Reply to "interactive comment of anonymous Referee #2"

We thank the referee for providing meaningful comments on the manuscript. In the revised manuscript, we would like to provide more detailed explanations about uncertainties in the origin and transport of OCRB-dominant plumes. The impacts of non-OCRB sources on carbonaceous aerosol correlations will also be discussed explicitly. This study was based on several published papers that have addressed OCRB-dominant pollution episodes at the summit of Mt.Tai during the MTX2006 campaign. For example, calculations using the Community Multi-scale Air Quality Modeling System (CMAQ) demonstrated that OCRB emissions in CEC were responsible for 79% and 80% of BC and OC mass concentrations respectively at the summit of Mt. Tai. In the revised manuscript we will revisit several published results and provided more discussions about the uncertainties related to: (1) the origin of OCRB plumes which impacted observations at the summit of Mt.Tai, (2) Characteristics of OCRB plumes from the source regions and (3) the transport time of OCRB plumes calculated using the WRF-FLEXPART model. Detailed replies to the comments of referee are listed below.

Comment: Although the data set is valuable for evaluating the influences of OCRB on the distribution of carbonaceous aerosols in that region, the regression fitting method used is inadequate for the expected results by authors in getting an estimation of lifetime of carbonaceous aerosols from OCRB, because the ratios of carbonaceous aerosol over CO calculated from measurements are the contributions of all air masses coming from various sources at the time, the air masses could be fresh or aged, from OCRB or industrial sources, but the transport time is estimated only for OCRB sources in the specific regions.

Reply: As implied by the referee, mixing with outflowed urban pollutions could alter the carbonaceous aerosol correlations of OCRB plumes measured at the receptor, resulting in large uncertainties in estimating their lifetime. However, its influence is relatively small in this study. During high-pollution episodes from June 5–7 and June 12–13, OCRB emissions in CEC were responsible for 79 and 80% of BC and OC mass concentrations measured at the summit of Mt. Tai based on calculations using the Community Multi-scale Air Quality Modeling System (shown in Auxiliary Figure 1). Also, the contributions from OCRB sources ranged from 65–87% (BC) and from 64–93% (OC) for the targeted episodes in this study. Variations in carbonaceous aerosol mass concentrations during urban air pollution-dominated periods (after June 15) were well-captured by the simulations, implying that uncertainties in the urban emissions inventory (REASv1.0, Ohara et al., 2007) had a very limited impact on simulations in the OCRB-dominant case. We also simulated the mass concentrations of carbonaceous aerosols based on a bottom-up emissions inventory (INTEX-B 2006, Zhang et al., 2009) and a source-receptor-relationship (SRR) determined using the FLEXPART_WRF model. Detailed descriptions of the calculation

of the SRR using backward simulations have been published (Stohl et al., 2009). Simulations assumed that transported particles did not suffer any loss from dry deposition and cloud scavenging. Even so, approximately 78% of the BC mass measured at the site could not be explained by anthropogenic emissions (industry, transport, domestic heating and power plants). We attributed that mass to OCRB sources. This result was generally consistent with the simulations using CMAQ models. The ratios of $\Delta EC/\Delta CO$ and $\Delta OC/\Delta CO$ for OCRB plumes can be quantitatively estimated assuming that the carbonaceous aerosol correlations measured at the site were weighted linear combinations of urban and OCRB pollution (equation: $\frac{\Delta[x]}{\Delta CO}\Big|_{obs}$

 $f_{urban} \frac{\Delta[x]}{\Delta CO}\Big|_{urban} + f_{OCRB} \frac{\Delta[x]}{\Delta CO}\Big|_{OCRB}$, where f represents the respective contribution and x refers to EC or OC. We assumed that the Δ EC/ Δ CO and Δ OC/ Δ CO ratios for urban pollution did not change significantly over 2 days). The ratios of Δ EC/ Δ CO (6.5 ng/m³/ppbv, Pan et al., 2011) and Δ OC/ Δ CO (23.5 ng/m³/ppbv) for urban pollution in CEC were adopted. The results demonstrated that the Δ EC/ Δ CO and Δ OC/ Δ CO ratios were 12 and 23% higher, respectively, than the observations. Applying the revised ratios to the calculation, we found that the lifetimes of EC and OC were generally consistent with the previous results, with means of 111.5 h (4.6 days) for EC and 25.8 h (1.1 days) for OC. Large variabilities in Δ EC/ Δ CO ratios for urban pollution because of diverse emission sources in different locations. To justify the results, we performed sensitivity tests using different Δ EC/ Δ CO ratios (4.8 ng/m³/ppbv in Beijing, reported by Han et al., (2009); 9.8 ng/m³/ppbv in Bangkok, reported by Sahu et al., (2011)) and Δ OC/ Δ CO ratios (14–84 ng/m³/ppbv, reported by Maria et al., (2003)) of urban pollution. We found that the estimated lifetimes of EC and OC had less than 10% variance, even if Δ EC/ Δ CO and Δ OC/ Δ CO ratios for urban pollution changed by a factor of 2. This suggests that the influence of urban pollution was limited.

In the present study, potential source regions of OCRB plumes with respect to the observations at the summit of Mt.Tai were estimated by multiplying OCRB emission fluxes (in units of kg/s) in each grid cell by the corresponding source-receptor-relationship (SRR as in Auxiliary Figure 2b, in units of s/kg). Here, OCRB emission fluxes were assumed to be proportional to the number of hotspots detected by MODIS (Auxiliary Figure 2a) in the target grid. SRR, calculated using the FLEXPART_WRF model, was proportional to the particle residence time in a particular grid cell. Herein, we divided the areas where burning events occurred into five different regions (A: $33-34^\circ$ N, $112-114^\circ$ E; B: $33-34^\circ$ N, $114-116^\circ$ E; C: $32-35^\circ$ N, $118-121^\circ$ E; D: $34-35^\circ$ N, $116-118^\circ$ E; S: $33-34^\circ$ N, $116-118^\circ$ E). The estimated potential OCRB source region (Auxiliary Figure 2c) was concentrated in region S (the region we used in the original manuscript, 75.3% contribution) based on observations at the summit of Mt.Tai, significantly higher than those from other regions (A (1.6%), B (10.6%), C (4.6%) and D (7.7%)). A sensitivity test of SRR employing different height criteria (< 500 m above the ground, instead of <100 m) in the base case also resulted in a similar contribution (74.4%) from region S.

The transport time uncertainties were also re-estimated by comparing results from the following scenarios: (Scenario 1) Releasing particles from region S homogeneously in time and space (as a base case in the previous manuscript). (Scenario 2) Temporal variations in emissions were accounted for in the number of hotspots detected by MODIS. Data gaps due to the overpass of satellites were filled using linear interpolations. (Scenarios 3-6) Region S was divided into four smaller sub-regions (S1: 33-34°N, 116-117°E; S2: 33-34°N, 117-118°E; S3: 33.5-34°N, 116–118°E; S4: 33–33.5°N, 116–118°E) and the particles were released only from one of the sub-regions at a time. As shown in Auxiliary Figure 3a, a good correlation (intercept = -0.2 h, slope = 1.1, r = 0.93) was obtained for scenarios 1 and 2, indicating that temporal variations in the emission strengths of OCRB had limited impact (variability ~10%) on the estimation of plume transport times. Estimated transport times of plumes from the sub-regions (S1, S2, S3 and S4) were approximately the same as those from region S (Auxiliary Figure 3b), with slopes ranging from 0.89–0.99 and intercepts ranging from 0.7–1.4 h, implying that the estimated transport time was almost independent of the area settings. Correlations among the transport times estimated for sub-regions S1, S2, S3 and S4 demonstrated that the maximum uncertainty related with the OCRB source regions was ~14% (Auxiliary Figures 3c and 3d), and there was approximately a 4h time lag (intercept of best linear fitting). These variations only slightly modified the estimated lifetimes and did not change the main conclusions of this paper. As shown in Auxiliary Figure 4, lifetime of EC was ranging 98.4-136.9 hours (4.1-5.7 days), and lifetime of OC was found to be 28.0-44.2 hours (1.2-1.8 days), comparable to previous results (4.3 days for EC, 1.1 days for OC).

Specifics 1: The baseline of carbonaceous aerosol and CO should be given.

Reply: As replied in the referee's previous comment, the baseline of carbonaceous aerosols and CO refers to the contributions from non-OCRB sources during the measurement period. As discussed above, less than 10% uncertainty was introduced to the estimates of the lifetimes of EC and OC because of impacts from urban pollution. In the revised manuscript, we will add more discussion on this issue.

Specifics 2: When comparing the EC data or the ratios of EC/CO from different investigators, authors should keep in mind that EC analyzed with Sunset OCEC analyzer may be different from other methods, as different instruments/methods may give different readings for BC or EC.

Reply: The referee makes an excellent point. As mentioned, systematic biases of BC measurements by different instruments (Sunset EC/OC analyzer with IMPROVE/NIOSH temperature protocol, multi-angle absorption photometer (MAAP), Aethalometer) have been studied and published (Kanaya et al., 2008) in this special issue. In the present study, we reported Δ EC/ Δ CO ratios from OCRB sources and investigated their dependence on transport time. Actually, uncertainties related to systematic biases inherent in the measurement methods were

investigated in the text. Calculations using $\Delta BC_e/\Delta CO$ and $\Delta BC_e/\Delta CO_2$ ratios (BC mass concentrations were determined using a MAAP instrument) demonstrated that the estimated lifetimes of BC were 116 h (4.8 days) and 104 h (4.3 days), respectively. The values were almost identical to those estimated based on EC, implying that any additional uncertainty was not large.

Specifics 3: P14373, Line5-6: "As shown in Fig. 2a and b, the upward transport pattern of OCRB plumes was excellently captured by the model", the comparison of model results with observations is needed.

Reply: Comparison of modeled and observed carbonaceous aerosol mass concentrations at the summit of Mt.Tai has been performed and published (Yamaji et al., 2010). Figures 2a and b in the manuscript were used to show the transport pattern of an OCRB plume from two snapshots of FLEXPART simulations. Upon the comment from the referee, we realized that this expression *"the upward transport pattern of OCRB plumes was excellently captured by the model"* could have been misleading. In the revised manuscript, we revised this section and reported modeled mass concentration of carbonaceous aerosols on the basis of a bottom-up emissions inventory (INTEX-B 2006, Zhang et al., 2009) and a source-receptor-relationship (SRR) determined using the FLEXPART_WRF model. Detailed descriptions of the calculation of SRR with backward simulations have been published (Stohl et al., 2009).

Some grammatical errors: grammatical and spelling errors in the text, such as "on basis of (page 14367, line 4)", symbols in Eq. (1) and Eq. (3) (Page 14373), "20<T30h (Page 14374)

Reply: We will correct the grammatical errors and thoroughly edit the manuscript to avoid any possible mistakes.

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Auxiliary Figures

Auxiliary Figure 1. Simulated mass concentrations of carbonaceous aerosols during the field campaign. This plot was published by Yamaji et al., 2010.



Auxiliary Figure 2. (a) Spatial distribution of hotspots detected by MODIS. (b) Source-receptorrelationship in a footprint region calculated by backward simulation of the FLEXPART_WRF model. (c) Potential source region (in arbitrary units) of OCRB plumes observed at the summit of Mt.Tai.



Auxiliary Figure 3. Scatterplots and their linear regressions for transport times under different emission scenarios.



Auxiliary Figure 4. Fitting results for the relationships between variations in $\Delta EC/\Delta CO$, $\Delta OC/\Delta CO$ and $\Delta OC/\Delta EC$ ratios and transport time for scenarios 3(a–c), scenarios 4(d–f), scenarios 5 (g–i) and scenarios 6 (j–l). Four figures in the rightmost panel indicate the source regions where tracer particles were released by the FLEXPART_WRF model. The gray lines in the figure represent the 90% confidence intervals.