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 $\mu g/m^3$) 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 $\mu g/m^3$). Surface concentrations of black carbon (BC) (mean value ~0.1 $\mu g/m^3$) 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

composition in CA demonstrate that this region is strategically located to characterize regional and intercontinental transport of pollutants. Aerosols at these sites are shown to reflect dust, biomass burning and anthropogenic sources from Europe, South, East and CA, and Russia depending on the time period.

Simulations for a reference 2030 emission scenario based on pollution abatement measures already committed to in current legislation show that $PM_{2.5}$ and BC concentrations in the region increase, with BC growing more than $PM_{2.5}$ on a relative basis. This suggests that both the health impacts and the climate warming associated with these particles may increase over the next decades unless additional control measures are taken. The importance of observations in CA to 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

undertaken between Russia, Kyrgyz Republic, and USA scientists to observe and model aerosols in the region. Measurements of particulate matter (PM) mass and composition were taken at two locations in the Kyrgyz Republic (Lidar Station Teplokluchenka (LST) and Bishkek) and modeling analysis was performed to assess the contributions of local, regional and distant sources to the PM concentrations in the region (Miller-Schulze et al., 2012, Chen et al., 2012, 2013).

In this paper we present a modeling analysis of $PM_{2.5}$, PM_{10}, ($PM_{2.5}$ refers to particles in the size range of less than 2.5 μm aerodynamic diameter (AD) and PM_{10} refers to particles in the size range of less than 10 μm AD), black carbon (BC) and organic carbon (OC) mass concentrations and aerosol optical depth (AOD) over the time period of April 2008 to July 2009. The Sulfur Transport and dEpostion Model (STEM), a hemispheric chemical transport model (D'Allura et al., 2011), is used to estimate spatial and temporal variations in PM in CA, and to assess the contributions to PM from wind-blown dust, open biomass burning, and anthropogenic sources, and different geographical source regions and source sectors (transportation, power, industry and residential). The simulated values are compared to surface measurements of AOD, $PM_{2.5}$, PM_{10}, BC, OC mass concentrations at the two regional sites in CA. The transport of aerosols into CA is also explored through three dimensional backward trajectory analysis. Transport from CA and their impacts on downwind areas are also analyzed via forward trajectory analysis. Finally we present results of how the PM concentrations may change in the future using emission scenarios for 2030 that reflect possible air quality and climate policies.

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).

In addition, the model prediction skill in simulating BC is evaluated using the BC measurements from the IMPROVE (Interagency Monitoring of Protected Visual Environments) surface site network (http://views.cira.colostate.edu/web/DataWizard/)

The Moderate Resolution Imaging Spectroradiometer (MODIS) collection 5.1 Level 2 AOD products (~ 10 km horizontal resolution) at 550 nm wavelength from Terra and Aqua satellites were used to compare the observed and simulated AOD. The MODIS Level 2 data were used and included land and ocean AOD retrieved via the dark target algorithm (Remer et al., 2005, Levy et al., 2007), and the Deep Blue AOD over land (Hsu et al., 2004, Hsu et al., 2006), which can retrieve AOD over bright and desert surfaces. This is particularly relevant for the CA region, which contains major dust sources in western China, South Asia, the Middle East, and Africa (Ginoux et al., 2001). The MODIS Level 2 to Level 3 averaging procedure outlined in Hubanks et al., (2008) was employed to interpolate the 10 km Level 2 AOD products to the 60 km horizontal model resolution on a daily basis. Level 2 QA flag weightings were used to reduce the uncertainty associated with the MODIS retrievals. In grid cells where both the MODIS dark target and Deep Blue AOD were retrieved, the mean value of the two was used.

2.2. Modeling System

2.2.1 Meteorological Model

The Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) version 3.2 was used to generate the meteorological fields needed for simulating the transport patterns in the STEM model (D'Allura et al., 2011). The WRF simulations for each day were initialized using the meteorological boundary conditions obtained from National Centers for Environmental

153 Prediction (NCEP) Final Analysis (FNL, http://rda.ucar.edu/datasets/ds083.2/). A daily 24 hour
154 spin up time for WRF was used (i.e. WRF was run for 48 hours each day and the first 24 hours
155 were treated as spin up and were discarded). The STEM model simulation time period was from
156 April 2008 to July 2009.

157 2.2.2 Chemical Transport Model

158 The STEM model (Carmichael et al., 2009) was used to simulate the mass of sulfate, BC,
159 OC, other primary emitted $PM_{2.5}$, and other primary emitted PM_{10} (i.e., non-carbonaceous PM
160 such as fly ash, road dust, and cement), which were simulated as a single mass component with
161 aerodynamic diameters less than or equal 2.5 μm, and between 2.5 and 10 μm (denoted as coarse
162 fraction), respectively and referred to in this paper as $OPM_{2.5}$ and OPM_{10}, dust (fine and coarse)
163 and sea salt (fine and coarse). Nitrate and secondary organic aerosols (SOA) were not included in
164 the model for this application. The nitrate aerosol is estimated to be a minor component of the
165 PM mass in CA (Baurer et al., 2007). The importance of SOA will be discussed later in the
166 paper. The dry deposition of aerosols was modeled using the "Resistance in Series
167 Parameterization" (Wesely and Hicks, 2000) and wet deposition was calculated as a loss rate
168 based on the hourly precipitation calculated from the WRF model. Further details of the wet
169 scavenging can be found in Adhikary et al., (2007). The modeled AOD at 550 nm wavelength
170 was calculated using the simulated three dimensional aerosol distributions and species specific
171 extinction coefficients as described in Chung et al., (2010).

172 2.2.3 Modeling Domain

The STEM and WRF computation domains were identical, with a 60 ×60 km horizontal resolution (249 ×249 horizontal grid cells) and 22 vertical layers up to 10 hPa. The domain (Fig. 1) covered much of the northern hemisphere in a polar stereographic projection, centered over the Arctic region and extended to 35° N to include the major emission regions of North America, Europe, and Asia. This modeling system has been applied to simulate aerosol distributions for ARCTAS field campaign as described in D'Allura et al., (2011) and further details describing the model can be found there. STEM was initialized with a one month spin up using March 2008. Much of the analysis for this paper is focused on the domain denoted by the rectangle centered over CA shown in Fig. 1. This domain has large gradients in topography (insert Fig. 1), which significantly impact the transport patterns in the region.

2.2.4 Air Mass Trajectories

The CA observation sites are impacted by dust, anthropogenic pollution, and biomass burning emissions from various source regions. To further understand the transport pathways and source region influences on the PM distributions at these sites, three dimensional ten day air mass trajectories (both forward and backward in time) from each site were calculated for the entire time period (April 2008 – July 2009). In this trajectory analysis, we utilized the three dimensional wind fields (including u, v and w components) along with the above ground level (AGL) altitude simulated by the WRF meteorological model consistent with Dallura et al., 2011 study. These trajectories describe the general flow patterns based on wind fields alone and provide useful information about the history of air mass particularly the influence of source regions over which the air mass had resided before arriving at the site of interest. Note that these

trajectories do not account for any other atmospheric processes such as diffusion or chemical evolution along its path (Kurata et al, 2004 and Guttikunda et al., 2005).

To understand the differences in transport patterns at the surface and aloft, and to study the impact of topographic gradients in the vicinity of the sites, trajectories were initialized at different altitudes (0.1 (100m), 0.3 (300m) 0.5 (500m), 1, 2, 3, and 5 km) a.g.l at the site locations (i.e. latitude and longitude) daily every 3 hours for a ten day period both backward and forward in time. The trajectories were terminated when they touched the ground, or went out of the model domain or exceeded the ten day calculation period. The trajectories (at or below 1 km) were used to characterize transport pathways impacting the surface concentrations at these sites, which are discussed later in section 3.5.

2.3. Emissions

2.3.1 Base emissions

Anthropogenic emissions of BC, OC, $PM_{2.5}$, PM_{10}, and SO_2 were based on the ARCTAS emissions described in D'Allura et al., (2011), but updated with newly available information. For India and China BC, OC, and SO_2 emissions from Lu et al., (2011) were utilized, and INTEX-B emissions were used for the rest of Asia (Zhang et al., 2009). For Europe, the EMEP 2008 (http://www.ceip.at/webdab-emission-database/officially-reported-emission-data) emissions were used for SO_2, $PM_{2.5}$, and PM_{10}, and the EUCAARI 2005 inventory was used for the carbonaceous particles (BC and OC) (Visschedijk et al. 2009 and Denier van der Gon et al. 2009). The shipping emissions came from the IIASA base year 2005 inventory (UNEP and WMO, 2011). Mass conservative regrinding tools including MTXCALC and MTXCPLE from

220 the IOAPI m3tools suite (http://www.baronams.com/products/ioapi/AA.html#tools) were used to
221 interpolate the input raw emissions described above on to the model grid.

222 Anthropogenic emissions for SO_2, BC and OC were available by major economic sectors;
223 i.e., transportation, residential, industry, and power. The industry and power sectors were treated
224 as small and large point sources, respectively, and emitted into the first 6 model levels (lowest 2
225 km). The residential and transportation emissions were treated as near surface area sources and
226 partitioned into the first two model levels from surface to ~ 100m (i.e. AGL altitude of level 2)
227 with a 90 – 10 percent split. Monthly emission allocation factors were applied over India and
228 China for the economic sectors from Lu et al., (2011). The rest of the domain (i.e. excluding
229 India and China) used same emission rates for all months due to unavailability of monthly
230 emission allocation factors.

231 The Fire Inventory from NCAR (FINN v1) was used for BC, OC, CO, SO_2, $PM_{2.5}$ and
232 PM_{10} biomass burning emissions from forest, grassland and crop residual fires. The FINN
233 database, which is based on MODIS fire detection as thermal anomalies, provides global
234 coverage of fire emissions at a spatial resolution of ~ 1km on a daily timescale (Wiedinmyer et
235 al., 2011). The WRF-Chem fire utility (http://bai.acd.ucar.edu/Data/fire/) was employed to
236 interpolate the speciated FINN emissions to the WRF model grid. The gridded two-dimensional
237 FINN emissions were used as input to the WRF-Chem (Grell et al., 2005) plume rise model
238 (Grell et al., 2011), which implements the Freitas et al., (2007) and Freitas et al., (2010)
239 algorithm to compute injection heights and to calculate the vertical distribution of fire emissions
240 at an hourly time step, which were further utilized as input to STEM model simulations.

241 Sea salt and dust emissions were calculated using the WRF meteorological fields based
242 on the methods described in Gong, (2003) and Uno et al., (2004), respectively. The dust
243 emissions were further constrained with snow cover (SNOWC variable from WRF output) and
244 only grid cells with snow cover < 1% were used for dust emission calculations.

245 Fig. 2 shows the annual gridded anthropogenic SO_2 and BC, dust, and biomass burning
246 $PM_{2.5}$ emissions in Gg per grid in and around CA. Large BC emission hotspots can be seen over
247 the Indo-Gangetic plain and eastern China. Significant BC emissions are also seen over Europe,
248 but are relatively lower in intensity than the Asian sources. The SO_2 emissions show Eastern
249 China as the largest source region followed by regions of South Asia, Europe, and Russia. The
250 major natural dust emission sources (Fig. 2c) include Africa, the Middle East, CA, Western India
251 boundaries, and Western China. The major sources of biomass burning are Eastern Europe,
252 portions of Siberian (between 40 – 60° N), Southeast Asia, Southern China and India (Fig. 2d).

253 Dust emissions have a strong seasonal cycle. The major dust sources in the region (Fig.
254 1) are located to the east, west and south of the observation sites and include the cool winter
255 deserts around the Aral and Caspian seas and those in western China and northern Pakistan as
256 well as the sub-tropical deserts in western India, around the Persian Gulf and northern Africa.
257 The emissions from the cool winter deserts occur when the surfaces are free of snow cover (from
258 March through October). Emissions from the sub-tropical deserts can occur throughout the year.

259 The open biomass burning emissions that impact CA also have a strong seasonality with
260 minimum impact in winter (Supplemental Materials Fig. S1). Fires typically begin in the spring
261 in Siberia along 50° N latitude and in northern India and South East Asia and in summer the high
262 latitude burning shifts to the west. In October the fire activity decreases and remains low until

263 spring, with the most active fire regions associated with agricultural burning in northern India
264 and southeast China.

265 **2.3.2 Future Emissions Scenarios**

266 In addition to the base emissions, a series of simulations were analyzed using emission
267 scenarios for 2030. These scenarios were developed for the WMO/UNEP report that looked at
268 short lived climate pollutants as described in Shindell et al., (2012) and Anenberg et al., (2012).
269 The reference scenario for 2030 was based on the implementation of control measures currently
270 approved in the various regions and assumed their perfect implementation. The 2030 reference
271 scenarios were developed from a reference global emissions inventory with a 2005 reference
272 year, and assumed significant growth in fossil fuel use relative to 2005, leading to increases in
273 estimated CO_2 emissions (45%). Abatement measures prescribed in current legislation were
274 projected to lead to reductions in air pollutant emissions, which varied by pollutant and region.
275 In the 2030 reference scenario, total primary $PM_{2.5}$ emissions remain approximately constant,
276 while BC and OC decline by a few percent. However, in the study domain emission changes
277 varied widely. BC emissions increased by 10 – 100% in CA, South and Southeast Asia and in
278 western China, and decreased in East Asia and Europe. The $PM_{2.5}$ emissions showed similar
279 regional changes but grew at smaller rates (10 – 40%). SO_2 emissions generally increased
280 throughout the region by 10 – 20%. Spatial maps of emission changes for the 2030 reference
281 scenario are presented in Figs. S2b, S3b, and S4b.

282 A series of emission control scenarios for 2030 were developed to evaluate the impact of
283 additional abatement measures designed to reduce the levels of short lived climate pollutants
284 (e.g., BC). The BC measures in the scenarios included two different sets of assumptions (low and

285 lowest). The first focused on reductions from incomplete combustion sources. These included
286 implementation of Euro 6 equivalent vehicle emission standards (requiring installation of diesel
287 particulate filters) and improving traditional biomass cook stoves in developing countries
288 (assuming 25% decrease in BC and 80 – 90% decreases in OC, CO, non-methane volatile
289 organic compounds(NMVOC), methane, and direct $PM_{2.5}$, relative to emissions from traditional
290 stoves). Under this scenario BC and $PM_{2.5}$ emissions in the study region are projected to decrease
291 throughout most of the domain, with SO_2 emissions showing almost no change (see Figs. S2c,
292 S3c, and S4c).

293 The lowest option assumed the additional elimination of high-emitting vehicles, biomass
294 cook stoves (in developing countries), and agricultural waste burning. These BC measures in the
295 lowest scenario reduced global anthropogenic BC emissions by 75%. Measures targeting BC
296 also substantially reduced total primary $PM_{2.5}$ (-50%), OC (-79%), NO_x (-27%), and CO (-44%).
297 These BC measures have little impact on SO_2 emissions. Projected emissions of BC and OC
298 under these scenarios are reduced most in Asia, followed by Africa. North America, and Europe.

299 These measures were also combined with a scenario designed to stabilize greenhouse
300 gases at 450 ppm of CO_2 equivalent (lowest+450ppm scenario), consistent with a global average
301 temperature increase of ~2° C. These CO_2 measures reduced SO_2 (-30%) (Fig. S4d) and NO_x (-
302 20%), but had little further impact on BC (~5% decline, Fig. S2d) since the major sources of CO_2
303 differ from those of BC. $PM_{2.5}$ emissions were substantially further reduced under this scenario
304 (Fig. S3d).

305 **2.4. Simulations Analyzed**

306 The CA region is strategically located and aerosols at these sites are shown to reflect
307 impact of varied dust (including CA, Western China, Africa and Middle East), biomass burning
308 (including Europe, Siberia, South Asia and North America) and anthropogenic sources
309 (including Europe, South, East and Central Asia, and Russia) depending on the time period. So
310 we choose different regions for the anthropogenic, dust and biomass burning in order to capture
311 the impact and the associated seasonality of these main regions on the regional distribution of
312 aerosols in this region. We have more anthropogenic regions to better understand how regional
313 changes in emissions may impact CA PM levels.

314 Several simulations were analyzed for this paper. The *base* simulation included all
315 sources and used the meteorology from the WRF model for the period April 2008 – July 2009.
316 To investigate the contributions from specific source sectors, additional simulations were
317 performed where emissions from one sector were set to zero everywhere. The contribution from
318 each sector was calculated as the difference between the base simulation and the simulation with
319 emissions from that particular sector set to zero. This was repeated for each sector and for
320 biomass burning. Additional simulations were performed to assess the source contribution from
321 specific regions to the particle levels in CA. The specific regions used are shown in Fig.1. In
322 these simulations all anthropogenic emissions were set to zero in that region. In a similar manner
323 regional dust and fire sources were also studied and the source regions are also shown in Fig.1.
324 Finally, simulations were performed using the *reference* 2005 and 2030 emissions and the *low*
325 and *lowest + 450* CO_2 2030 scenarios (described above). For these runs, the meteorology, open
326 biomass burning, and dust emissions were the same as in the base run (i.e., 2008/2009).

327 **3. Results and Discussion**

We have evaluated this model framework in other regions outside CA including arctic region and continental US previously, which are summarized below. The domain configuration and settings of the WRF/STEM modeling system used in this study have been applied to provide meteorology and air quality forecasting during the ARCTAS field campaign in 2008 (Dallura et. al., 2011) as mentioned earlier in sections 2.2.3 and 2.3.1 respectively. The study by Dallura et al., used WRF v2.2 and pre-emission ARCTAS emissions data (http://mic.greenresource.cn/arctas_premission), real time biomass burning emissions from the RAQMS modeling group (http://raqms-ops.ssec.wisc.edu/) and evaluated the performance of the WRF-STEM modeling framework (used in this study) through the case study of the ARCTAS mission DC-8 flight conducted on July 9 2008. The WRF model compared well with observed meteorological variables (including temperature, pressure, wind speed and wind direction) along the flight and correctly simulated the various meteorological processes (i.e. cyclonic circulation, low pressure system, anticyclones and geostrophic wind flows) that facilitate transport of pollutants into the Arctic region. This study also compared CO along the flight and found that the simulated values were able to capture the magnitude and temporal variability seen in observed values. The study also found that the fires from North Asia and anthropogenic CO from China were the major sources contributing to observed CO levels along the flight suggesting that the midlatitude pollution and summertime forest fires/biomass burning transport events were well represented in this modeling system.

Huang et al, 2012 used a subset of this domain (covering the region of continental North America) to simulate summertime sectoral and regional contributions to BC over continental US using the WRF/STEM modeling framework. Note that the Huang et al., 2012 study used the same WRF meteorology, ARCTAS emissions and RAQMS biomass burning emissions used in

351 the Dallura et al., 2011 study, but used the full-chemistry version of the STEM model. The
352 comparison of the simulated aerosol BC from the STEM tracer model with corresponding BC
353 values from the full chemistry version of the STEM model showed that the simulated tracer BC
354 was able to capture trend and the same relative variability seen in the full chemistry predictions.
355 Huang et al., 2012 evaluated the model performance aloft and at surface using measurements
356 from ARCTAS mission DC8 flights conducted on June 20 and 22 and 24 that sampled over
357 California and IMPROVE surface site network. Huang et al., 2012 found that the predicted BC
358 values captured many of the vertical features seen in the flight observations with a tendency to
359 over predict surface BC and > 4km. The comparison of predicted BC with observed values from
360 the IMPROVE network showed that the model was able to capture the gradients in BC values
361 with high concentrations in western and eastern NA regions with slight positive and negative
362 biases over the mountain regions and the eastern US/California.
363 These studies indicate that the simulated transport patterns are able to capture the main
364 important features of the hemispheric flows at the scale hemispheric flows impacting Central
365 Asia, North America and the Arctic region, at the scale discussed. The prediction of aerosol mass
366 and composition at the hemispheric scales are able to capture important aspects of horizontal
367 gradients and variability, but has considerably higher uncertainties associated with emission
368 estimates (in particular forest fires/biomass burning and natural dust emissions) and wet removal
369 processes (Bates et al., 2006).
370
371 **3.1 Comparison with surface BC observations from IMPROVE network**

372 We evaluated the simulated BC values with IMPROVE BC observations for the simulation
373 time period (i.e. April 2008 – July 2009) used in this study. The IMPROVE BC analysis is
374 limited to 27 sites on the US west coast (i.e. west of 120 W) to be consistent with the focus of
375 this paper (i.e., long range transport of pollutants on a hemispheric scale). The mean observed
376 and modeled BC for the West Coast sites are ~0.16 and 0.19 μg m−3 (n = 3759 points) with
377 mean bias and RMSE of 0.03 μg m−3and ~ 32 % respectively. The mean simulated/observed BC
378 at surface sites is ~ 1.2, which compares well with the corresponding ratio of 1.6 reported in the
379 global model performance over North America region by Koch et al., 2009. The detailed analysis
380 of the IMPROVE BC sites and associated seasonality will be the addressed in a future paper. The
381 rest of this section will focus on discussing the regional aerosol distributions in the CA region.

382 **3.2. CA Regional Perspective**

383 CA is a region with high aerosol loadings as shown in the mean MODIS retrieved AOD
384 at 550 nm for the time period of study (April 2008 – July 2009) (Fig. 3).AOD (period mean)
385 throughout CA (~ 45-90° E, 35-50° N) are greater than 0.25, with the highest regional values
386 around the desert areas near the Caspian and the Aral seas. There are also high values (>0.6)
387 along CA's eastern border, which reflect the deserts and rapidly developing cities in western
388 China, and to the south over Pakistan and northern India.

389 The predicted period mean AOD spatial distribution shown in Fig. 4d captures the main
390 observed features. The period-mean predicted surface concentrations of $PM_{2.5}$, BC, and total dust
391 (fine and coarse) are also plotted. The period mean $PM_{2.5}$ concentrations in CA (10 to 35$\mu g/m^3$)
392 have a similar geographical distribution as AOD. Dust is the major component of predicted PM_{10}
393 in CA and concentrations are high (25-100 $\mu g/m^3$). The BC levels in CA are typically less than

0.3 $\mu g/m^3$ and its spatial pattern reflects contributions from both anthropogenic and biomass burning sources.

3.3 Comparison with surface observations in CA

The surface observations at the two CA sites provide the opportunity for the first time to evaluate the performance of chemical transport models in estimating the distribution of aerosols in CA and to assess the emission estimates in the region. A comparison of the predicted and observed meteorology is presented in Fig. 5, where the distributions of key meteorological parameters for the entire measurement period are shown as box-plots. The model accurately predicted the magnitude and variability in temperature and relative humidity. For example the model mean value of temperature and relative humidity are 279.3K and 61.6% in comparison to the observed values of 280.3K and 59.2% at the LST site. The model wind speeds were biased high by about 30% (mostly in winter, see section 3.4) and the direction had a southwest bias. These biases are due in part to the 60 km model grid size, which is too coarse to capture the steep gradients in topography in and around the observation sites, and to the site placement in small N-S oriented valleys. More details regarding the evaluation of the modeled meteorology can be found in the Supplemental Materials, Table S1.

The observed and modeled distributions of AOD and PM are compared in Fig. 6. The AOD observations based on the LE on average are ~50% larger than those from the SP. Modeled AOD on average are ~20-30% higher when compared to SP at the Bishkek and LST sites and ~1 % lower when compared to the LE values. The variability in the predictions is slightly under-estimated. $PM_{2.5}$ is over predicted (~50%) and the spread is accurately captured, while PM_{10} is over predicted by ~70%. This leads to an underestimation of the $PM_{2.5}/PM_{10}$ (0.4 predicted

versus 0.5 observed) and also helps account for the overestimation in modeled AOD (by ~20-30%).

Chemical analysis of the filter and soil samples in the CA dust regions have been used to estimate the dust contribution to measured PM at the two sites and to help identify source regions of importance (Park et al., 2014). The emission regions within CA, including around the Aral Sea, and western China were identified as the most important dust sources, which is consistent with the regions identified in the simulations. Dust was estimated to comprise between 5-40% of $PM_{2.5}$ mass at the LST site and to vary by season (minimum values in winter). The observation-based estimates of dust percentage contribution suggest that modeled dust is over predicted by ~2 times. Thus it appears that dust is a main reason for the over prediction of $PM_{2.5}$ and PM_{10}, and that dust emission models need to be refined for CA applications.

The overestimation in PM mass at the surface could also be impacted by errors in the modeled PBL heights. The PBL height as determined by the Lidar aerosol profiles varies seasonally and is highest in the summer (from 2-4 km AGL) and lowest in the winter (Nov-Feb, 0.5-1.5 km AGL) (Fig. S5). The predicted PBL heights show a similar seasonal cycle with a tendency to under-predict the heights in all seasons as indicated by the comparison of the distributions of the observed and predicted values (Fig. 5), and this occurs in all seasons (Fig S5). The lower PBL height in the model contributes a systematic high bias in surface concentrations driven by near surface emissions. Further statistical details of the model-observation comparison can be found in the supplemental materials (Tables SM2 & 3). The seasonal variability in the observations is discussed in further detail later (section 3.4).

3.4 Source Contributions to $PM_{2.5}$

438 Model simulations were performed to identify the component, source region and
439 emission sector contributions to $PM_{2.5}$ mass. Period means for the spatial average over the entire
440 CA region (see Fig. 1) and for the grid cells for the Bishkek and LST observation locations are
441 presented in Fig.7, and their comparison provides insights into the spatial variability of PM and
442 its sources within CA, and how representative the observation sites are at characterizing CA PM.
443 The component contribution to AOD at the sites and for the CA average are similar, with the
444 major contributions coming from fine dust, sulfate, and OC. Spatial maps of mean percent
445 contributions of the various components (i.e., BC, OC, sulfate, OPM, dust and sea salt) to AOD
446 and $PM_{2.5}$ mass are presented in Figs. S6 & S7, respectively. Coarse particles contribute ~10% to
447 mean AOD. Dust accounts for >60% of the calculated $PM_{2.5}$ mass at the observation sites and for
448 the CA region. The dust source regions (see Fig. 1) contributing to $PM_{2.5}$ vary within CA. Dust
449 from the CA source regions has the largest influence on the region mean dust- $PM_{2.5}$ mass. At the
450 LST site, which is located in the far east of CA, western China dust sources have their largest
451 influence (~40%). African and Middle East source regions have their largest influence on the
452 Bishkek site (20 and 15%, respectively), and collectively contribute ~25% to regional CA dust
453 $PM_{2.5}$.

454 The source region contributions to the non-dust $PM_{2.5}$ are very similar for the Bishkek
455 and LST sites, with CA sources making the largest contribution (~50%) followed by Europe
456 (~20%), the Middle East (~15%), and biomass burning (~15% from all sources). For the entire
457 CA region the European source contribution is as large as the CA sources (~30% each), with
458 larger contributions from biomass burning and Russia sources and smaller contributions from
459 Middle East emissions than at the observation sites. The significant contribution from Europe
460 sources reflects their large anthropogenic emissions and the general westerly transport patterns.

On average the impact from South Asia sources are small. North America sources contribute between 3 to 7% to $PM_{2.5}$ in CA. Of the biomass burning contribution to $PM_{2.5}$, the Siberian and European fires (see Fig. 1 for fire regions) contribute 63 and 25%, respectively, with contributions from South/Southeast Asia and North America fires each contributing ~5%. The power and industrial sectors are identified by the simulations as the largest contributors (~40% each) to non-dust $PM_{2.5}$ mass in CA.

3.5. 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 the stations. This transport pattern occurs episodically throughout the year, with contributions from western China sources as large as 20 to 50%. The dust seasonal cycle is in turn influenced by the seasonal variations in meteorology that drives the dust emissions and transport. The seasonal changes in the dust source regions can be seen in the seasonal spatial maps of AOD (Fig. S8). Throughout the domain, AOD in the dust regions are highest in March – October and lowest in winter (Fig. S8) as the nearby dessert regions are snow covered.

Biomass burning also adds to the seasonal cycle, and its contribution is minimum in the winter. South Asia sources can impact the sites in the winter time. The periods when North America sources impact the site are associated with strong transport events across the Atlantic and subsequent subsidence towards the surface associated with high pressure systems as they move towards CA. The transport pathways are discussed in more detail in Sect. 3.7.

3.6 Source Contributions to BC

Because of its dual role as an air pollutant and as a climate warming agent there is special interest in understanding the regional and sector contributions to BC (Ramanathan and

Carmichael, et al., 2008). BC comprises on average only about 1-2% of $PM_{2.5}$ mass in CA. The period mean predicted BC surface concentrations are ~0.1 $\mu g/m^3$ at the two observation sites and 0.15 $\mu g/m^3$ for the CA regional average. As shown in Fig. 7, European emissions contribute ~50% to the mean BC concentrations in CA, while Middle-eastern and biomass burning sources each contribute ~15-20%. Residential and transport are the most important sectors each contributing ~30% to BC in CA, followed by industry (~20%), and with power the least important. This is in contrast to the sector contributions to non-dust $PM_{2.5}$ mass, where power and industry are the most important sectors. On average biomass burning contributes ~10% to BC mass, with Siberian and European fires accounting for 61% and 33%, respectively. The source contributions to OC are shown in Fig. S9.

There is also large seasonal variability in BC concentrations and source sector/region contributions (Fig. 12). BC surface concentrations show the highest values in fall/winter (as do the observations), when there is maximum contribution from the residential sector, reflecting the wide-spread use of biofuels and coal for heating in the region. The source region contributions vary by season, with maximum contributions from Europe and China. South Asia sources contribute in the winter. Biomass burning also is an important source of BC and plays an important role in influencing daily and seasonal variability in BC concentrations.

Predicted BC captures the seasonality and the magnitude of the spring and summer values as observed, but concentrations are biased low in the fall/winter. Median BC concentrations (and variability) are underestimated by a factor of 2 at both observation sites (Fig. 6 and Tables S2&3)). The high wind speed bias in winter (~ factor of 2), should result in too rapid dispersion and could contribute to the negative bias, but the negative bias in the PBL

524 heights should lead to higher predicted concentrations. Thus this negative bias is likely related to
525 emissions (an indication of an underestimation of the heating fuel use).

526 The OC concentrations follow a similar seasonal cycle as BC and are also under
527 predicted (Fig. 6 and Tables S2 & 3)). Furthermore the OC/BC ratio is under predicted by a
528 factor of ~3 (Fig. 6). The observed OC/BC ratio follows a seasonal cycle with values >15 in
529 summer and ~5 in September through April. Part of this under prediction in OC and the OC/BC
530 ratio is due to the fact that SOA is not estimated in the model. However a source contribution of
531 OC using the filter data and chemical mass balance (CMB) approach found that SOA sources
532 were very low in winter and only ~ 20% in summer (Miller-Schulze et al., 2011). Thus SOA
533 cannot account for the model under prediction of winter values. There appears to be an
534 underestimation of regional OC primary emissions. SOA can however help account for the large
535 values of OC/BC observed in the summer and not predicted.

536 Biomass burning emissions cannot account for the underestimation in winter BC and
537 OC. The largest impact of fires at the observation sites is in the late summer, when the fires are
538 concentrated in western Russia and the wind direction is such that the smoke is transported into
539 CA. Fires from South Asia can impact the sites associated with the fires and high pollution levels
540 in northern India and with winds from the south, which can occur in late fall, but not frequently.

541 The fact that BC and OC are systematically under predicted in the winter suggests that
542 local/regional emissions during the heating season may be underestimated. This is supported by
543 the results of the CMB analysis of OC discussed above that found the contribution from biofuel
544 combustion increased 2-3 times in the fall and winter periods. The uncertainty in emissions can

545 also be partly caused by the lack of seasonal emissions over this region as described earlier in
546 section 2.

547

548 **3.7 Vertical Distributions**

549 Figure 13 shows the predicted weekly averaged vertical distributions of $PM_{2.5}$, dust, and
550 BC for the entire simulation period at the LST site. These plots show more clearly that much of
551 the variability in the PM loadings is associated with dust and biomass burning episodes (as
552 represented by the enhancements in BC). Typically the high PM episodes show elevated PM
553 mass that extend from the surface to 2 to 4 km. The vertical extents show a seasonality
554 associated with seasonal variations in the PBL heights. These vertical distributions indicate that
555 much of the transport of aerosols in CA occurs via low altitude pathways. In some cases there are
556 large amounts of dust and biomass burning aerosol in the 3-6 km altitude range that are
557 decoupled from the surface (e.g., dust in early May 2009), reflecting that some aerosols are lifted
558 out of the boundary layer and are transported at high altitude over CA, enhancing AOD but not
559 contributing to ground-level mass concentrations at the observation sites. These vertical features
560 are confirmed by the aerosol extinction profiles observed at the LST site as discussed in Chen et
561 al., (2012 b). The variation in weekly averaged AOD can be significant (Fig. 13 bottom panel)
562 and is driven by variations in dust and biomass burning emissions.

563

564 **3.8 Transport Pathways**

565 The three dimensional ten day air mass trajectories (described in section 2.2) were utilized to
566 further understand the transport pathways of air masses entering into and exiting out of the CA
567 region and its subsequent impact of source regions on the aerosol distributions at the CA sites.

568

569 3.7.1 Transport into CA

570 The air mass transport into CA is discussed through back trajectories associated with the
571 five events labeled on Fig. 13. These five events represent transport episodes with elevated
572 surface $PM_{2.5}$ (averaged over the three hour time window consistent with trajectory time step)
573 with varying contributions from biomass burning, anthropogenic pollution, and dust sources. In
574 each trajectory figure (Figs. 14 and 15), the regions with active dust (blue diamond hatches) and
575 biomass burning emissions (green square hatches) for the event time period and prior ten days
576 are identified and MODIS AOD (values printed in black) overlaid. The trajectories are color
577 coded by region to distinguish the source impacts. The regions include: Africa (blue), Middle
578 East (green), CA (yellow), North Asia biomass (> 50° N, black), Europe (brown), China (red)
579 and South Asia (orange).

580 Figure 14 shows the first two events for August 2008. The first event in early August
581 (Fig. 13, event labeled 1) is a high dust event associated with trajectories from the west passing
582 over the dust regions of the Aral Sea (~45N, 55E) and eastern trajectories passing over the
583 Taklimakan region of western China (90-100E, 40N). These were both regions with active dust
584 emissions as indicated by the elevated MODIS AOD values. Thus the PM increases during this
585 event were due largely to dust emitted from CA and western China deserts. The second event in

586 the latter half of August is characterized by high levels of BC without dust. During this episode
587 the transport to the site was under the influence of a high pressure system located to the
588 northwest and air masses were transported over the active fire region in western Russia.

589 Figure 15 shows winter and spring events. The November episode (event 3) is a period
590 with elevated BC and $PM_{2.5}$ from pollution sources from South Asia (including some fires) and
591 western China and low fire and dust emission activity. The January episode (event 4) is a period
592 of elevated BC with air masses coming from Europe, indicating the influence of anthropogenic
593 pollution coming from this industrialized region, and from CA sources. Dust emissions from CA
594 and Africa were low during this period. The final illustrative episode is for April 2009 (event 5),
595 a period with both elevated levels of dust from western China, CA and Africa and BC from both
596 fire and anthropogenic pollution from Europe, CA and Russia sources.

597 These examples provide insights into the source region contributions to PM mass in CA
598 as presented in Figs. 11 & 12). CA is an ideal location to observe a variety of source regions as it
599 is at the crossroad of transport patterns with air masses impacted from dust, anthropogenic
600 activity and biomass burning from different geographical regions.

601 3.7.2 Long range transport of CA sources

602 The transport pathways out of CA were also evaluated by calculating forward trajectories
603 from the observation sites. Selected forward trajectories initialized at or below 1 km are used to
604 represent the transport of boundary layer PM from CA and these are shown for summer, winter
605 and spring periods in Fig. 16. In these plots the MODIS AOD, dust, and fire emissions plotted
606 for each event represent values averaged over the subsequent ten days and trajectories were

607 stopped if they impacted the surface. During the summer, outflow from CA is towards the north
608 in association with the summer monsoon system. Figure 16a shows the subset of forward
609 trajectories that reside for at least 3 days over the region 48 - 65° N during June 2008.
610 Trajectories typically pass over Russia and reach into the Arctic and also can be caught in
611 westerly storm tracks and reach the higher latitudes of the northern hemisphere. During the
612 winter, air mass transport out of CA is associated with the winter monsoon system and the
613 transport pathway is over Mongolia and then over eastern China as seen in Fig. 16b. In this
614 figure, forward trajectories that pass over the region west of 130° E during late December 2008
615 are shown. Figure 16c illustrates transpacific transport of CA air masses. Plotted are the forward
616 trajectories that stay within the 30 – 50° N region for at least 3 days during the month of April
617 2009. During the spring transport from CA is dominated by strong westerly flows and air masses
618 are transported over China, Korea, and Japan and then across the Pacific, reaching North
619 America in 7 – 10 days.

620 PM arising from dust and anthropogenic emissions from CA impact the entire northern
621 hemisphere as illustrated in the hemispheric maps of seasonal average surface dust
622 concentrations due to CA emissions only (Fig. 17). The predominant westerly flows in all
623 seasons in the northern hemisphere result in the bulk of the CA emissions being transported to
624 the west. The contribution of CA sources to surface PM concentrations of 0.35 $\mu g/m^3$ or greater
625 covers large portions of the northern hemisphere, including the Arctic, all of Asia, much of
626 Europe, and portions of the continental US.

627 There are episodic and seasonal components to the intercontinental transport as shown in
628 the time series of the vertical profiles of $PM_{2.5}$ at Mt. Bachelor, Oregon (43.97N, 121.69W,

629 2700m agl) (Fig. 18). The bulk of the CA particle transport takes place in the free troposphere
630 and impacts surface concentrations in the US as the boundary layer grows and entrains "plumes"
631 aloft. This occurs most frequently in spring, summer and fall. The episodic contributions of CA
632 sources to surface concentrations can exceed 1.5μg/m^3. In the fall, there is also strong transport
633 of dust from CA across Europe and out into the Atlantic.

634

635 **3.9 Future Scenarios**

636 How might PM levels change in CA over the next few decades? To address this question,
637 simulations were repeated for various emission scenarios developed and used in the
638 WMO/UNEP assessment on short-lived climate pollutants (Shindell et al., 2012) as described in
639 Sect. 2. Dust and biomass burning emissions and meteorology were the same as those used in the
640 2008/2009 simulations. The period mean changes in surface BC and $PM_{2.5}$ concentrations in
641 2030 for the reference scenario are shown in Fig. 19a, d, respectively. This scenario reflects all
642 present agreed policies affecting emissions and assumes that they are fully implemented. Under
643 this scenario $PM_{2.5}$ increases significantly in South Asia and western China (>50%) and in parts
644 of CA, including the area where the sampling sites are located. $PM_{2.5}$ decreases in Western
645 Europe and Eastern China (< 10%). BC surface concentrations show a similar pattern to $PM_{2.5}$,
646 although covering larger portions of CA with relatively larger increases in BC than in $PM_{2.5}$.
647 These results suggest that health impacts and climate warming due to BC and $PM_{2.5}$ may increase
648 in coming decades unless additional emission control measures are implemented.

649 Results for two other scenarios are also presented in Fig. 19. One scenario specifically
650 targets BC emission reductions in recognition that BC is also a major contributor to atmospheric
651 warming (Ramanathan and Carmichael, 2008). These additional measures significantly reduce
652 2030 BC concentrations by greater than 35% throughout most of the domain, with only a few
653 regions (e.g., Myanmar and eastern Afghanistan) showing increases in BC relative to 2005
654 levels. This scenario assumes that all BC emission reduction measures are perfectly implemented
655 and 100% effective. BC measures also impact emissions of co-emitted pollutants (e.g., OC and
656 SO_2). $PM_{2.5}$ concentrations under this scenario (Fig. 19e) are reduced, but by much smaller
657 amounts, and concentrations still increase relative to 2005 over large regions of South Asia and
658 western China, and parts of CA. These results suggest that health impacts in these regions may
659 increase due to the $PM_{2.5}$ increases whereas positive radiative forcing and health effects due to
660 BC may decrease. When the BC measures are used along with greenhouse gas measures aimed at
661 keeping CO_2 levels below 450 ppm, the $PM_{2.5}$ levels in South Asia are lower than 2005 levels
662 (Fig. 19f), with few exceptions (one being Myanmar region). This is due to the large decreases in
663 SO_2 and NO_x emissions under this scenario, resulting in significant reductions in particulate
664 nitrate and sulfate (e.g., a ~30% increase in particulate sulfate in CA).

665 To get a regional perspective of how the future emission changes would specifically
666 impact the CA region (See Fig. 1 for the CA region definition used in this study), we have
667 summarized the changes in emissions and the corresponding concentrations in Table 1. The
668 reference 2030 emissions scenario reports an increase in emissions of BC, SO_2 and $PM_{2.5}$ over
669 Central Asia by ~ 22, 17 and 14 % respectively w.r.t to base 2005 levels. However, the
670 corresponding BC, SO_4 and anthropogenic portion of $PM_{2.5}$ decrease by ~ 5, 12 and 5 %
671 respectively. This suggest that on average the concentration levels go down even though CA

emissions increase due to long range transport of pollutants into CA region from the surrounding regions, but locations within CA still increase when dominated by local sources. The Low GWP emission scenario predicts decrease in BC and $PM_{2.5}$ emissions by 31% and 10% respectively with the corresponding decrease in concentrations by ~ 37% and 10 % respectively, while SO_2 emissions remained same as the reference 2030 scenario. The lowest + 450 ppm scenario shows a consistent decreasing trend in emissions and concentration of all the species. This analysis suggests that the impact of the changes in major source region emissions and their subsequent transport to Central Asia will play a major role in determining the future aerosol levels in CA.

4. Summary

AOD in CA during the period April 2008 through July 2009 averaged ~0.3 and displayed a seasonal cycle with the lowest values in the winter and highest values in spring to mid-summer with observed values of AOD > 0.6 and even > 0.8. Surface $PM_{2.5}$ measured at two sites in eastern CA averaged ~10 $\mu g/m^3$ but with large variability (hourly values from 2 - 90 $\mu g/m^3$). Surface concentrations of PM also showed a seasonal cycle with peak values and largest variability in the spring/summer, and lowest values and variability in the winter. BC at these sites averaged ~0.1 $\mu g/m^3$ with peak values (~ 1 $\mu g/m^3$) in the winter.

The seasonal cycles and source sector and source region contributions to PM in CA were analyzed using the STEM chemical transport model. Dust was the largest component of the $PM_{2.5}$ and PM_{10} mass in the region in all seasons except winter, whereas sulfate was the largest anthropogenic component of the $PM_{2.5}$ mass. Dust was also found to be the major driver of the seasonal cycles of AOD and PM concentrations. On an annual basis the power and industrial sectors were the most important contributors to $PM_{2.5}$, while residential and transportation were

694 the most important sectors for BC. Open biomass burning within and outside the region also
695 contributed to elevated PM and BC concentrations and to the temporal variability.

696 The model simulations showed a systematic over prediction of PM mass. This is most
697 likely due in large part to the over prediction in dust. Carbonaceous PM was underpredicted and
698 it is speculated that the winter emissions associated with residential heating may be
699 underestimated in the current emissions inventory. The predicted wind speeds were biased high
700 (by ~30%) and the direction had a southwest bias. The high bias in wind speeds may also
701 contribute to the over-prediction in PM_{10}, as dust emissions depend strongly on wind speed.
702 Efforts to improve the dust emissions and to improve the wind speed and direction predictions
703 using a finer model resolution are planned. Additional efforts are needed to improve the
704 anthropogenic emissions estimates for CA.

705 Currently there are few measurements in CA that can be used to quantify the
706 intercontinental transport of pollution from Europe to Asia. The analysis of the transport
707 pathways and variations in PM mass and composition observed at the two sites in CA
708 demonstrate that this region is strategically located to characterize regional and intercontinental
709 transport of pollutants. Aerosols at these sites were shown to reflect dust, biomass burning, and
710 anthropogenic sources from South, East, and CA, Europe, and Russia depending on the time of
711 year. For example, during the spring fine particles from Europe and Africa were transported to
712 CA, on to eastern Asia, and then across the Pacific to North America.

713 Observations of PM and its composition in this region are of growing importance as it is
714 estimated that $PM_{2.5}$ levels are likely to increase significantly in Central and South Asia and
715 western China over the next few decades. Simulations for a reference 2030 emission scenario

showed that BC concentrations had a larger relative increase than $PM_{2.5}$ concentrations. This suggests that health impacts and climate warming associated with these pollutants may increase over the next decades unless additional control measures are implemented. Continued pollutant observations in CA will help to characterize the changes that are rapidly taking place in the region.

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7. Table Captions

Table:1 Summary of changes in Central Asia (CA) region (See figure 1 for CA region definition) emissions and concentration under the future emission scenarios (See Sect. 3.9 for more details)

8. 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

938 percentile values respectively. The whiskers denote the maximum and minimum values. The
939 triangle marker denotes the mean value.

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

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

951 Fig. 9. Comparison of simulated PM mass with filter-based observations at Central Asia sites (a)
952 $PM_{2.5}$ (LST) along with TEOM non-volatile measurements, (b) $PM_{2.5}$ (Bishkek), (c) PM_{10} (LST),
953 and (d) PM_{10} (Bishkek) in ($\mu g/m^3$). Lidar denotes the LST (Lidar Station Teplokluchenka) site.

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

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

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

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

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

974 Fig. 15. Same as Fig. 14 but for November 2008, January 2009 and April 2009 (events 3, 4, 5
975 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.

9. Supplemental Materials Figure and Table Captions

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

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

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

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

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

Fig. S3. Spatial distribution of a) base year 2005 $PM_{2.5}$ emissions (Gg/yr/grid) along with percent change (w.r.t to base year 2005) in b) Reference 2030 $PM_{2.5}$ emissions c) 2030 $PM_{2.5}$ emissions with BC measures and d) 2030 $PM_{2.5}$ emissions with BC and greenhouse gas measures aimed at

keeping CO_2 levels below 450ppm. The triangle and circle markers denote locations of the LST and Bishkek sites. Refer to Sec. 2.3 for more details on emission scenarios.

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

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

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

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

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

Fig S9. Summary of period mean contributions by source regions and sectors for OC 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 and fire source regions.

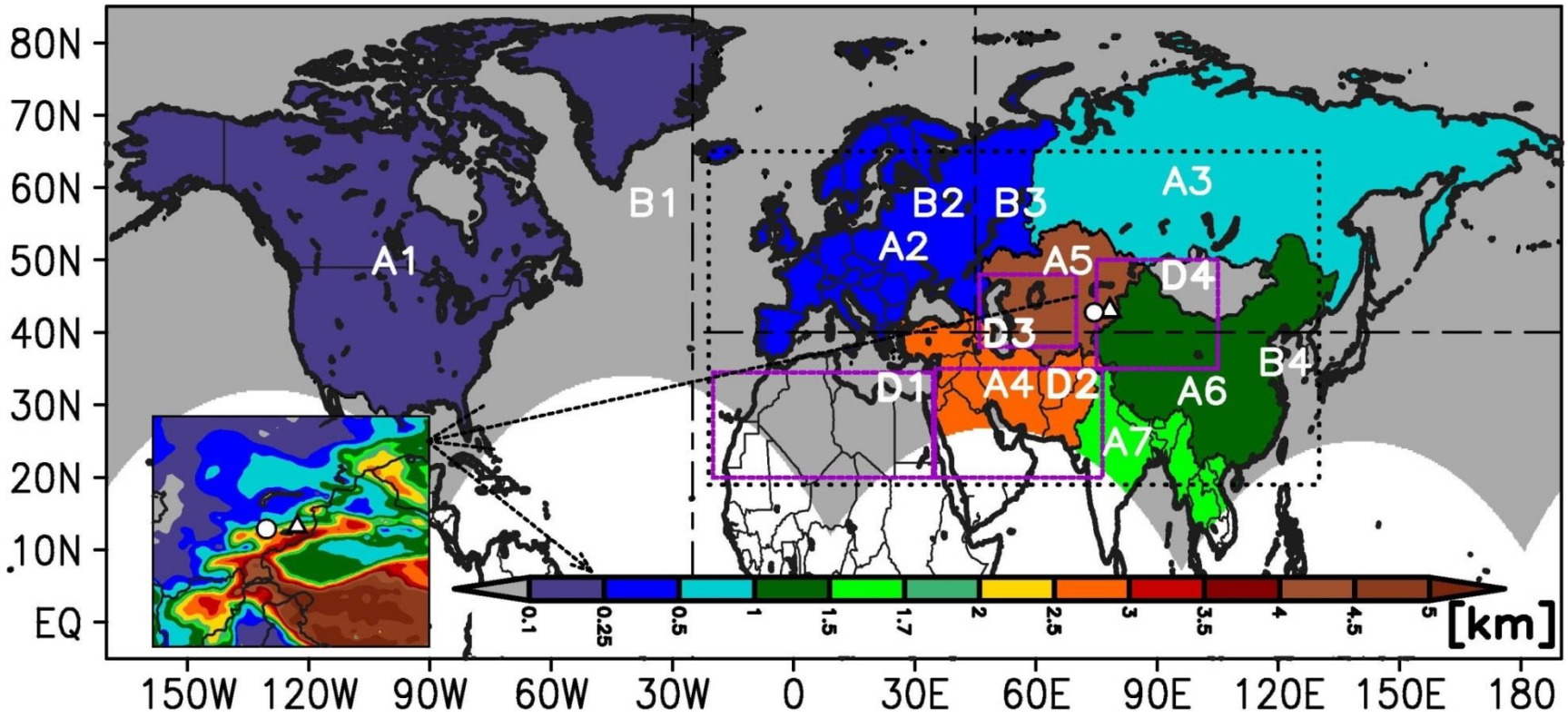
1031 Table:1 Summary of changes in Central Asia (CA) region (See figure 1 for CA region definition)
1032 emissions and concentration under the future emission scenarios (See Sect. 3.9 for more details)
1033

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

1034

1035



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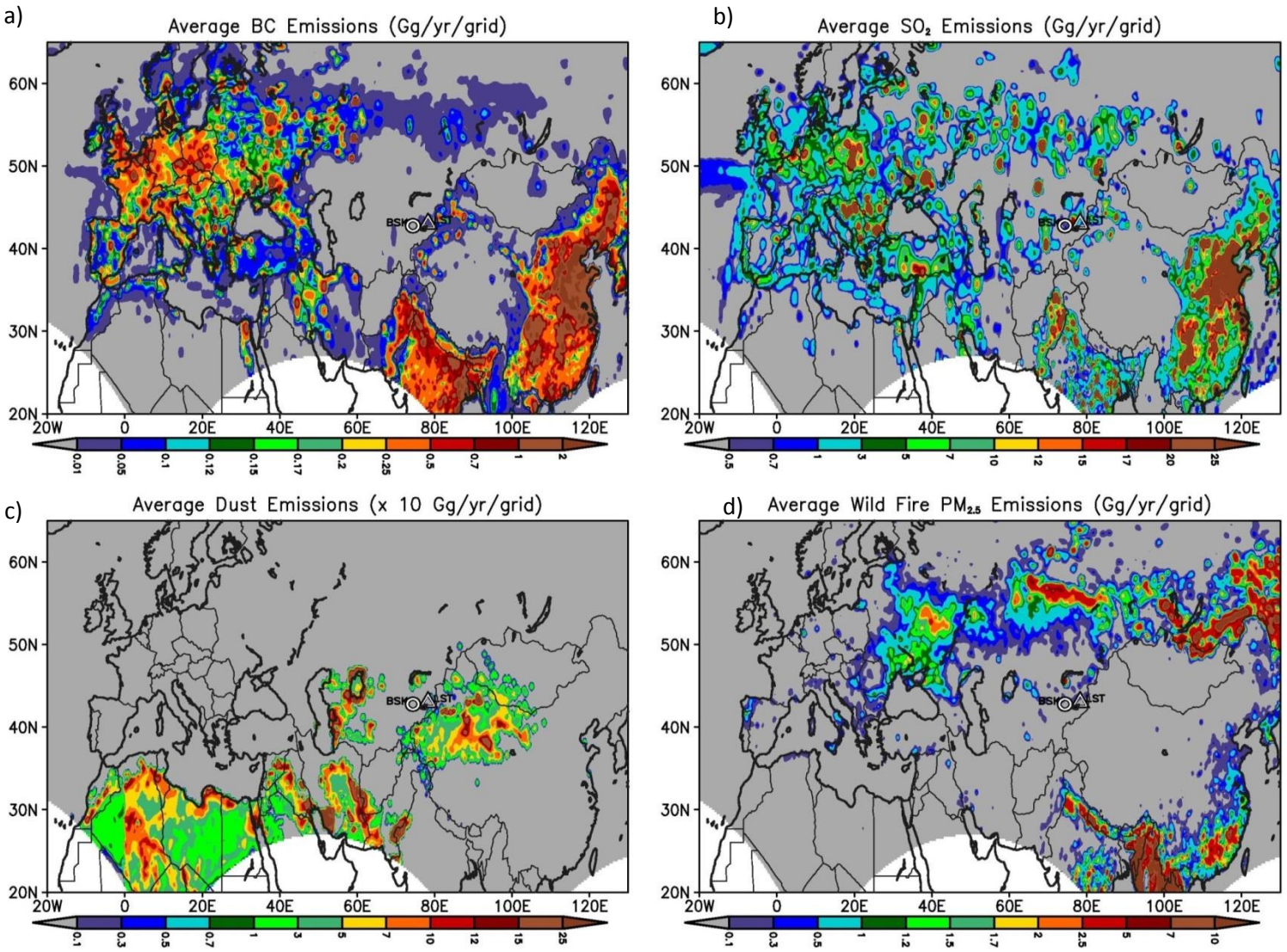

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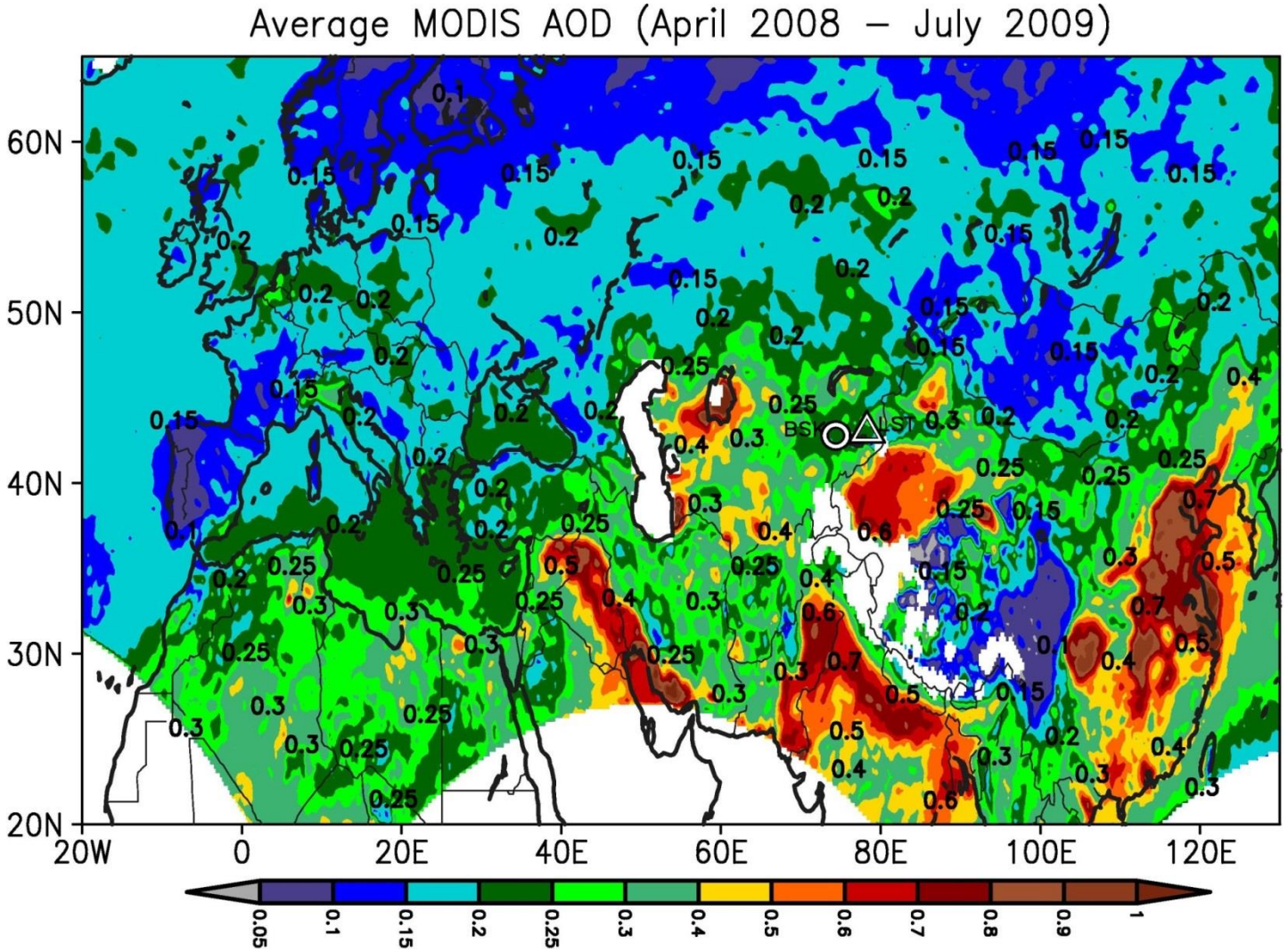

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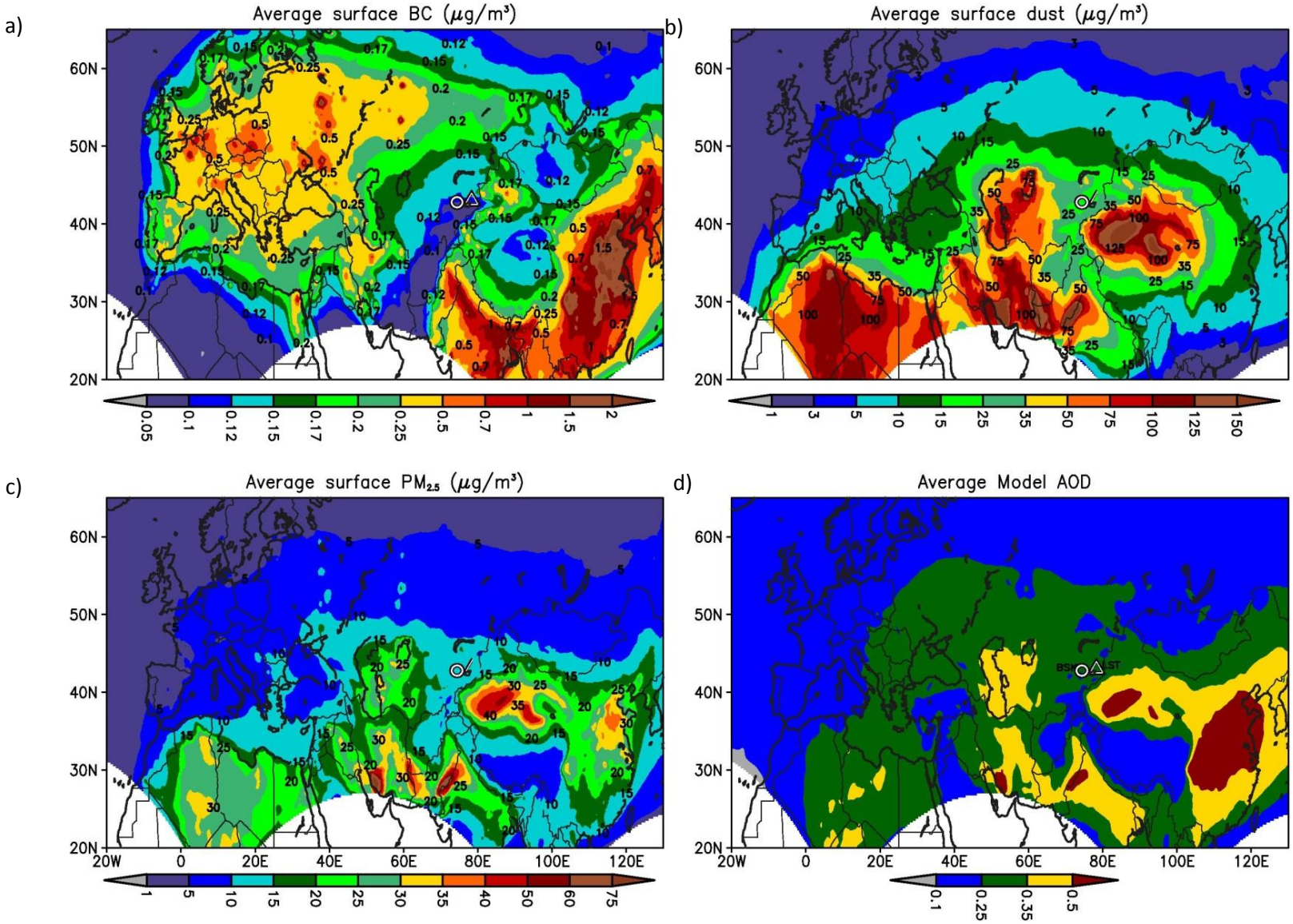

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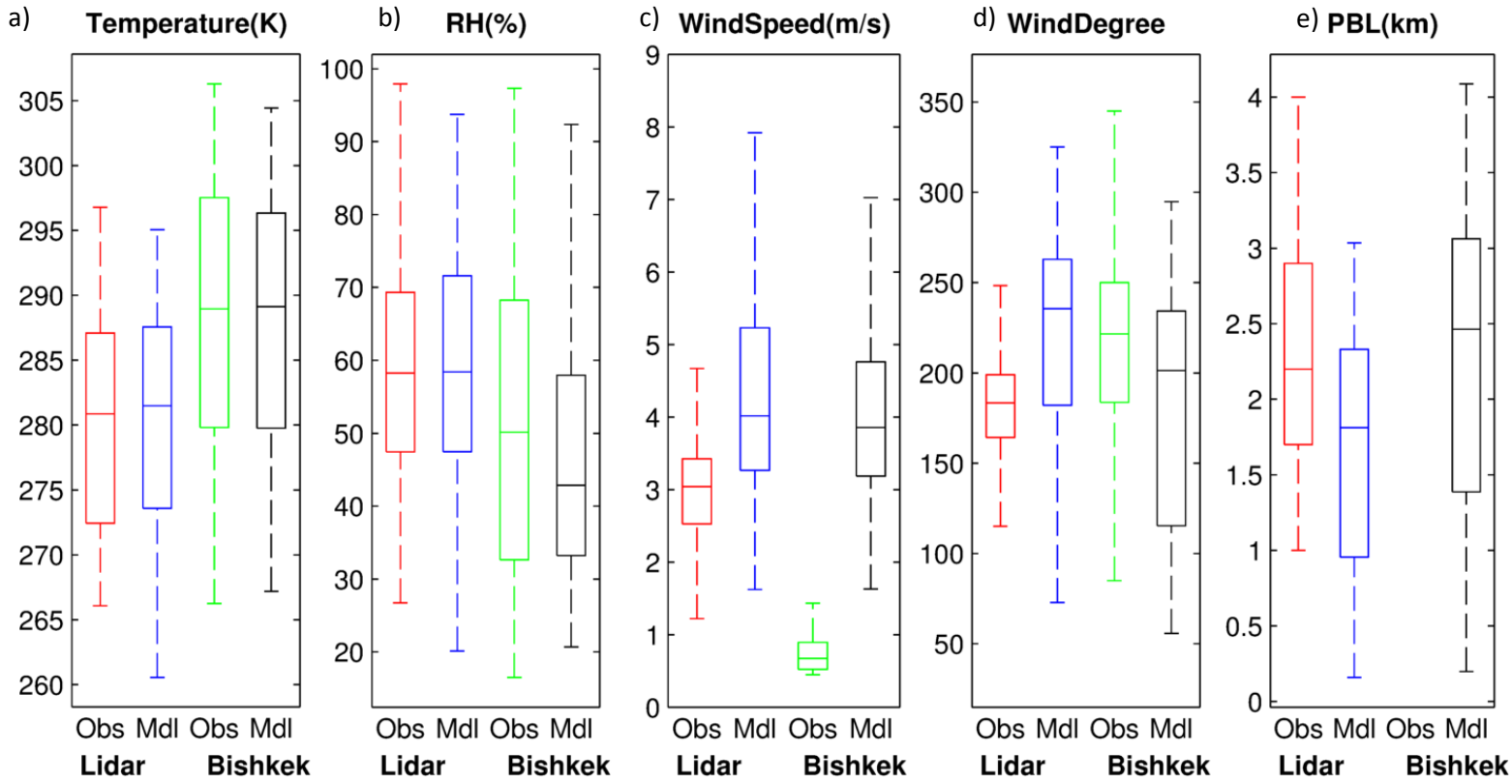

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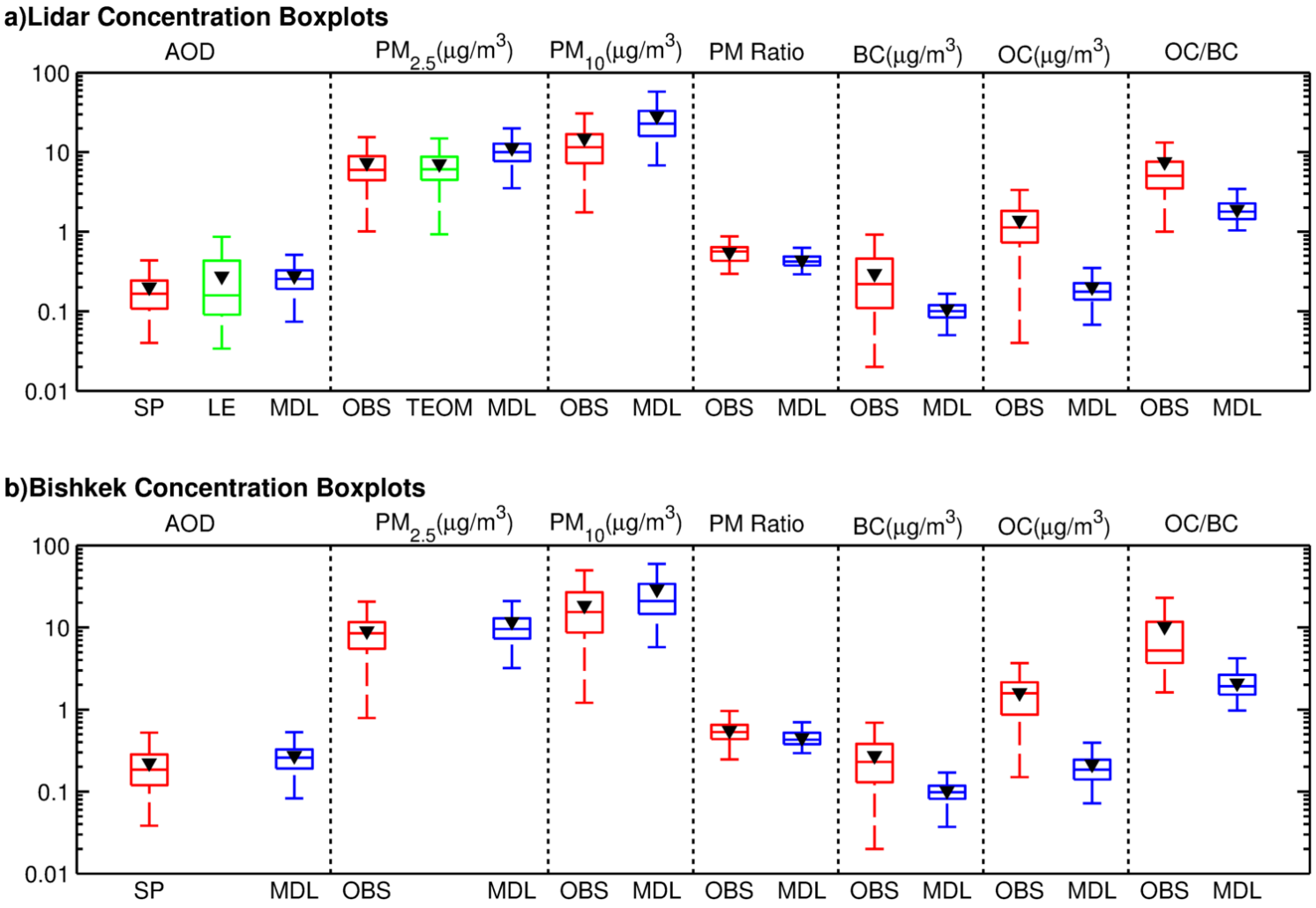

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box represent 25th and 75th percentile values respectively. The whiskers denote the maximum and minimum values. The triangle marker denotes the mean value.

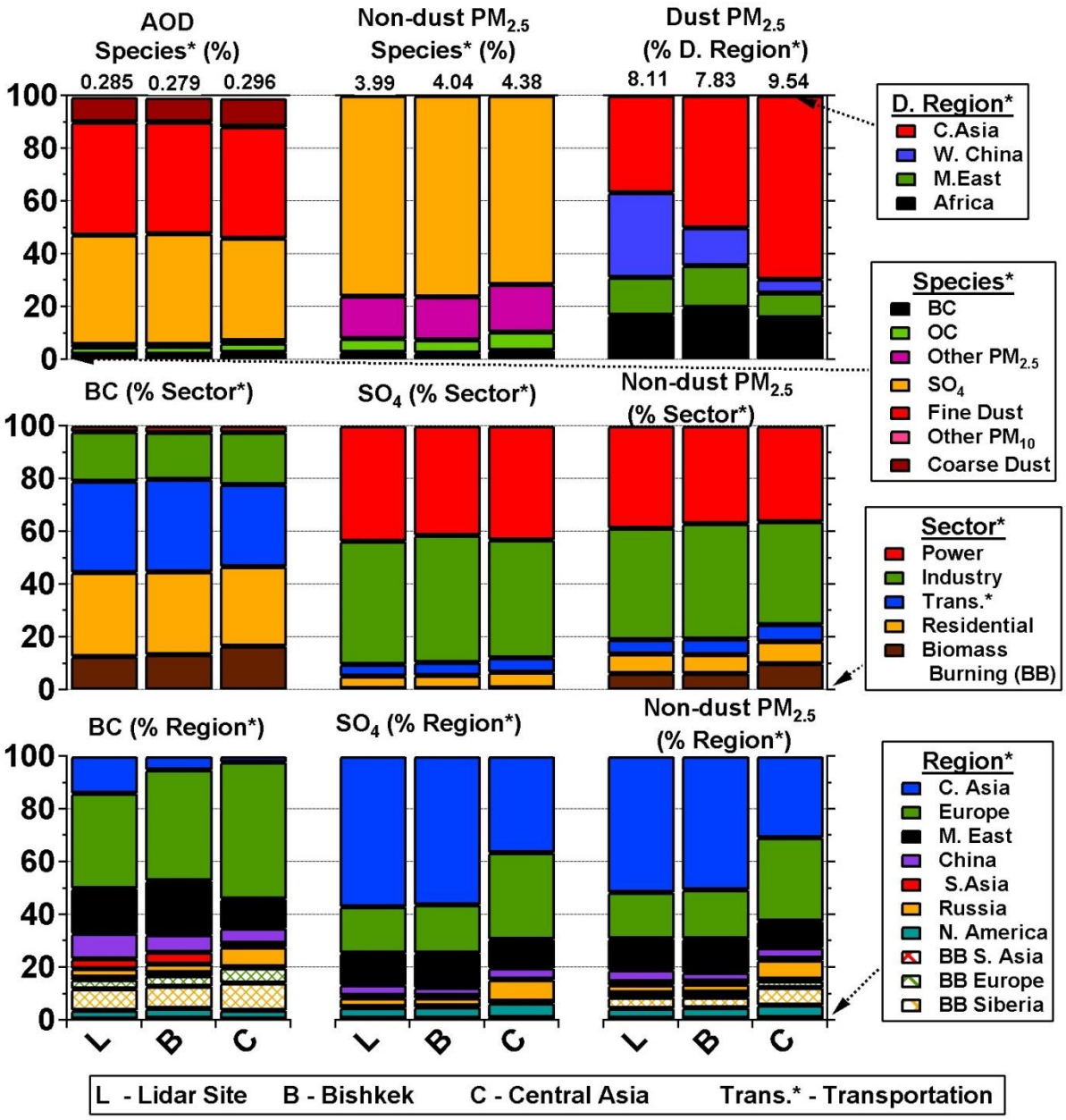

Fig. 7. Summary of period mean contributions by source regions and sectors for AOD, $PM_{2.5}$, dust, non-dust $PM_{2.5}$, sulfate and BC in % for the grid cells containing the Bishkek and LST observation sites, and spatially averaged over the Central Asia region. See Fig. 1 for anthropogenic, dust and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

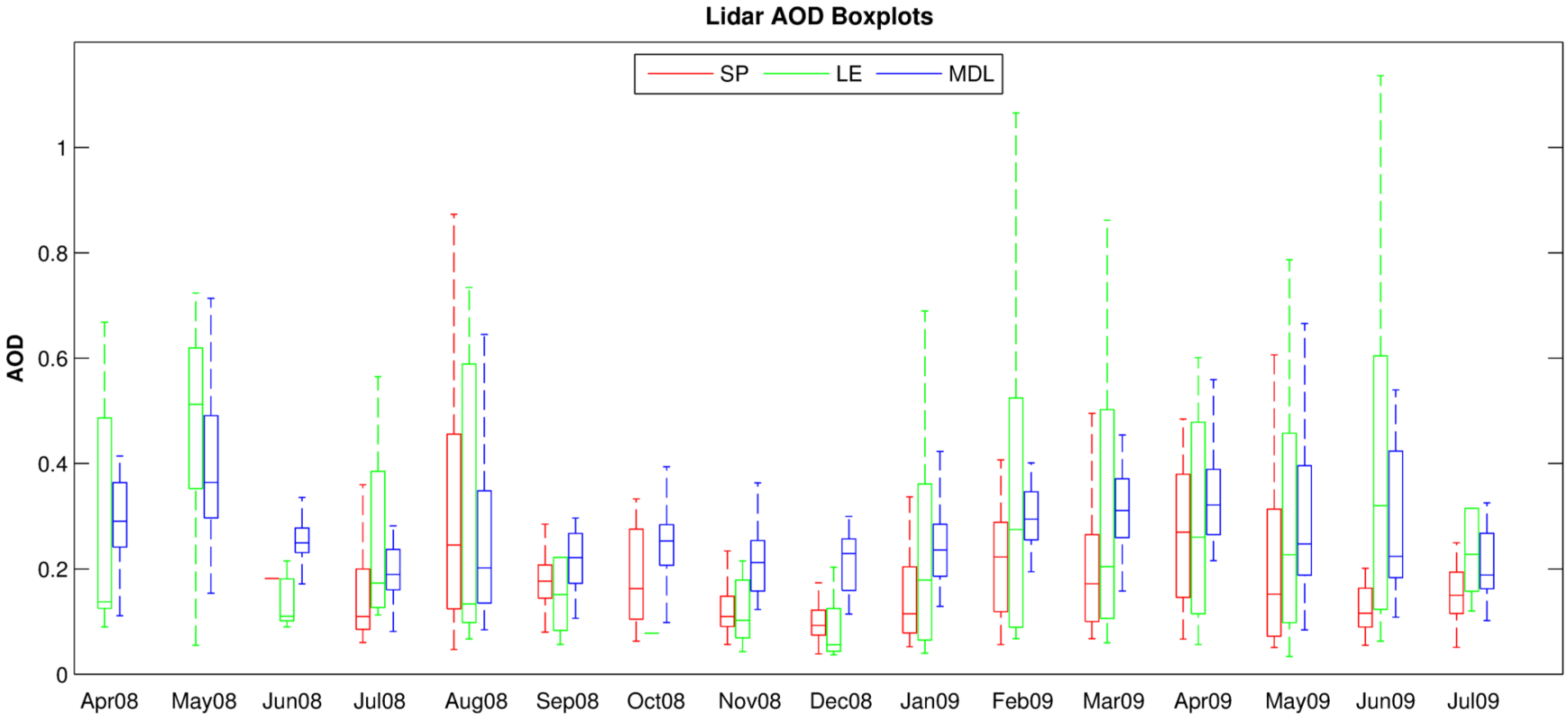

Fig. 8. Temporal variability in simulated AOD compared with observations at the LST site. The box-plots of monthly values are shown. AOD from the sun-photometer (SP) and integrated from the vertical extinction profiles (LE) are shown. MDL denotes modeled values. Lidar denotes the LST (Lidar Station Teplokluchenka) 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.

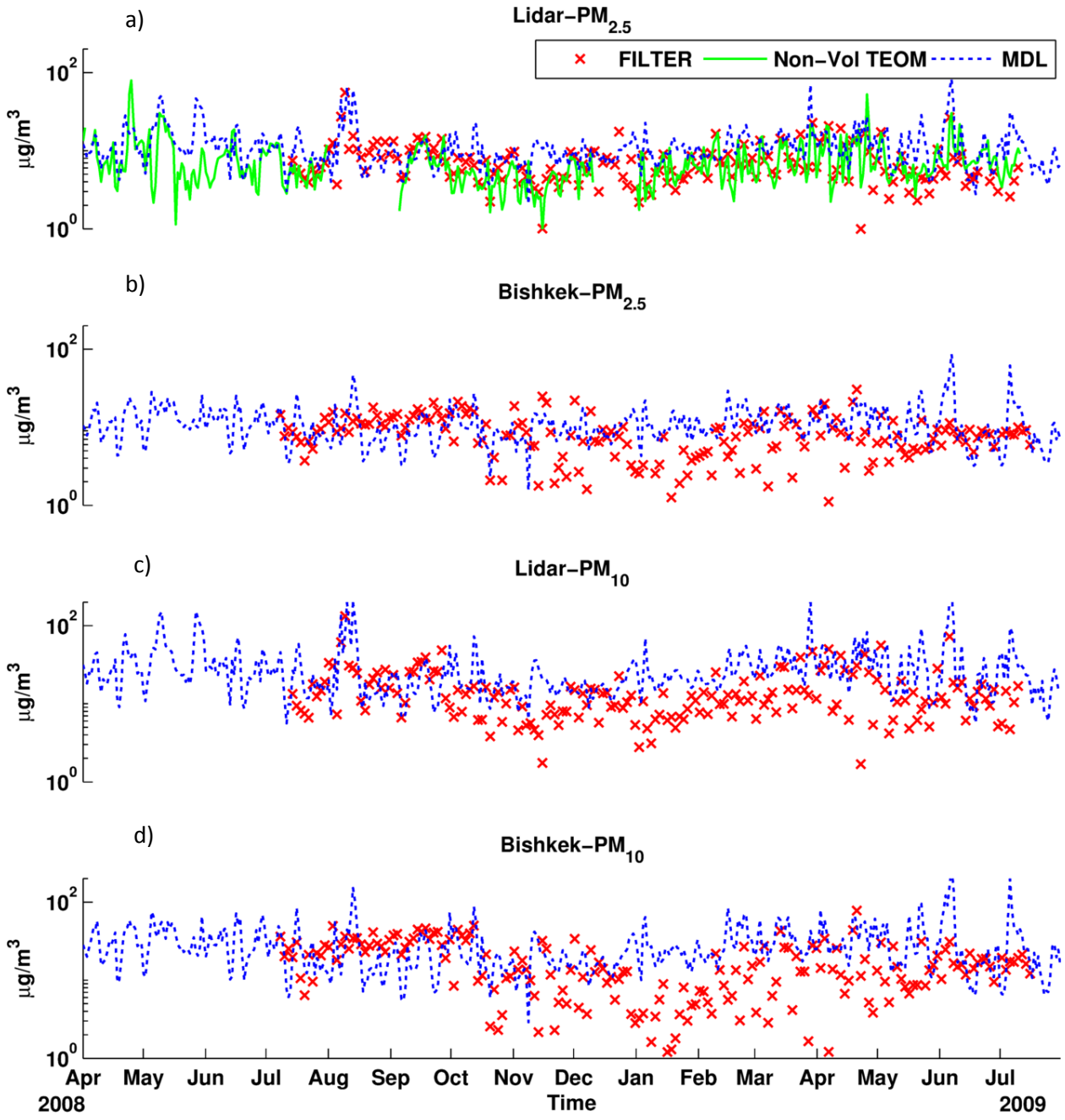

Fig. 9. Comparison of simulated PM mass with filter-based observations at Central Asia sites (a) $PM_{2.5}$ (LST) along with TEOM non-volatile measurements, (b) $PM_{2.5}$ (Bishkek), (c) PM_{10} (LST), and (d) PM_{10} (Bishkek) in (µg/m^3). Lidar denotes the LST (Lidar Station Teplokluchenka) site.

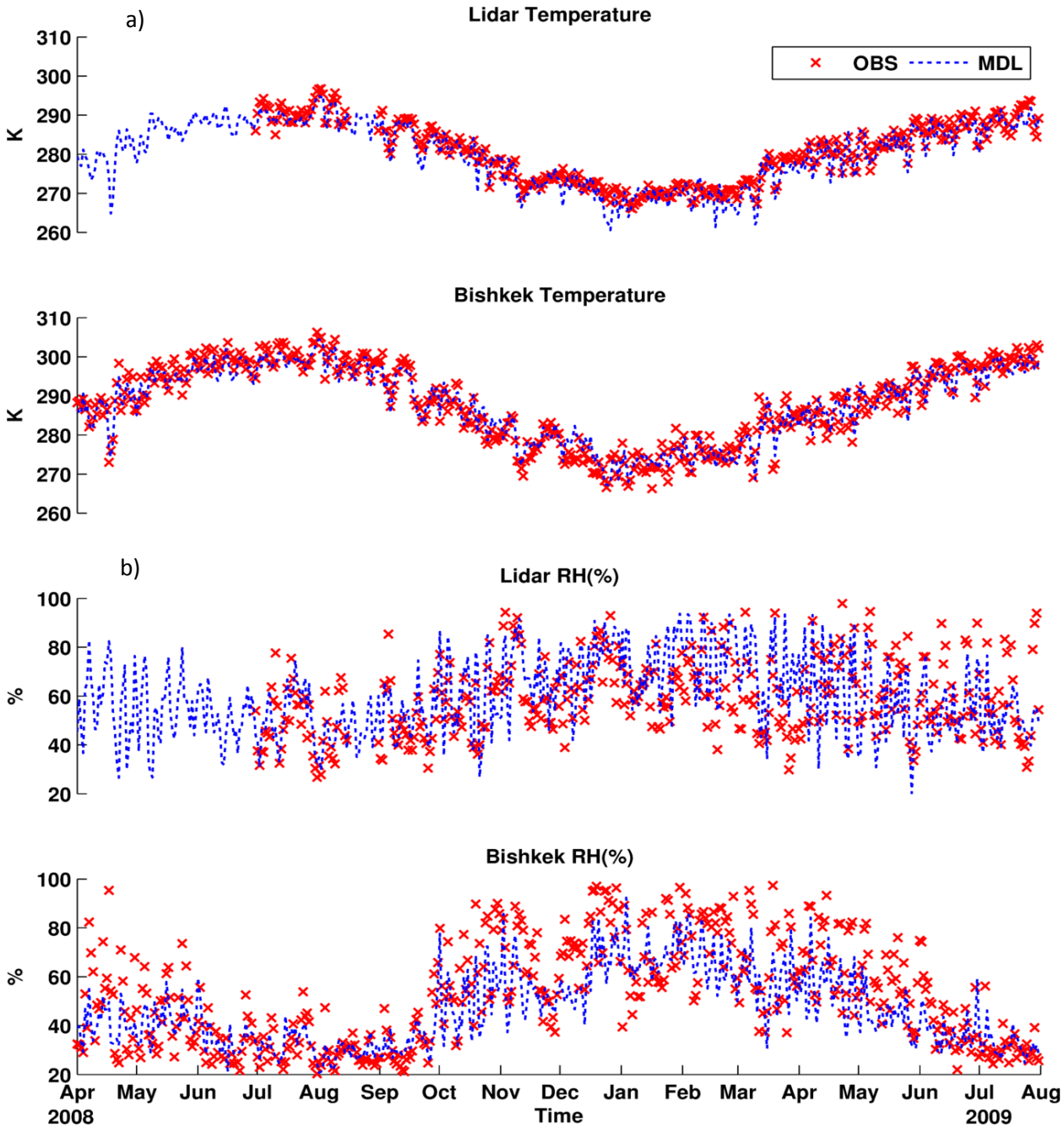

Fig. 10. Temporal variability in simulated Temperature (K) and (b) Relative Humidity RH (%) from WRF model compared with observations at the LST and Bishkek sites. Lidar denotes the LST (Lidar Station Teplokluchenka) site.

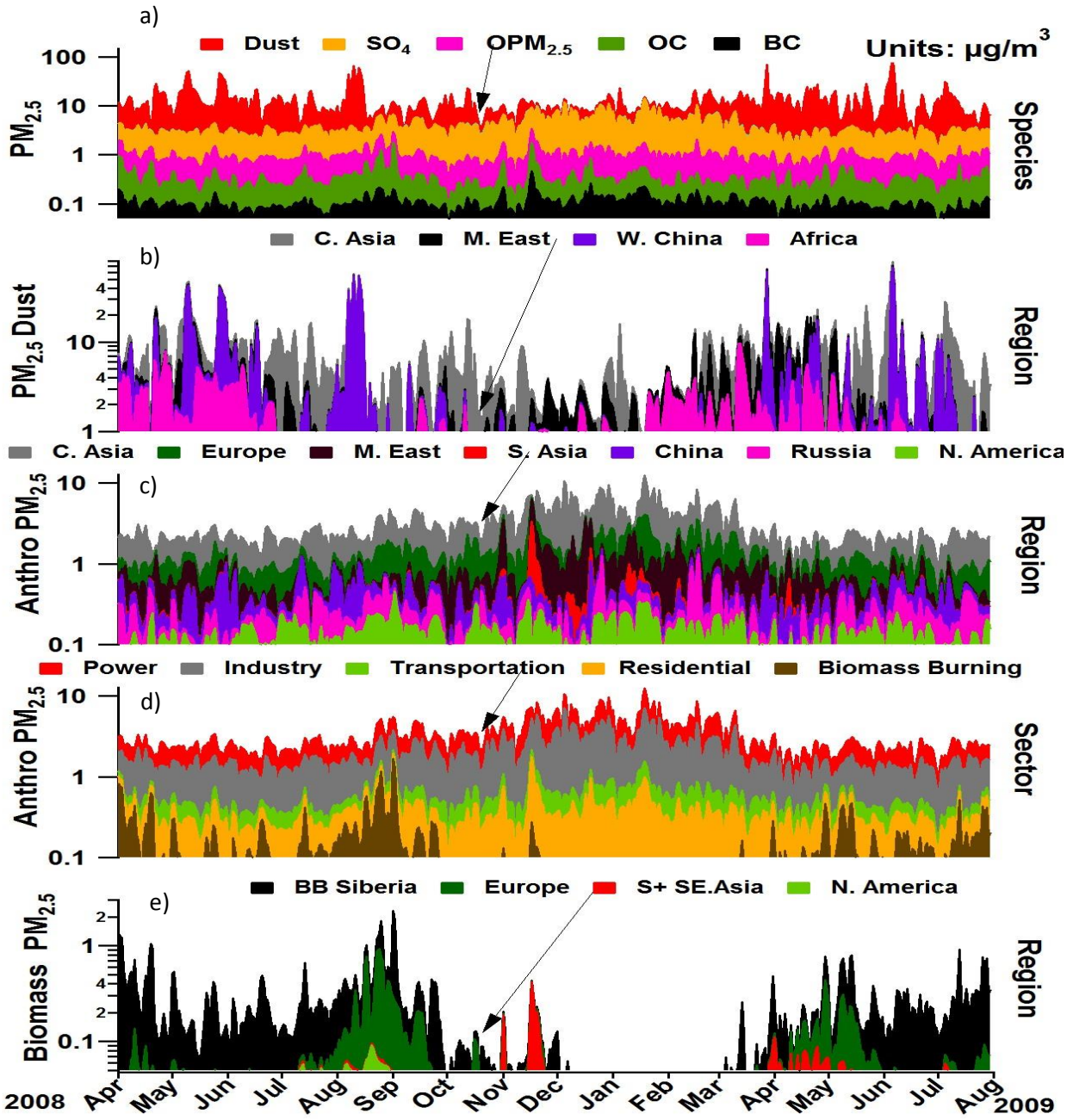

Fig. 11. Simulated composition of $PM_{2.5}$ at the LST site by a) Species (µg/m^3), b) Source sector (µg/m^3), and c) Source regions (µg/m^3). The contributions from source region and sectors denote the non – dust portion of $PM_{2.5}$ mass.

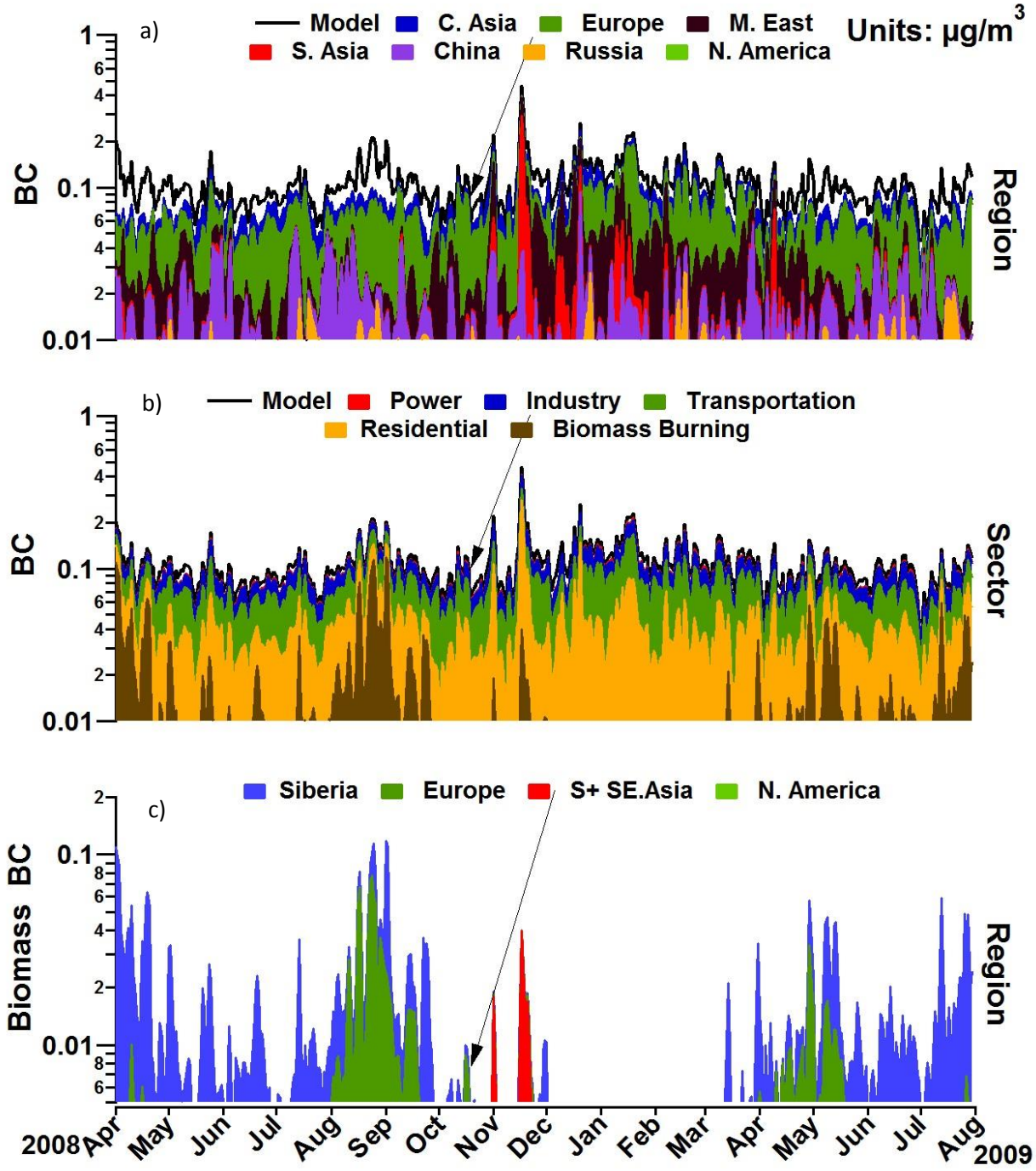

Fig. 12. Simulated composition of BC at the LST site by a) Source regions (µg/m^3), b) Source sector (µg/m^3), and c) Biomass burning source regions (µg/m^3).

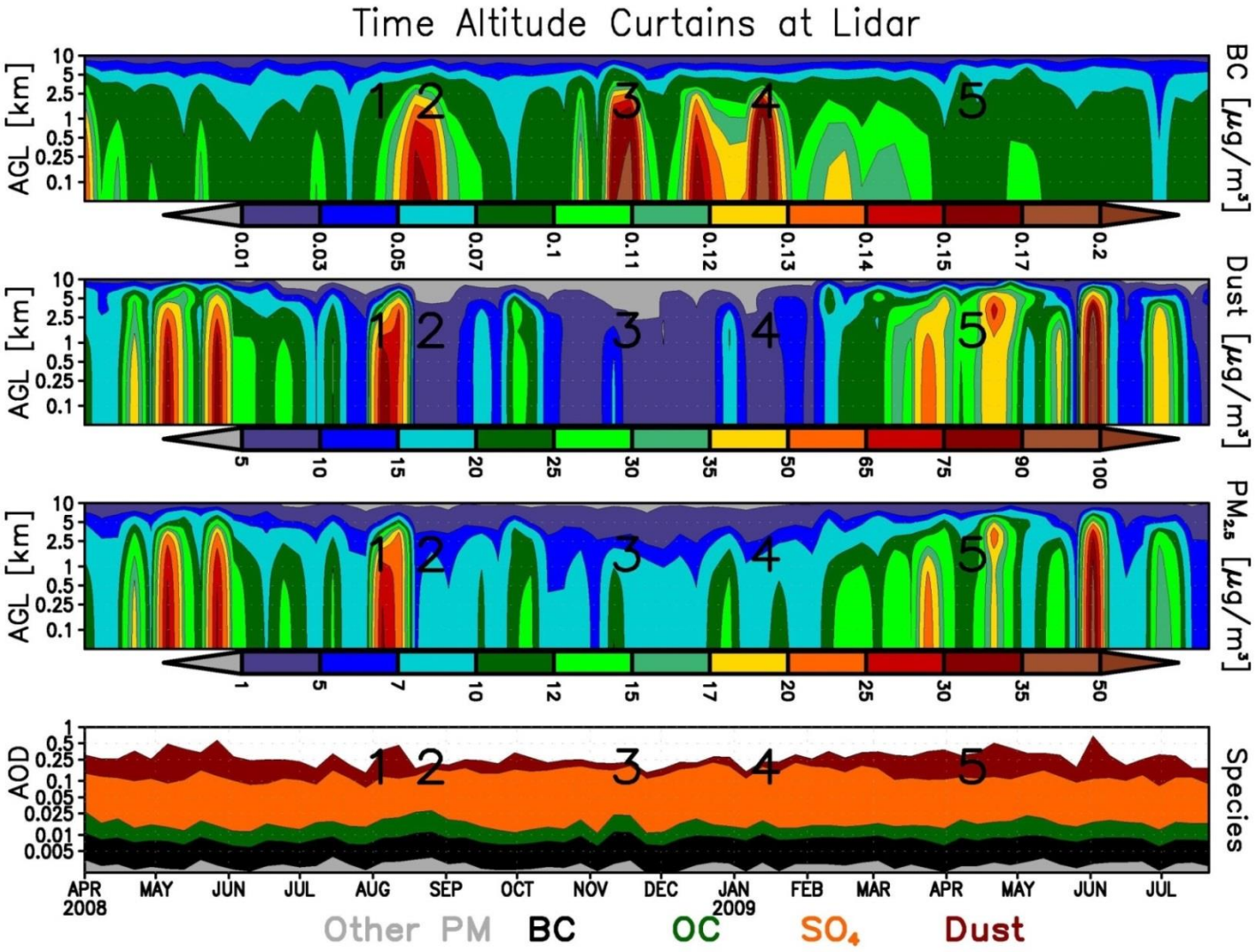
Time Altitude Curtains at Lidar
AGL [km]
BC [μg/m³]
Dust [μg/m³]
PM2.5 [μg/m³]
AOD
Species
1
2
3
4
5
APR 2008
MAY
JUN
JUL
AUG
SEP
OCT
NOV
DEC
JAN 2009
FEB
MAR
APR
MAY
JUN
JUL
Other PM
BC
OC
SO4
Dust

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.

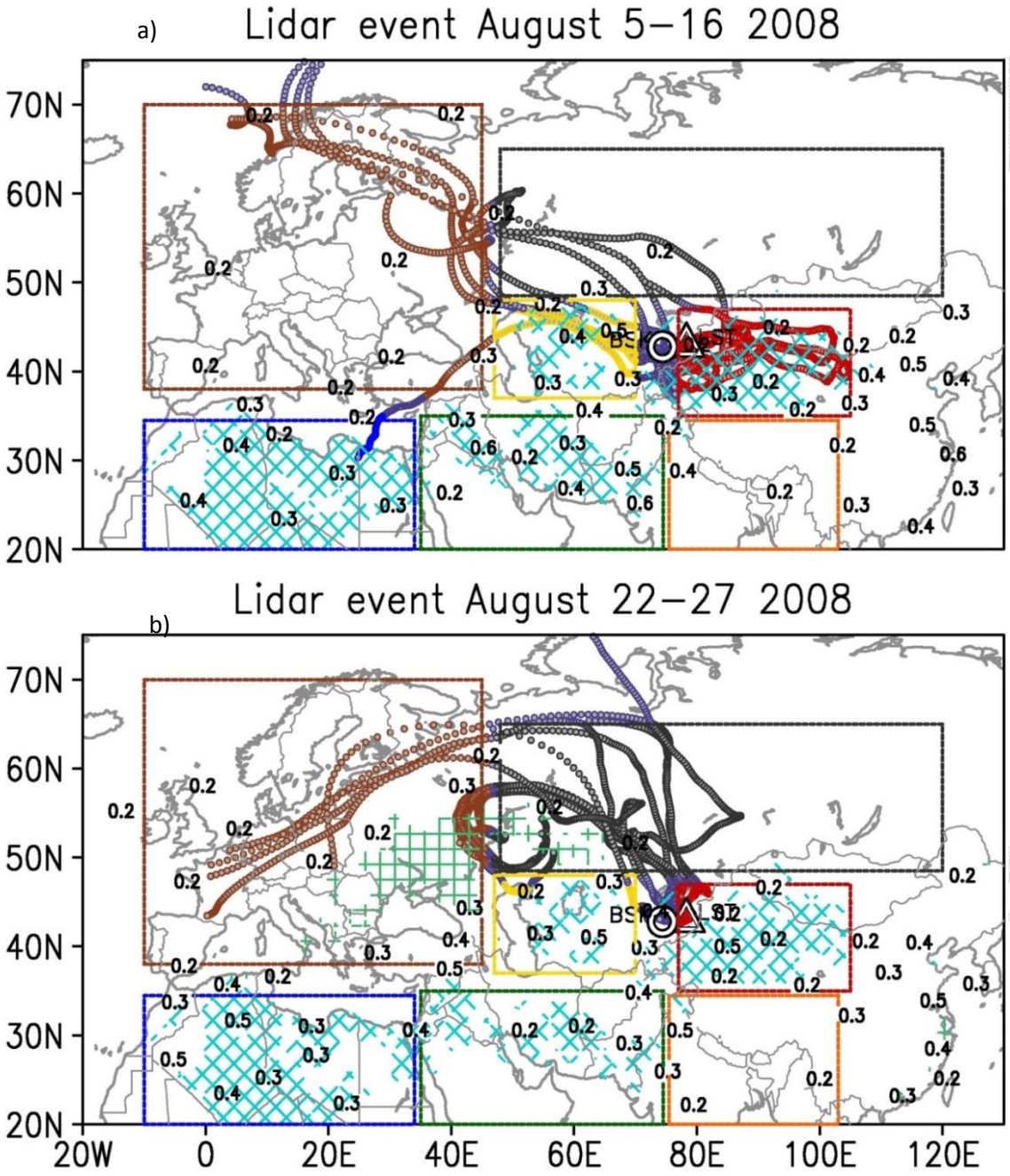

Fig. 14. Ten day air mass back trajectories for August 2008 (events 1 and 2 as denoted in Fig. 11) color coded by source regions. The blue diamond and green square hatched areas denote the natural dust and biomass burning emission sources while the number in black denote MODIS AOD contours averaged over 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,

black), Europe (brown), China (red) and South Asia (orange). Lidar denotes the LST (Lidar Station Teplokluchenka) site.

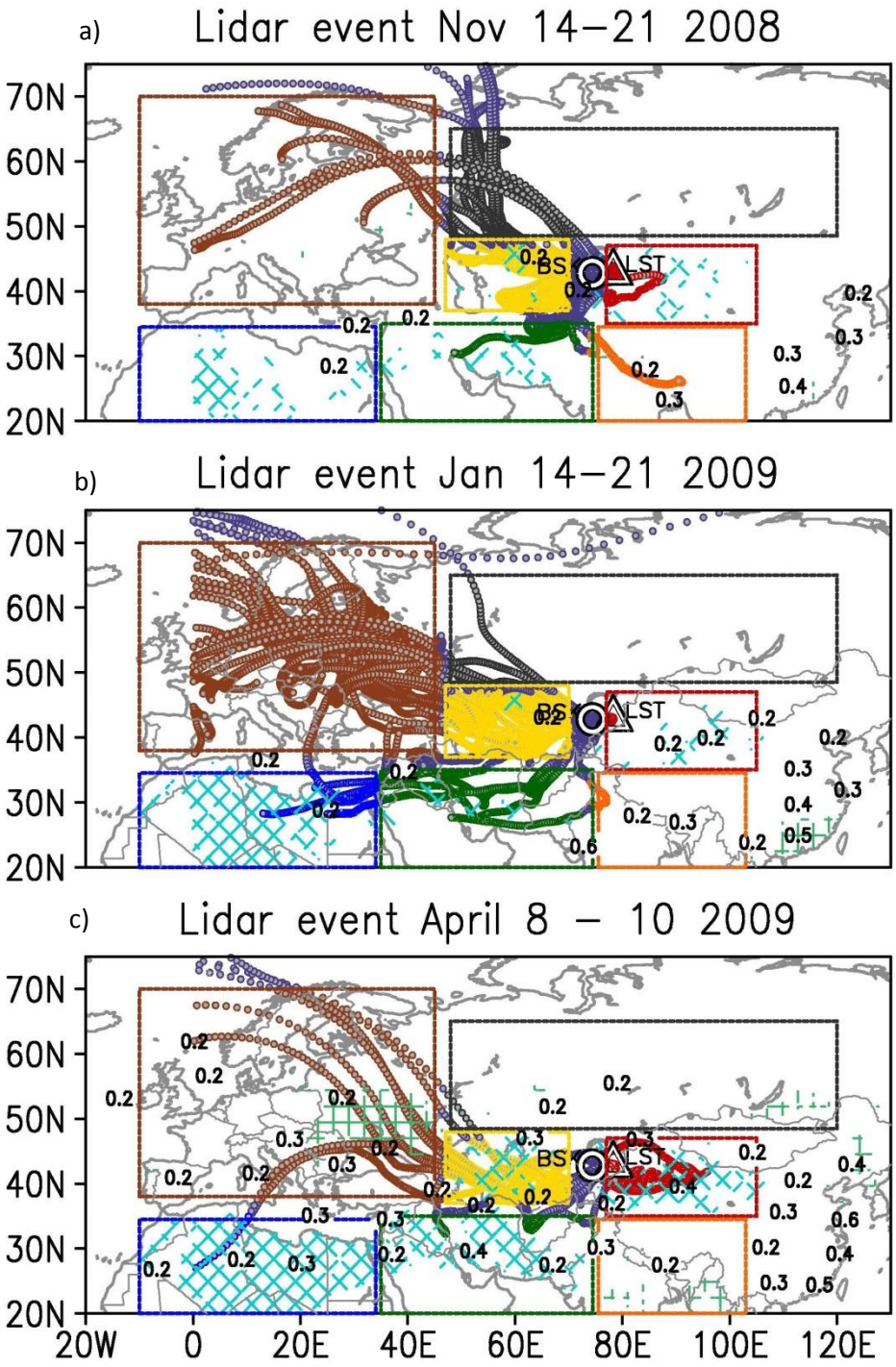

Fig. 15. Same as Fig. 14 but for November 2008, January 2009 and April 2009 (events 3, 4, 5 denoted in Fig. 13).

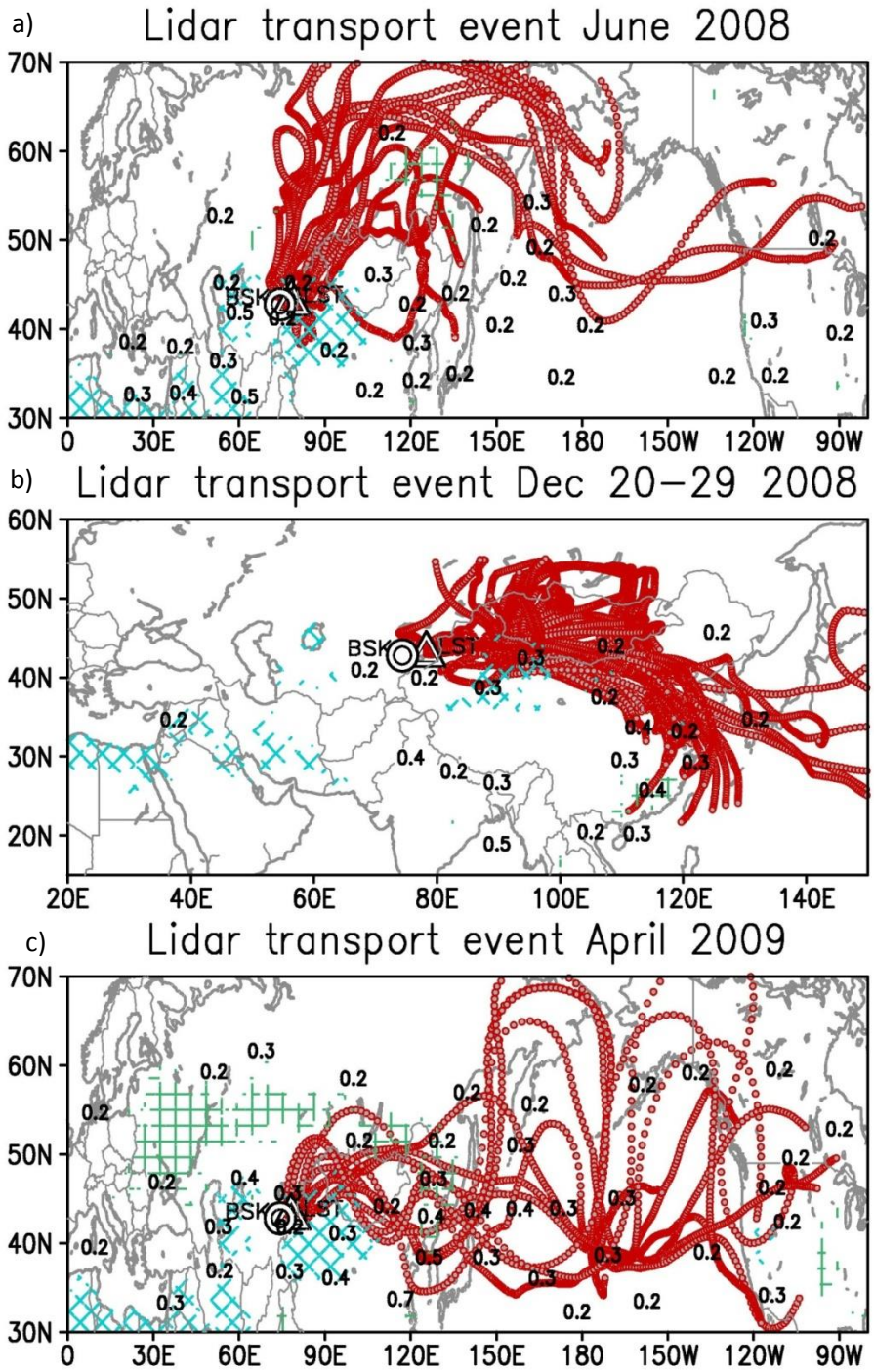

Fig. 16 Ten day air mass forward trajectories illustrating the seasonality in transport pathways out of CA for a) June 2008, b) December 2008, and c) April 2009. The blue diamond, green square hatched areas denote the natural dust and biomass burning emission sources while the number in black denote MODIS AOD contours, respectively, averaged over the event time

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

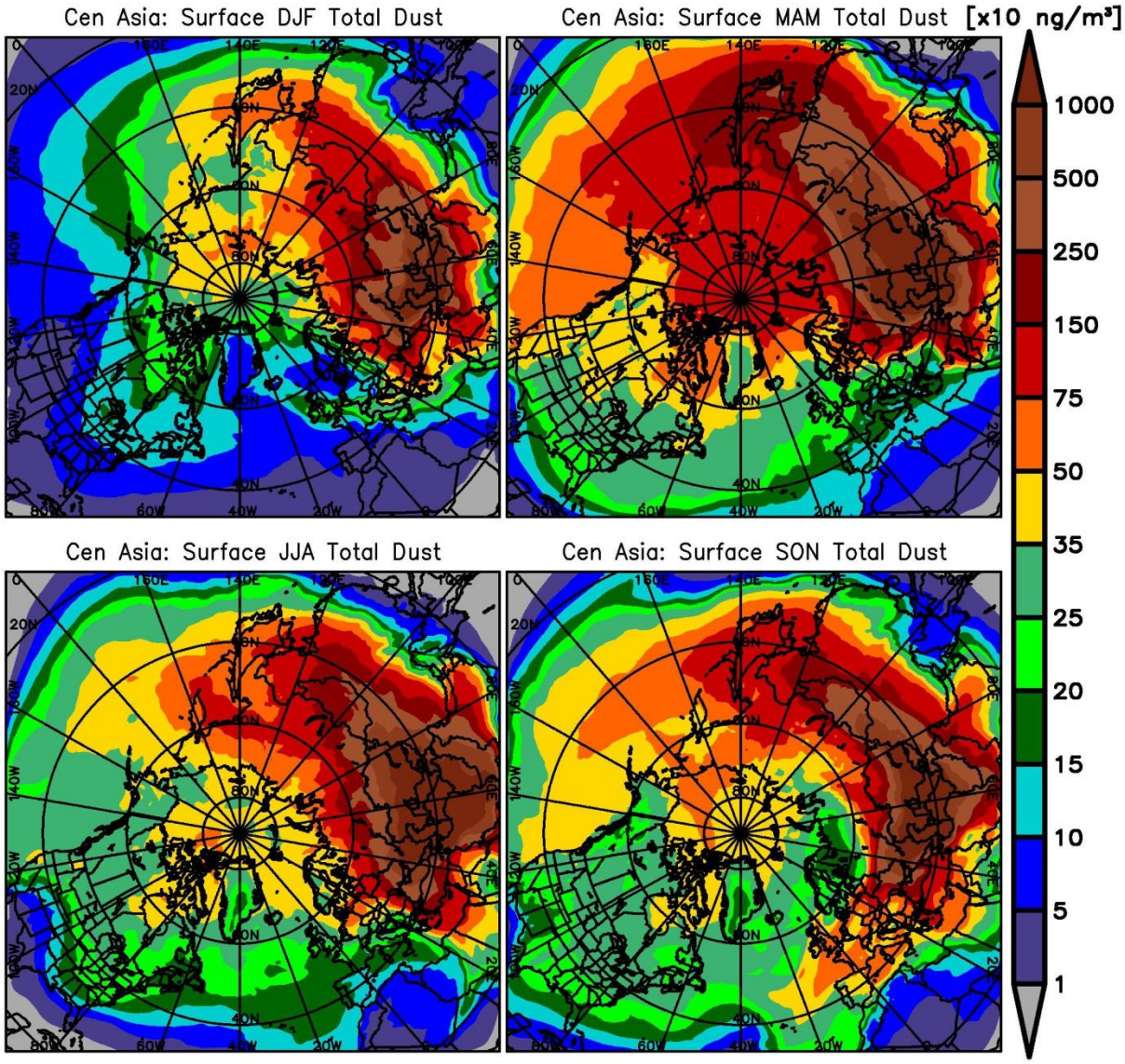

Fig. 17. Seasonally averaged surface total dust (PM_{10}) concentrations from Central Asia dust emissions. DJF (top left panel) denotes the average for the months of December, January and February. MAM (top right panel) denotes the average for months of March, April and May. JJA (bottom left panel) denotes the average for months of June, July and August while SON (bottom right panel) denotes average for months of September, October and November.

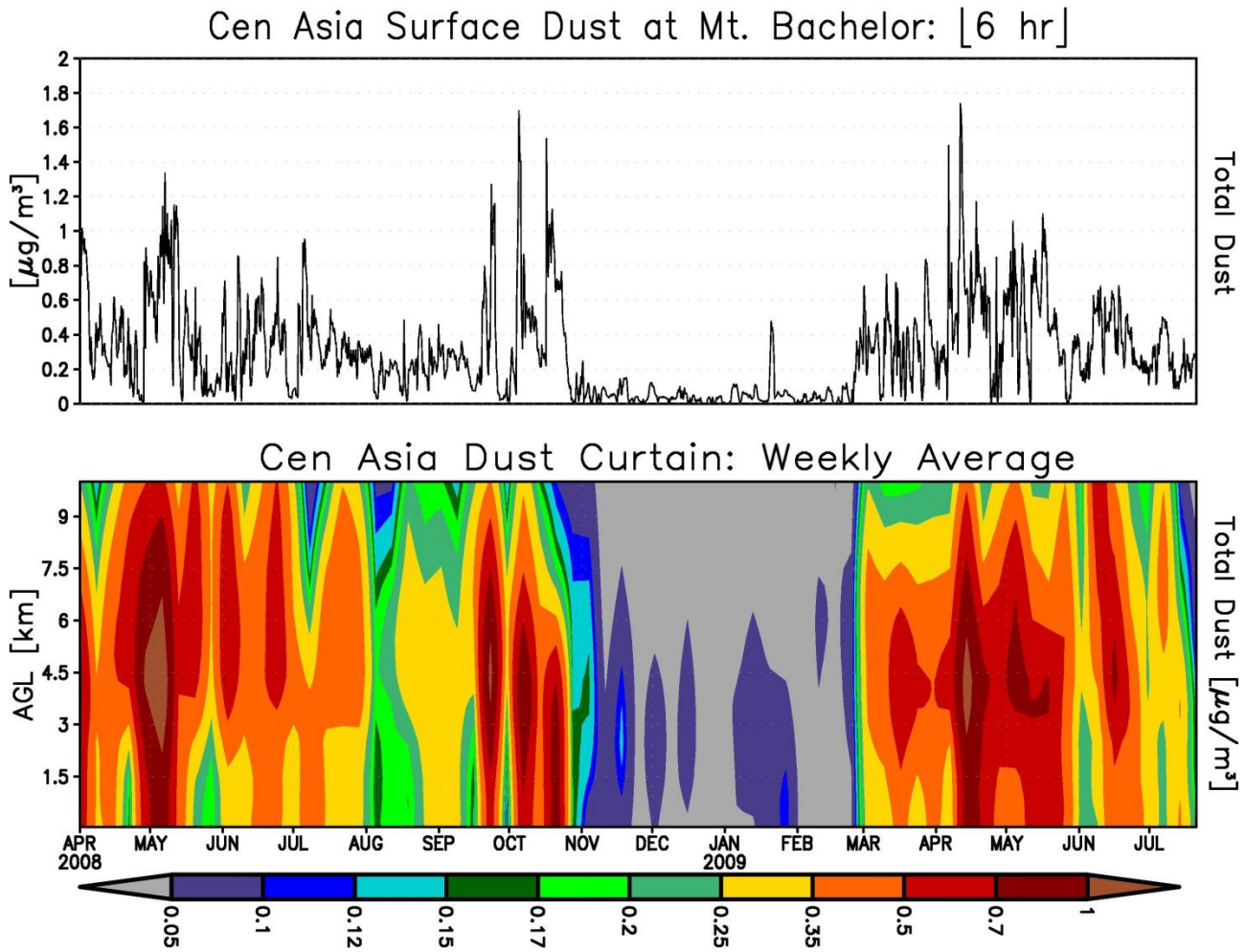

Fig. 18. Time series of predicted surface concentration at 6 h time step (top panel) and weekly averaged time altitude cross sections (bottom panel) of total dust (PM_{10}) from Central Asia dust emissions at Mt. Bachelor, Oregon.

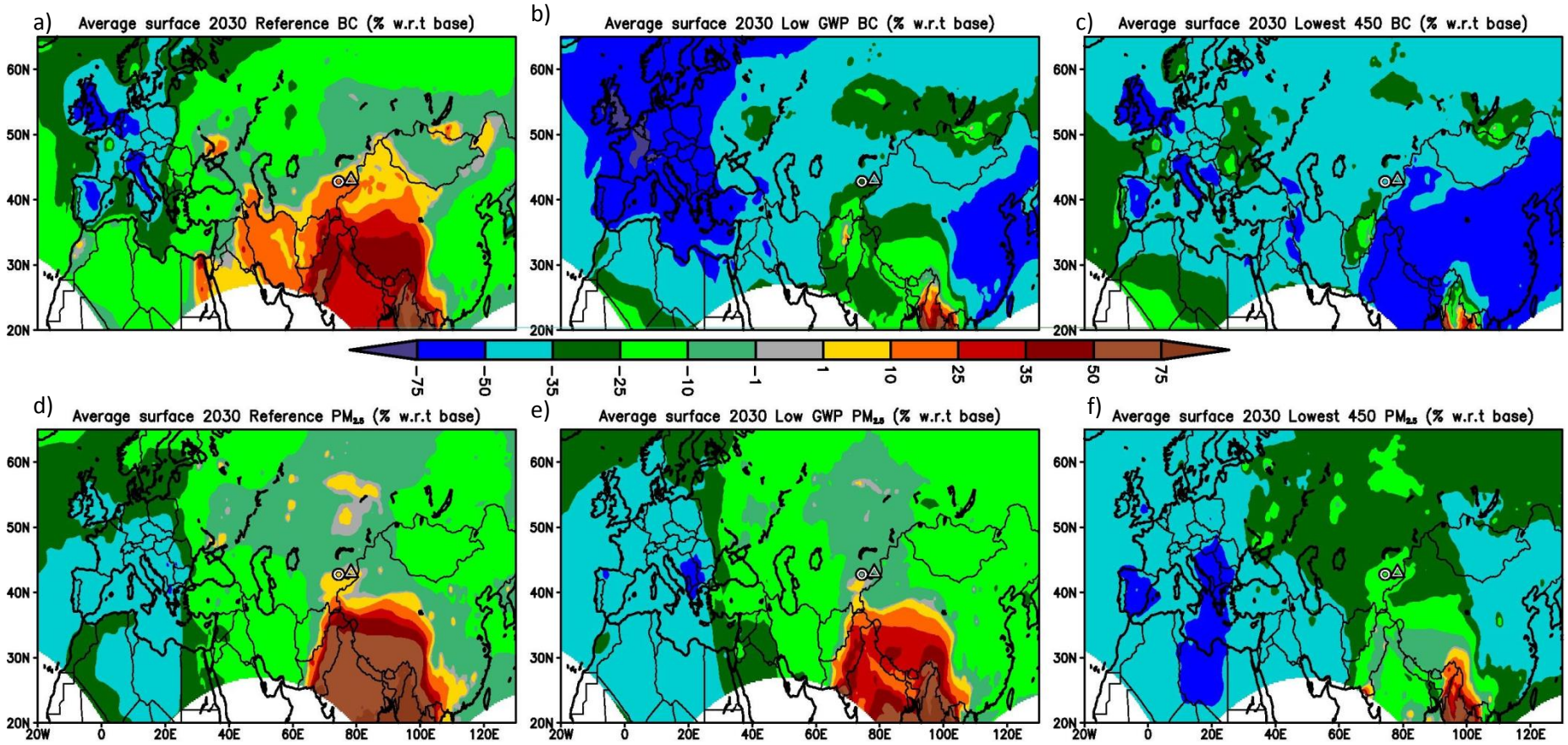

Fig. 19. Percent change in simulated period mean surface BC and $PM_{2.5}$ concentrations for future 2030 emission scenarios relative to the base year (2005) a),d) reference 2030, a),e) BC measures (low) and c),f) BC (lowest) and greenhouse gas measures aimed at keeping CO_2 levels below 450 ppm. Refer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