
872   doi:10.5194/gmd-4-625-2011, 2011.

873   Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park,
874   I. S., Reddy, S., Fu, J. S., Chen, D., et al.: Asian emissions in 2006 for the NASA INTEX-B
875   mission., Atmos Chem Phys, 9(14/2), 5131–5153, doi:10.5194/acp-9-5131-2009, 2009.

876   **7. Figure Captions**

Fig. 1. WRF-STEM modeling domain set up and source region definition used in the simulations. Modeling analysis was done on the hemispheric scale. The black dotted rectangular box denotes the subset of the modeling domain used in this analysis. The triangle and circle markers denote locations of the LST and Bishkek observation sites. The anthropogenic source regions are denoted by colored regions with the A notation: A1 (N. America), A2 (Europe), A3 (Russia), A4 (Middle East), A5 (Central Asia), A6 (China) and A7 (South + Southeast Asia). The dust source regions are indicated by the magenta rectangles with the D notations: D1 (Africa), D2 (Middle East), D3 (Central Asia) and D4 (Western China). The biomass burning sources regional boundaries are indicated by the long dashed black lines with B notations: B1 (N. America i.e. region west of 25 W), B2 (Europe i.e. region west of 45E and 40 N and above), B3 (Siberia : i.e east of 45 E and 40 N and above) and B4 (South Asia i.e. below 40 N). The insert shows the topography around the observation sites.

Fig. 2. Spatial distribution of a) BC b) SO_2 c) Natural dust d) Biomass burning $PM_{2.5}$ emissions averaged over the simulation period in Gg/yr/grid.

Fig. 3. Spatial distribution of MODIS AOD averaged over the simulation period. The triangle and circle markers denote the location of LST and Bishkek sites. The numbers on the map denote contour values at sharp gradients.

Fig. 4. Spatial distribution of simulated a) BC ($\mu g/m^3$), b) Dust ($\mu g/m^3$), c) $PM_{2.5}$ ($\mu g/m^3$), and d) AOD averaged over the simulation period. The values on the map denote contour values at sharp gradients.

Fig. 5. Comparison of predicted meteorological variables from WRF model with observations shown as box and whisker plots over the simulation period (a) Temperature (K), (b) Relative Humidity RH (%), (c) Wind Speed (m/s), (d) Wind Direction (°), and (e) PBL height (m). Lidar denotes the LST (Lidar Station Teplokluchenka) site. In each box whisker panel, the middle line denotes the median value, while the edges of the box represent 25th and 75th percentile values respectively. The whiskers denote the maximum and minimum values.

Fig. 6. Comparison of predicted aerosols with observations shown as box and whisker plots over the simulation period at a) LST and b) Bishkek sites. SP and LE denote the AOD from the sun-photometer (SP) and integrated fromthe vertical extinction profiles (LE). OBS and TEOM denote filter and TEOM measurements while the MDL denotes the modeled values respectively. Lidar denotes the LST (Lidar Station Teplokluchenka) site. In each box whisker panel, the middle line denotes the median value, while the edges of the box represent 25th and 75th percentile values respectively. The whiskers denote the maximum and minimum values. The triangle marker denotes the mean value.

Fig. 7. Summary of period mean contributions by source regions and sectors for AOD, $PM_{2.5}$, dust, non-dust $PM_{2.5}$, sulfate and BC in % for the grid cells containing the Bishkek and LST observation sites, and spatially averaged over the Central Asia region. See Fig. 1 for anthropogenic, dust and fire source regions. Lidar denotes the LST (Lidar Station Teplokluchenka) site.

916 Fig. 8. Temporal variability in simulated AOD compared with observations at the LST site. The
917 box-plots of monthly values are shown. AOD from the sun-photometer (SP) and integrated from
918 the vertical extinction profiles (LE) are shown. MDL denotes modeled values. Lidar denotes the
919 LST (Lidar Station Teplokluchenka) site. In each box whisker panel, the middle line denotes the
920 median value, while the edges of the box represent 25th and 75th percentile values respectively.
921 The whiskers denote the maximum and minimum values.

922 Fig. 9. Comparison of simulated PM mass with filter-based observations at Central Asia sites (a)
923 $PM_{2.5}$ (LST) along with TEOM non-volatile measurements, (b) $PM_{2.5}$ (Bishkek), (c) PM_{10} (LST),
924 and (d) PM_{10} (Bishkek) in ($\mu g/m^3$). Lidar denotes the LST (Lidar Station Teplokluchenka) site.

925 Fig. 10. Temporal variability in simulated (a) Temperature (K) and (b) Relative Humidity RH
926 (%) from WRF model compared with observations at the LST and Bishkek sites. Lidar denotes
927 the LST (Lidar Station Teplokluchenka) site.

928 Fig. 11. Simulated composition of $PM_{2.5}$ at the LST site by a) Species ($\mu g/m^3$), b) Fine dust
929 source regions($\mu g/m^3$) c) Anthropogenic $PM_{2.5}$ source regions ($\mu g/m^3$) d) Anthropogenic $PM_{2.5}$
930 source ($\mu g/m^3$) e) Biomass $PM_{2.5}$ source regions($\mu g/m^3$).The contributions from source region
931 and sectors denote the non – dust portion of $PM_{2.5}$ mass.

932 Fig. 12. Simulated composition of BC at the LST site by a) Source regions ($\mu g/m^3$), b) Source
933 sector ($\mu g/m^3$), and c) Biomass burning source regions ($\mu g/m^3$).

934 Fig. 13. Time altitude cross sections of weekly averaged predicted BC, dust and $PM_{2.5}$ for the
935 simulation period at the LST site. The numbers denote the specific episodes of aerosol
936 enhancements for back trajectory analysis in Fig. 14 and Fig. 15. Lidar denotes the LST (Lidar
937 Station Teplokluchenka) site.

938 Fig. 14. Ten day air mass back trajectories for August 2008 (events 1 and 2 as denoted in Fig.
939 13) color coded by source regions. The blue diamond and green square hatched areas denote the
940 natural dust and biomass burning emission sources while the number in black denote MODIS
941 AOD contours averaged over the event time period and ten day prior time window. The
942 trajectories are color coded by source regions including Africa (blue), Middle East (green),
943 Central Asia (yellow), North Asia biomass (> 50° N, black), Europe (brown), China (red) and
944 South Asia (orange). Lidar denotes the LST (Lidar Station Teplokluchenka) site.

945 Fig. 15. Same as Fig. 14 but for November 2008, January 2009 and April 2009 (events 3, 4, 5
946 denoted in Fig. 13).

947 Fig. 16 Ten day air mass forward trajectories illustrating the seasonality in transport pathways
948 out of CA for a) June 2008, b) December 2008, and c) April 2009. The blue diamond, green
949 square hatched areas denote the natural dust and biomass burning emission sources while the
950 number in black denote MODIS AOD contours, respectively, averaged over the event time
951 period and subsequent ten day time window. Lidar denotes the LST (Lidar Station
952 Teplokluchenka) site.

953 Fig. 17. Seasonally averaged surface total dust (PM_{10}) concentrations from Central Asia dust
954 emissions. DJF (top left panel) denotes the average for the months of December, January and

955 February. MAM (top right panel) denotes the average for months of March, April and May. JJA
956 (bottom left panel) denotes the average for months of June, July and August while SON (bottom
957 right panel) denotes average for months of September, October and November.

958 Fig. 18. Time series of predicted surface concentration at 6 h time step (top panel) and weekly
959 averaged time altitude cross sections (bottom panel) of total dust (PM_{10}) from Central Asia dust
960 emissions at Mt. Bachelor, Oregon.

961 Fig. 19. Percent change in simulated period mean surface BC and $PM_{2.5}$ concentrations for future
962 2030 emission scenarios relative to the base year (2005) a),d) reference 2030, a),e) BC measures
963 (low) and c),f) BC (lowest) and greenhouse gas measures aimed at keeping CO_2 levels below
964 450 ppm. Refer to Sec. 2.3 for more details on emission scenarios.

965
966 **8. Supplemental Materials Figure and Table Captions**

967 Table S1. Comparison of the observed and predicted surface meteorology at the observation
968 sites.

969 Table S2. Comparison of observations and model of AOD, $PM_{2.5,}$ PM_{10}, BC and OC at the
970 Bishkek site.

971 Table S3. Comparison of observations and model of AOD, $PM_{2.5,}$ PM_{10}, BC and OC at the LST
972 site.

973

974 Fig. S1. Seasonal variability in spatial distribution of biomass burning $PM_{2.5}$ emissions in
975 Gg/month/grid (a) April 2008, (b) August 2008, (c) November 2008, and (d) May 2009.

976 Fig. S2. Spatial distribution of a) base year 2005 BC emissions (Gg/yr/grid) along with percent
977 change (w.r.t to base year 2005) in b) Reference 2030 BC emissions c) 2030 BC emissions with
978 BC measures (low) and d) 2030 BC emissions with BC (lowest) and greenhouse gas measures
979 aimed at keeping CO_2 levels below 450ppm. The triangle and circle markers denote locations of
980 the LST and Bishkek sites. Refer to Sec. 2.3 for more details on emission scenarios.

981 Fig. S3. Spatial distribution of a) base year 2005 $PM_{2.5}$ emissions (Gg/yr/grid) along with percent
982 change (w.r.t to base year 2005) in b) Reference 2030 $PM_{2.5}$ emissions c) 2030 $PM_{2.5}$ emissions
983 with BC measures and d) 2030 $PM_{2.5}$ emissions with BC and greenhouse gas measures aimed at
984 keeping CO_2 levels below 450ppm. The triangle and circle markers denote locations of the LST
985 and Bishkek sites. Refer to Sec. 2.3 for more details on emission scenarios.

986 Fig. S4. Spatial distribution of a) base year 2005 SO_2 emissions (Gg/yr/grid) along with percent
987 change (w.r.t to base year 2005) in (b) Reference 2030 SO_2emissions c) 2030 SO_2 emissions with
988 BC measures and d) 2030 SO_2 emissions with BC and greenhouse gas measures aimed at
989 keeping CO_2 levels below 450ppm. The triangle and circle markers denote locations of the LST
990 and Bishkek sites. Refer to Sec. 2.3 for more details on emission scenarios.

991 Fig. S5. Comparison of observed and predicted PBL heights (m) at the LST site. Observed PBL
992 heights were determined from the Lidar profiles.

993 Fig. S6. Spatial distribution of predicted species contributions (%) to AOD averaged over the
994 simulation period a) Carbonaceous aerosols (BC+OC), b) SO4, c) Other PM, and d) Dust.

995 Fig. S7. Spatial distribution of predicted species contributions (%) to $PM_{2.5}$ averaged over the
996 simulation period a) Carbonaceous aerosols (BC+OC), b) SO4, c) Other $PM_{2.5}$, and d) Dust.

997 Fig. S8. Seasonal variability in spatial distribution of MODIS and simulated AOD averaged over
998 the simulation period.

999 Fig S9. Summary of period mean contributions by source regions and sectors for OC in % for the
1000 grid cells containing the Bishkek and LST observation sites, and spatially averaged over the
1001 Central Asia region. See Fig. 1 for anthropogenic and fire source regions.

1002 Table:1 Summary of changes in Central Asia (CA) region (See figure 1 for CA region definition)
1003 emissions and concentration under the future emission scenarios (See Sect. 3.8 for more details)
1004

Emission Scenario	Emissions Gg yr^{-1}			Emissions change (%) w.r.t to base 2005		
	BC	SO$_2$	PM$_{25}$	BC	SO$_2$	PM$_{25}$
Base 2005	39	1130	197			
Reference 2030	47	1326	224	22.1	17.3	13.9
Low GWP 2030	27	1325	177	-31.1	17.3	-10.1
Lowest GWP + 450	21	1058	115	-46.9	-6.4	-41.5

	Concentration (μg m^{-3})			Concentration change (%) w.r.t to base 2005		
	BC	SO$_4$	PM$_{25}$	BC	SO$_4$	PM$_{2.5}$
Base 2005	0.17	3.04	4.26			
Reference 2030	0.16	2.65	4.03	-4.2	-12.6	-5.3
Low GWP 2030	0.11	2.64	3.85	-36.6	-13	-9.5
Lowest GWP + 450	0.10	2.04	3.06	-38.5	-32.9	-28

1005

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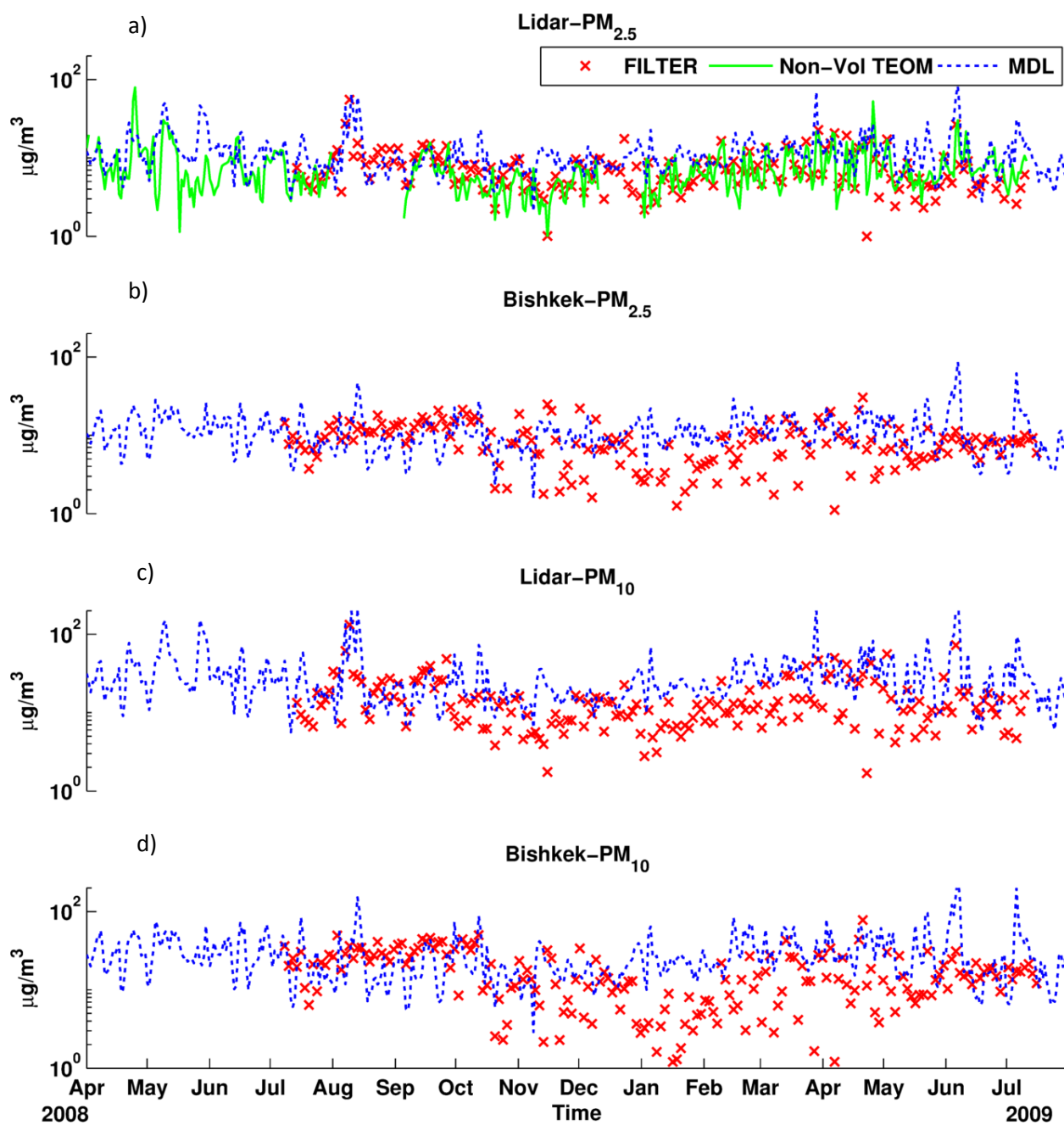
a)Lidar Concentration Boxplots

b)Bishkek Concentration Boxplots

Fig. 6. Comparison of predicted aerosols with observations shown as box and whisker plots over the simulation period at a) LST and b) Bishkek sites.SP and LE denote the AOD from the sun-photometer (SP) and integrated from the vertical extinction profiles (LE). OBS and TEOM denote filter and TEOM measurements while the MDL denotes the modeled values respectively. Lidar denotes the LST (Lidar Station Teplokluchenka) site. In each box whisker panel, the middle line denotes the median value, while the edges of the

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Fig. 7. Summary of period mean contributions by source regions and sectors for AOD, PM$_{2.5}$, dust, non-dust PM$_{2.5}$, sulfate and BC in % for the grid cells containing the Bishkek and LST observation sites, and spatially averaged over the Central Asia region. See Fig. 1 for anthropogenic, dust and fire source regions. Lidar denotes the LST (Lidar Station Teplokluchenka) site.

Lidar AOD Boxplots

Fig. 8. Temporal variability in simulated AOD compared with observations at the LST site. The box-plots of monthly values are shown. AOD from the sun-photometer (SP) and integrated from the vertical extinction profiles (LE) are shown. MDL denotes modeled values. Lidar denotes the LST (Lidar Station Teplokluchenka) site. In each box whisker panel, the middle line denotes the median value, while the edges of the box represent 25th and 75th percentile values respectively. The whiskers denote the maximum and minimum values.

Fig. 9. Comparison of simulated PM mass with filter-based observations at Central Asia sites (a) $PM_{2.5}$ (LST) along with TEOM non-volatile measurements, (b) $PM_{2.5}$ (Bishkek), (c) PM_{10} (LST), and (d) PM_{10} (Bishkek) in ($\mu g/m^3$). Lidar denotes the LST (Lidar Station Teplokluchenka) site.
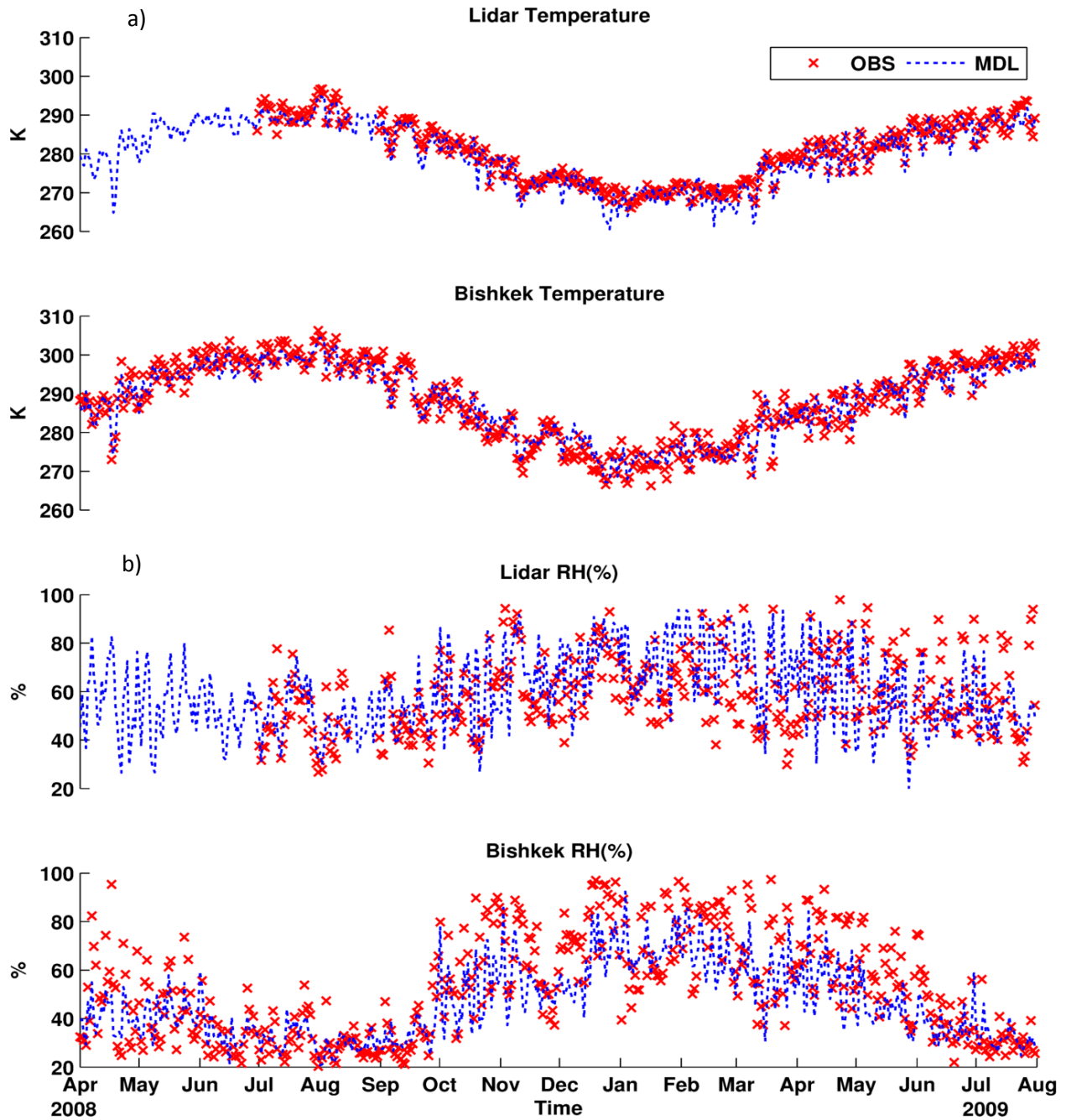
Fig. 10. Temporal variability in simulated Temperature (K) and (b) Relative Humidity RH (%) from WRF model compared with observations at the LST and Bishkek sites. Lidar denotes the LST (Lidar Station Teplokluchenka) site.
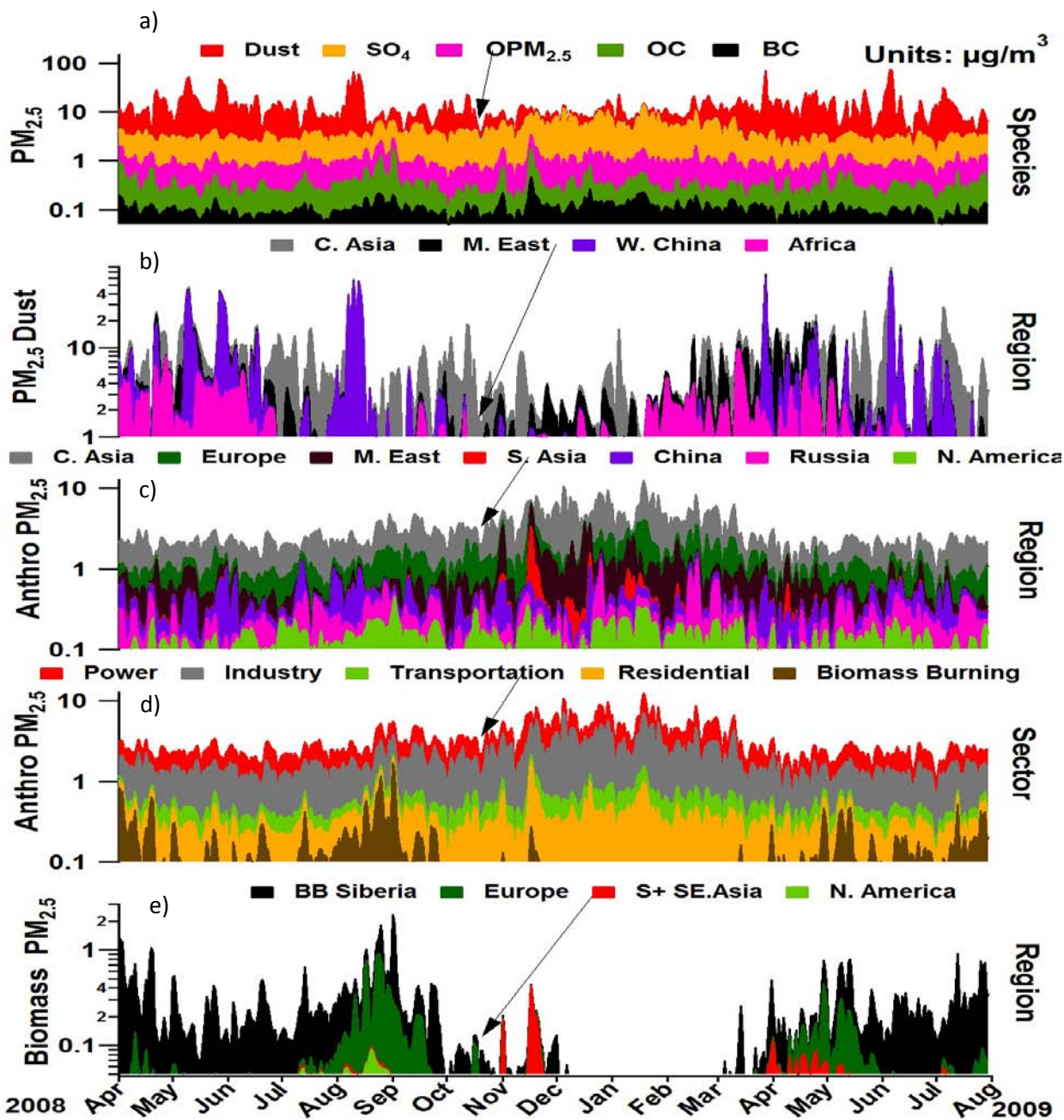
Fig. 11. Simulated composition of $PM_{2.5}$ at the LST site by a) Species ($\mu g/m^3$), b) Source sector ($\mu g/m^3$), and c) Source regions ($\mu g/m^3$). The contributions from source region and sectors denote the non – dust portion of $PM_{2.5}$ mass.

Fig. 12. Simulated composition of BC at the LST site by a) Source regions (µg/m³), b) Source sector (µg/m³), and c) Biomass burning source regions (µg/m³).

Time Altitude Curtains at Lidar
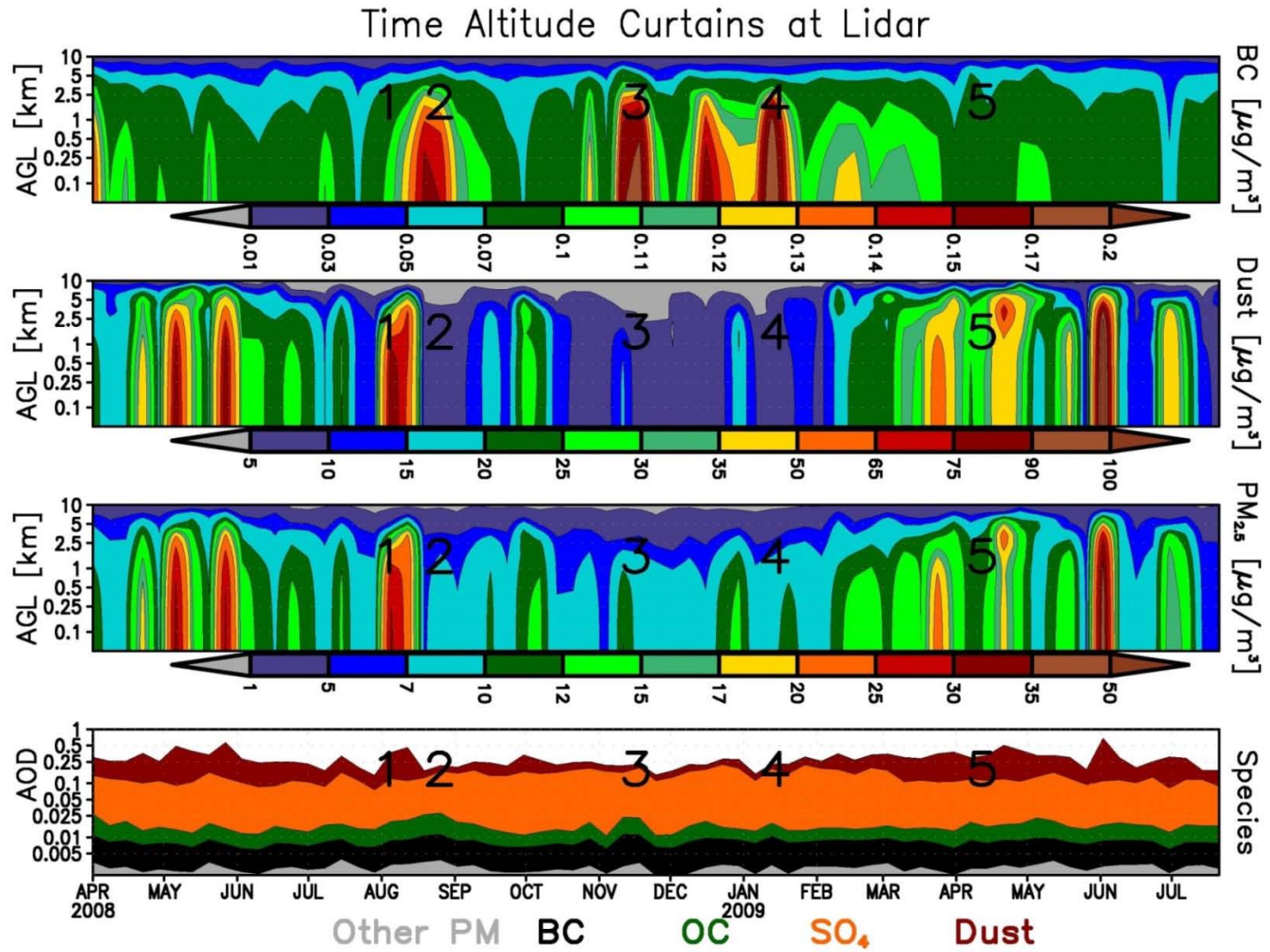
Fig. 13. Time altitude cross sections of weekly averaged predicted BC, dust and $PM_{2.5}$ for the simulation period at the LST site. The numbers denote the specific episodes of aerosol enhancements for back trajectory analysis in Fig. 14 and Fig. 15. Lidar denotes the LST (Lidar Station Teplokluchenka) site.
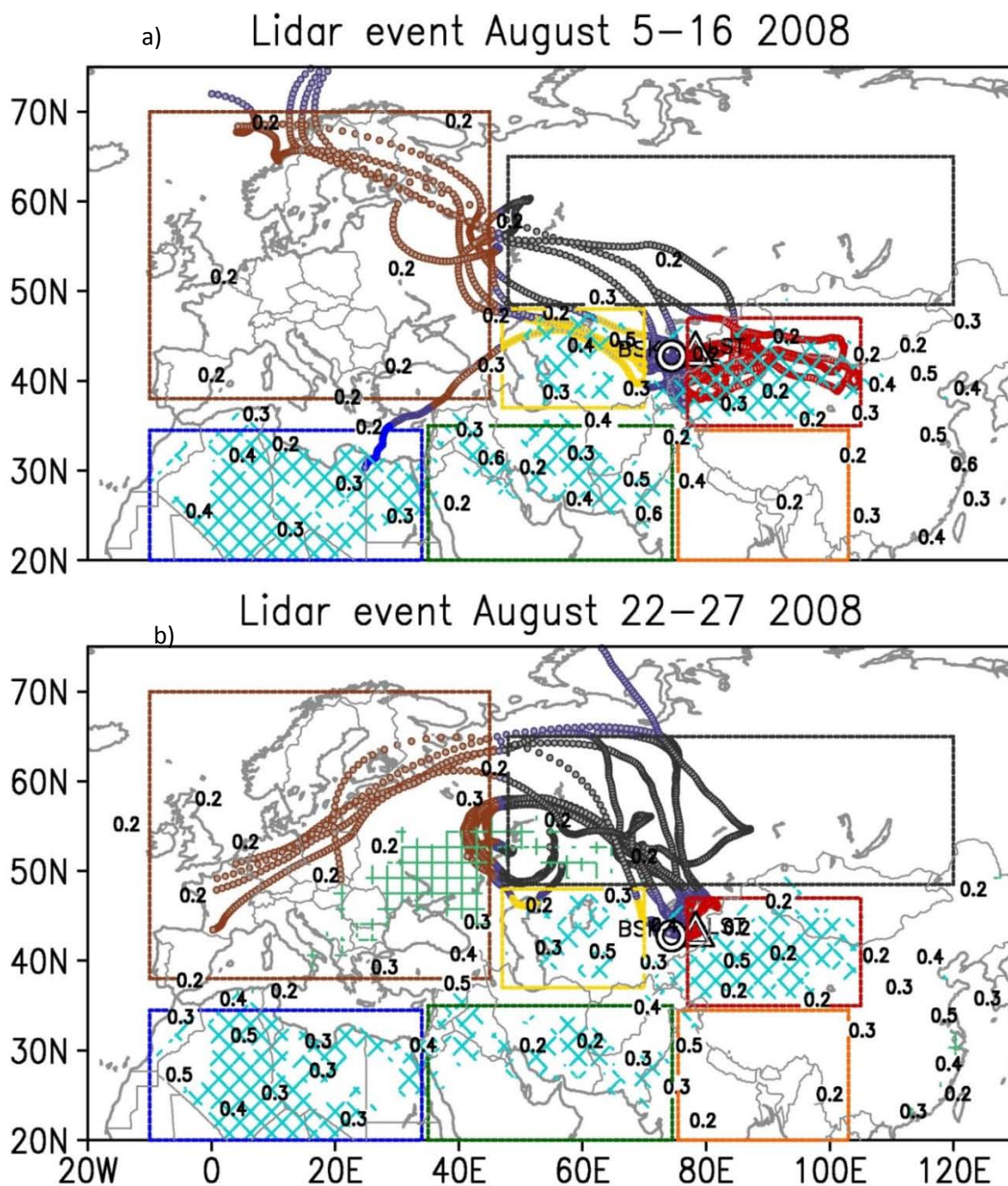
Fig. 14. Ten day air mass back trajectories for August 2008 (events 1 and 2 as denoted in Fig. 11) color coded by source regions. The blue diamond and green square hatched areas denote the natural dust and biomass burning emission sources while the number in black denote MODIS AOD contours averaged over the event time period and ten day prior time window. The trajectories are color coded by source regions including Africa (blue), Middle East (green), Central Asia (yellow), North Asia biomass (> 50° N, black), Europe (brown), China (red) and South Asia (orange). (green), Central Asia (yellow), North Asia biomass (> 50° N,

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black), Europe (brown), China (red) and South Asia (orange). Lidar denotes the LST (Lidar Station Teplokluchenka) site.
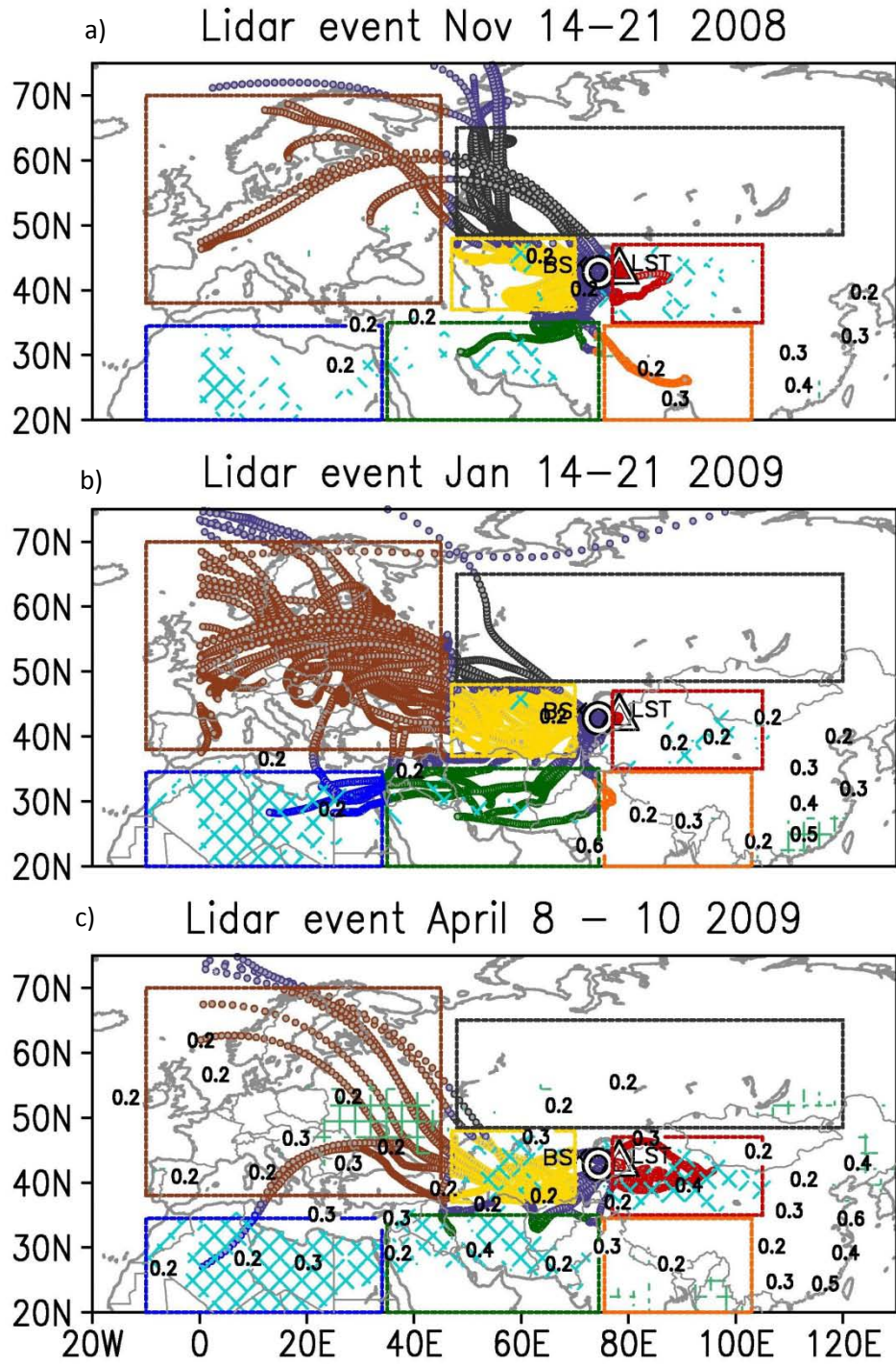
Fig. 15. Same as Fig. 14 but for November 2008, January 2009 and April 2009 (events 3, 4, 5 denoted in Fig. 13).
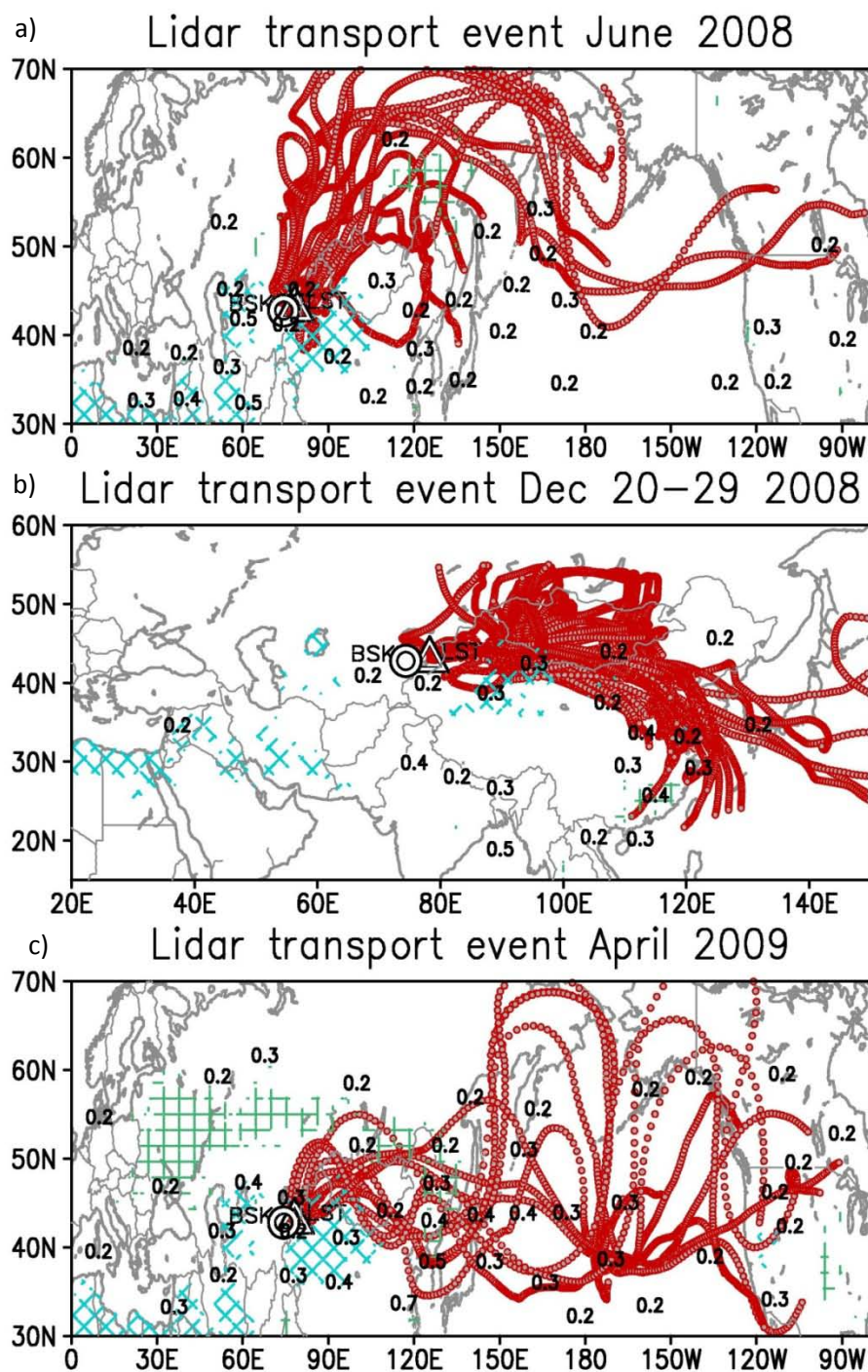
Fig. 16 Ten day air mass forward trajectories illustrating the seasonality in transport pathways out of CA for a) June 2008, b) December 2008, and c) April 2009. The blue diamond, green square hatched areas denote the natural dust and biomass burning emission sources while the number in black denote MODIS AOD contours, respectively, averaged over the event time

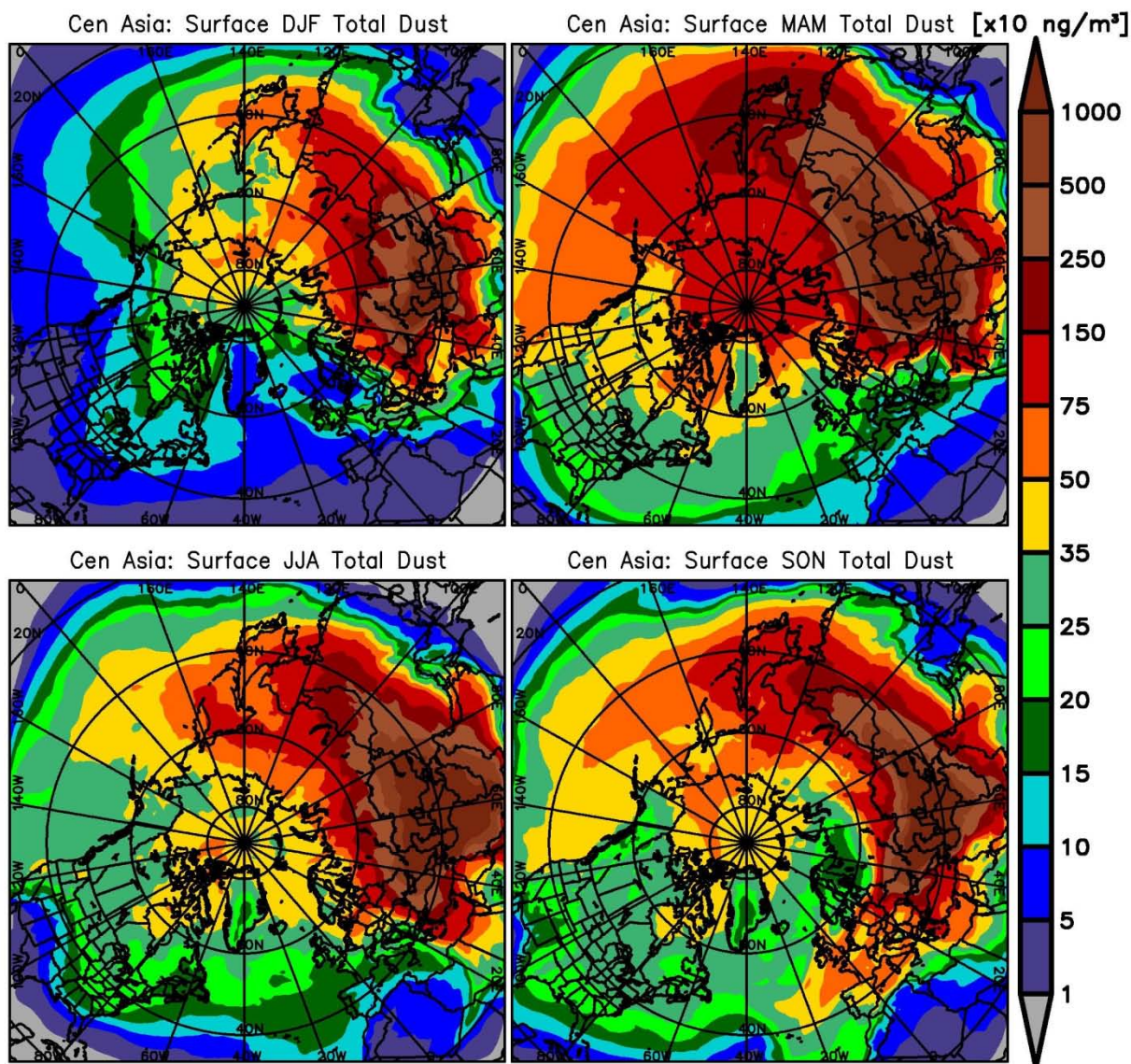period and subsequent ten day time window. Lidar denotes the LST (Lidar Station Teplokluchenka) site.

Fig. 17. Seasonally averaged surface total dust (PM_{10}) concentrations from Central Asia dust emissions. DJF (top left panel) denotes the average for the months of December, January and February. MAM (top right panel) denotes the average for months of March, April and May. JJA (bottom left panel) denotes the average for months of June, July and August while SON (bottom right panel) denotes average for months of September, October and November.
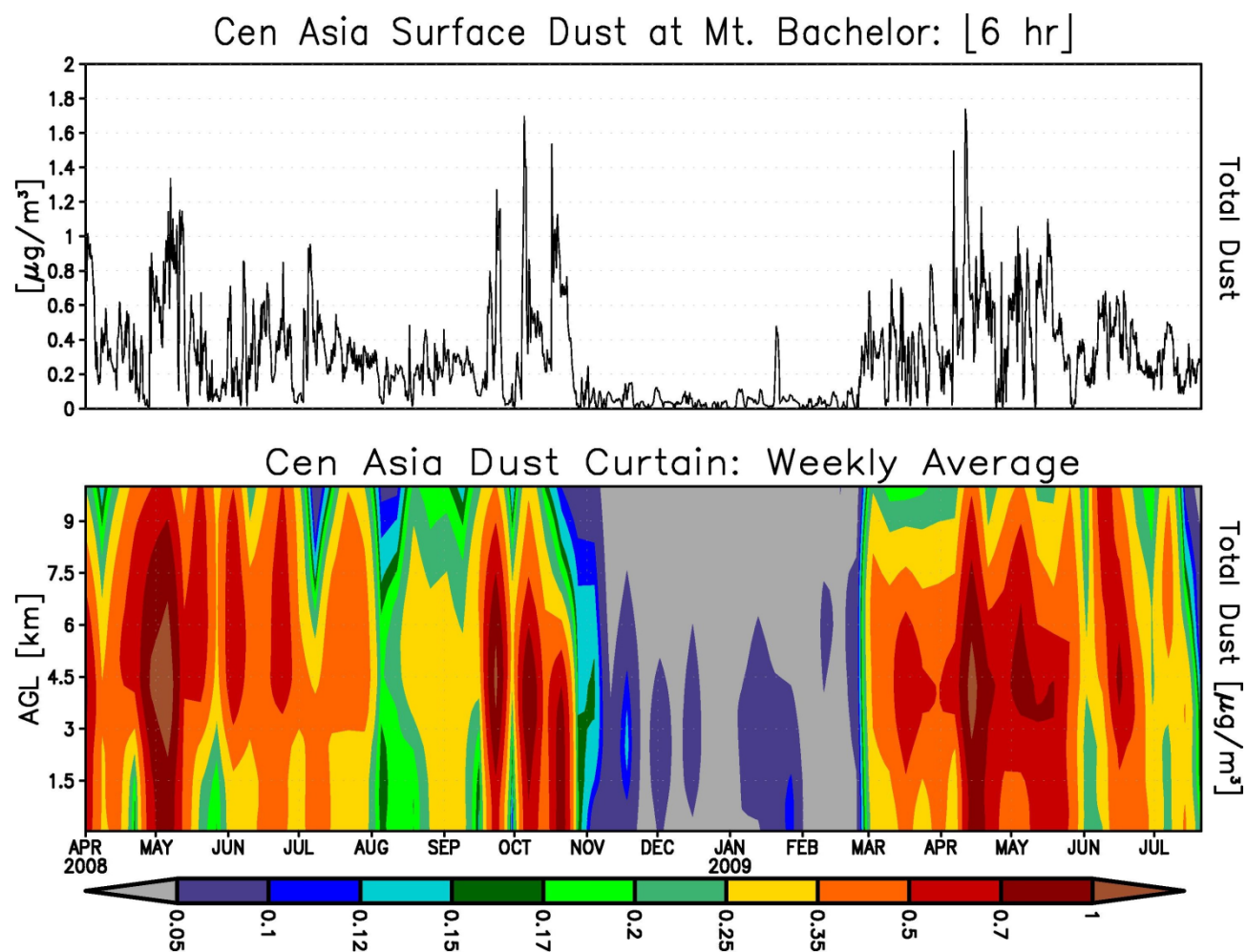
Fig. 18. Time series of predicted surface concentration at 6 h time step (top panel) and weekly averaged time altitude cross sections (bottom panel) of total dust (PM$_{10}$) from Central Asia dust emissions at Mt. Bachelor, Oregon.
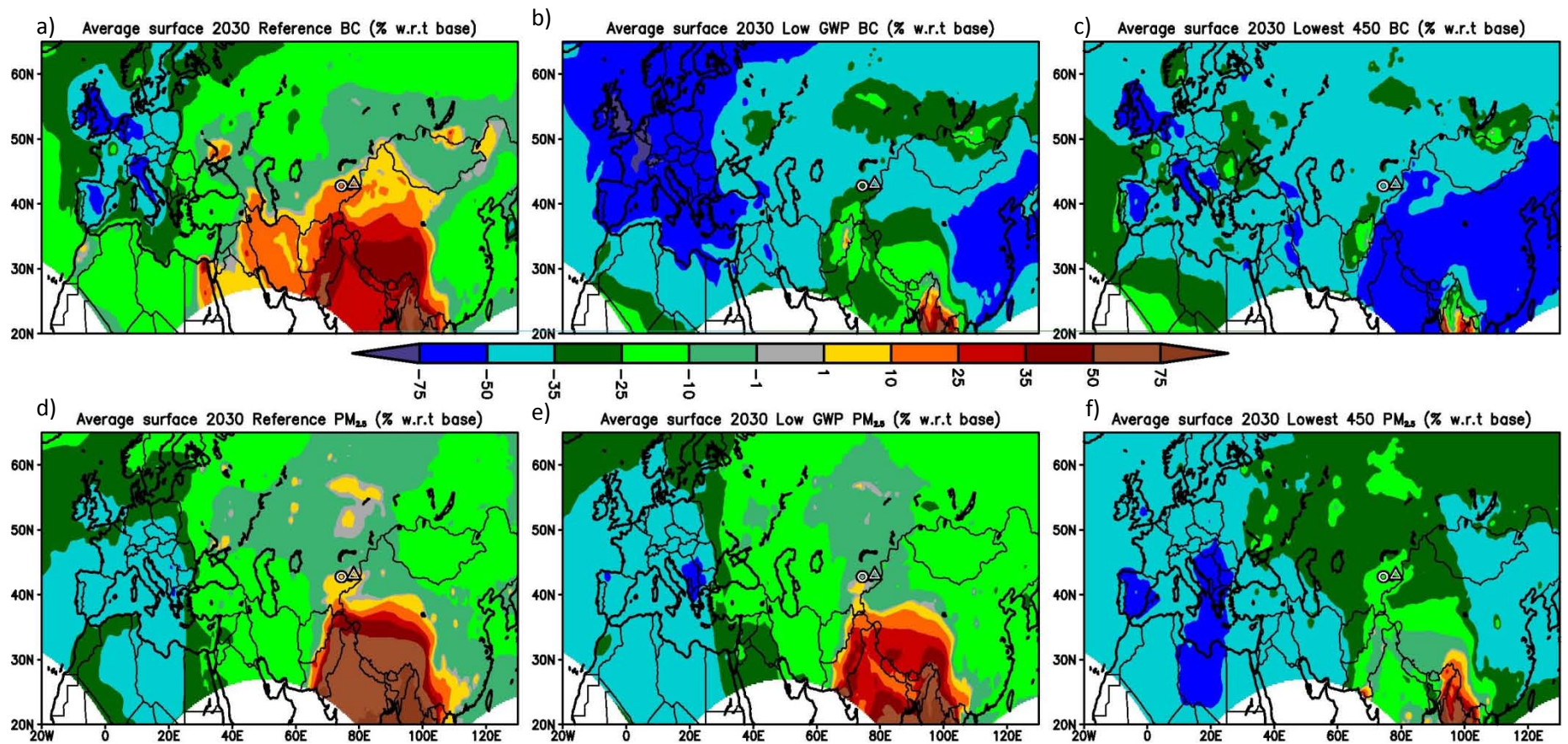
Fig. 19. Percent change in simulated period mean surface BC and $PM_{2.5}$ concentrations for future 2030 emission scenarios relative to the base year (2005) a),d) reference 2030, a),e) BC measures (low) and c),f) BC (lowest) and greenhouse gas measures aimed at keeping CO_2 levels below 450 ppm. Refer to Sec. 2.3 for more details on emission scenarios.