

Exploring wintertime regional haze in Northeast China: role of coal and biomass burning

Zhang et al.,

We are grateful for Referee#2's comments. These comments are all valuable and helpful for improving our paper. We answered the comments carefully and have made corrections in the submitted manuscript. The corrections and the responses are as following:

In the revised manuscript and supplement, the red color was marked as the revised places.

General comment

1. On the other hand, in the discussion, I would argue another possibility for the particle source in the Haze-I, in addition to the possible local source that the authors discuss in this manuscript. The 24-hour back-trajectories in Fig-S4 show possible contributions from the Beijing area during the Haze-I (heavy) period. The air mass with Beijing pollutants seems to be brought by a low-pressure system on November 4-5 (Fig. S3). Then the air mass changed to Northwest with biomass burning pollutants. If this hypothesis is reasonable, the discussion that the changes in particle composition were due to regional atmospheric chemical reactions (line 427-429) needs to be revised. I suggest to consider the possibility and revised the discussion if needed.

Reply: We appreciated the reviewer's comments and carefully considered them. We added three figures in Figure S5, which shows air mass backward trajectories during the haze period and concentration-weighted trajectory (CWT) plots of PM_{2.5} on 50 m and 500 m heights during Haze-I event. Figure S5b shows that the contribution of PM_{2.5} was high from Shenyang city and its surrounding areas and rather low from other regions during the moderate Haze-I period. Figures S5c-d show that compared with transport from Beijing-Tianjin-Hebei region, local emission and transformation mainly contributed to the heavy Haze-I formation. Therefore, we could determine that PM_{2.5} at the urban and mountain sites were mainly from local emission and transformation during the Haze-I event based on the CWT plots. The detailed discussion as follows.

P17 L425-431: "Through the meteorological data (Figures S4a-b), air mass backward trajectory (Figure 7c), and concentration-weighted trajectory (CWT) plots of PM_{2.5}

(Figures S5b-d) analyses, we inferred that the air quality at the two sampling sites during the Haze-I event was mostly influenced by Shenyang city and its nearby surrounding emissions, although transport from Beijing-Tianjin-Hebei region slightly contributed to the heavy Haze-I formation on 4 DT November.”

P17 L436-438: “Figures S5c-d further indicate that compared with transport from Beijing-Tianjin-Hebei region, local secondary transformation was one major factor to cause the heavy Haze-I formation.”

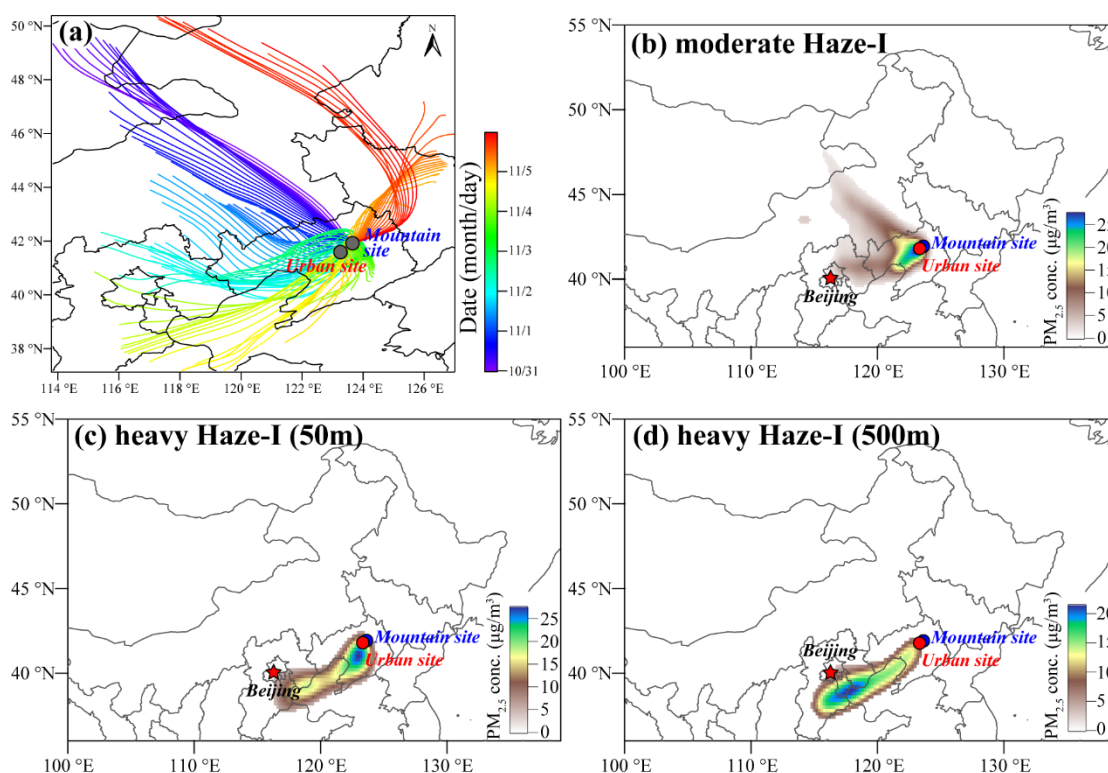


Figure S5. (a) 24-h air mass backward trajectories on 500 m height before arriving at Shenyang during 31 October-5 November. Concentration-weighted trajectory (CWT) plots of PM_{2.5} at the urban and mountain sites during the Haze-I event: (b) moderate Haze-I on 500 m height; (c-d) heavy Haze-I on 50 m and 500 m heights.

Specific comments

- Line 32 and 36 (S-OM and K-OM): Spell out.

Reply: We explained S-OM and K-OM particles as follows.

P2 L32-33: “S-rich internally mixed with OM (named as S-OM) particles”

P2 L38: “K-rich internally mixed with OM (named as K-OM) particles”

3. Line 43-45: grammatically something wrong.

Reply: We revised this sentence as follows.

P2-3 L45-48: “Our study highlights that large numbers of light-absorbing tarballs significantly contribute to winter haze formation in Northeast China and they should be further considered in climate models.”

4. Line 245: Define DT and NT.

Reply: We thanked the reviewer. We defined DT and NT in section 2.1.

P7 L154-156: “To better explore the variation of PM_{2.5} composition and individual particles, we collected the daytime (DT, 8:30-20:00 (local time)) PM_{2.5} and nighttime (NT, 20:30-8:00 (next day)) PM_{2.5}...”

5. Line 364-367: Please see my general comment. It seems to me that the composition change is mainly due to the air mass change rather than a process of secondary sulfate and nitrate formations.

Reply: We added figures of CWT plots of PM_{2.5} in Figure S5 to indicate the geographic origins of haze particles. Figures S5c-d show that compared with transport from Beijing-Tianjin-Hebei region, local emission and transformation mainly contributed to the heavy Haze-I formation.

We revised this sentence as follows.

P15 L375-378: “Figures 5a-b show that SO₄²⁻/EC and NO₃⁻/EC factors reached their maximum values at the two sampling sites during the heavy Haze-I event, suggesting that secondary sulfates and nitrates significantly increased during Haze-I evolution (details in Section 4.1).”

6. Line 391-392: Please indicate spherical OM and domelike OM in the figure 6 c-d. I found these OM particles in the clean day in figure 6b but not in figure 6 c-d.

Reply: We carefully considered the comment. To make the figure clearer, we indicated spherical OM and domelike OM with red arrows and green arrows in Figures 6a-f as

below.

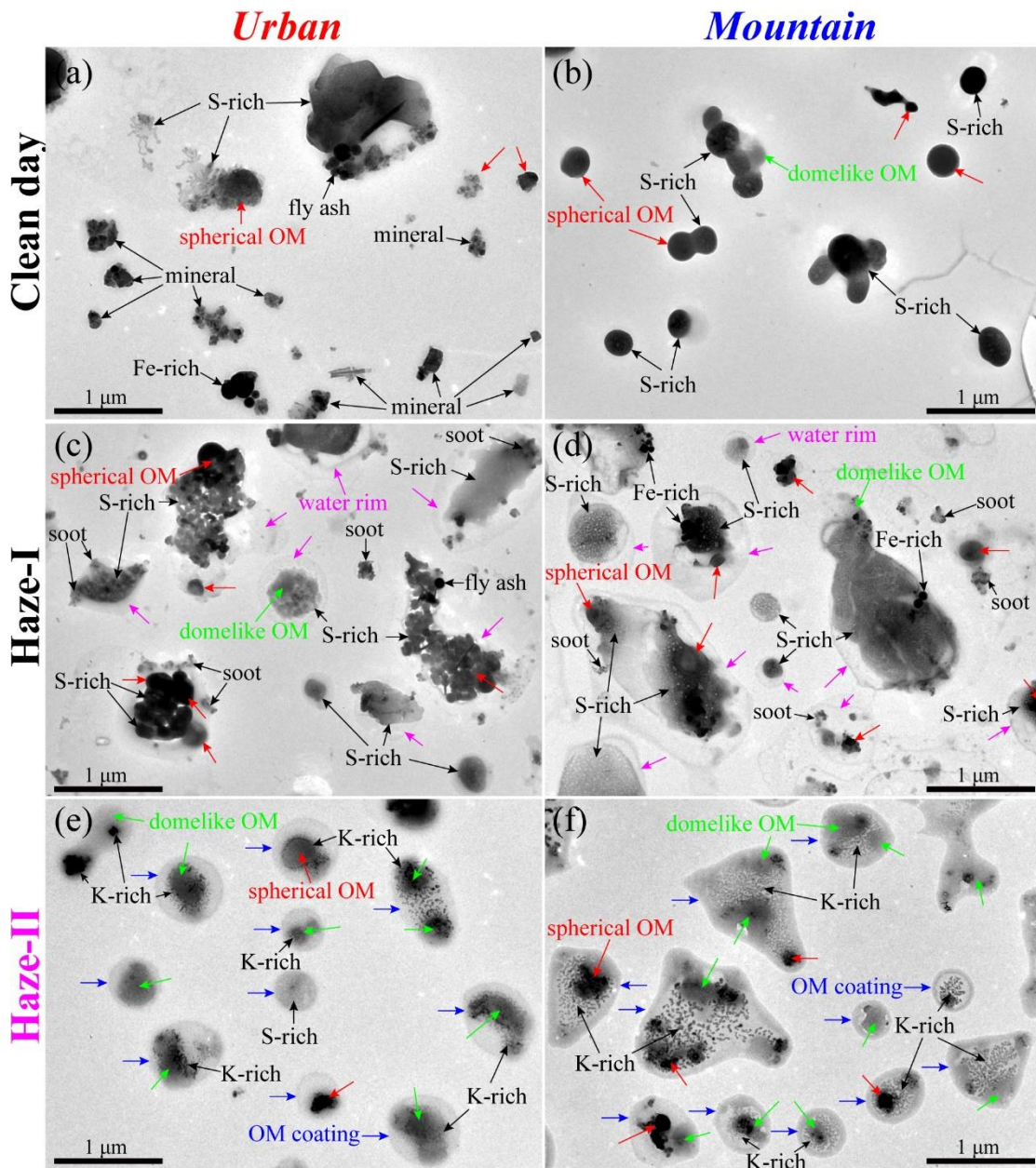


Figure 6. Typical TEM images of individual aerosol particles at low magnification at the urban and mountain sites: (a-b) clean day; (c-d) Haze-I; (e-f) Haze-II. The red, green, blue, and pink arrows represent spherical OM, domelike OM, OM coating, and water rim, respectively.

7. Line 405-410 (figure 7 and S4): Are the back trajectories in Fig. 7 and S4 the same? The Fig. 7 suggests local source but the S4 shows long-range transportation (possibly from Beijing area?). Figure S2 also suggests broader regional pollution events on November 4 rather than a local event. Please also see my general comment.

Reply: Yes, the backward trajectories in Figure 7 and Figure S5a are same. Figure S5a

shows hourly backward trajectories during the haze period, and Figure 7c shows two typical backward trajectories calculated from 8:00 (local time) on 3 November during the moderate Haze-I event. We added figures of CWT plots of PM_{2.5} in Figures S5b-d to indicate the geographic origins of haze particles. The results show that compared with transport from Beijing-Tianjin-Hebei region, local emission and transformation mainly contributed to the heavy Haze-I formation.

We revised some discussion about the heavy Haze-I formation as follows.

P17 436-438: “Figures S5c-d further indicate that compared with transport from Beijing-Tianjin-Hebei region, local secondary transformation was one major factor to cause the heavy Haze-I formation.”

P18 L453-456: “Therefore, we concluded that atmospheric heterogeneous reactions under high RH (> 70%) and local primary emissions together induced the heavy Haze-I formation in Northeast China.”

8. Line 427-429 and line 570-572: This statement also needs to be reconsidered if my general comment is reasonable.

Reply: Please see our reply in Q1. Figures S5c-d show that local emission and transformation mainly contributed to the heavy Haze-I formation. We revised these two sentences as below.

P18 L453-456: “Therefore, we concluded that atmospheric heterogeneous reactions under high RH (> 70%) and local primary emissions together induced the heavy Haze-I formation in Northeast China.”

P23-24 L606-608: “Production of secondary aerosols via heterogeneous reactions at high RH (> 70%) and local primary emissions caused the transition from the moderate Haze-I to the heavy Haze-I events.”

9. Line 486: This section includes not only Atmospheric implications but also broader discussion such as health issues. Please reconsider the title.

Reply: Thanks. We revised the section title as follows.

“Atmospheric and health implications”

10. Line 503-526: I do not see spherical and domelike OM particles in Figure 6c-f when biomass-burning or coal burning were the dominant sources. Please indicate which particles are spherical and domelike OM particles. When were the particles in Fig 3 collected? The figure shows spherical and domelike OM particles without sampling periods.

Reply: We indicated spherical OM and domelike OM with red arrows and green arrows in Figures 6a-f. The particles in Figures 3a-h, 3k and Figures 3i-j were collected during the Haze-I and Haze-II events, respectively. We added the sampling periods of particles in Figure 3 caption.

P35 L1015-1016: “Particles in (a-h, k) and (i-j) were collected during the Haze-I and Haze-II events, respectively.”

11. Figure 5: Please indicate the Y-axis label on the left.

Reply: We added the left Y-axis label in Figure 5 as below.

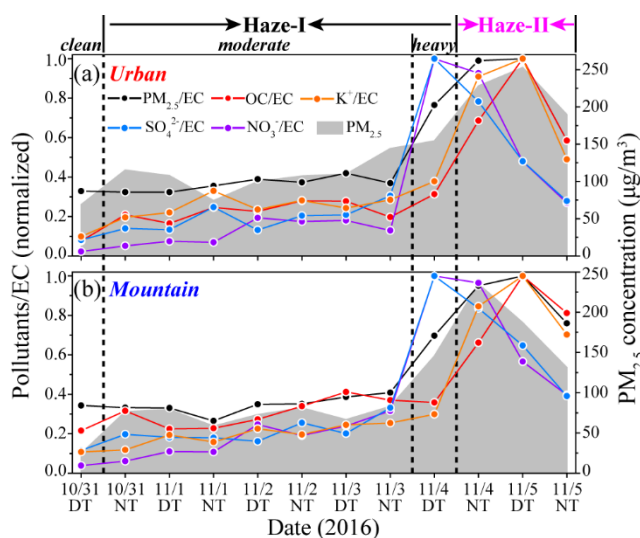


Figure 5. Variation in $PM_{2.5}/EC$, OC/EC , SO_4^{2-}/EC , NO_3^-/EC , and K^+/EC factors at the urban site (a) and mountain site (b). These factors are normalized.

12. Table S1: please indicate the error range (or standard deviation).

Reply: We added the standard deviation in Table S2.

Table S2. The ratios of OC/EC in Northeast China and North China Plain (NCP) during winter haze days.

Sampling site	Period	OC/EC	References
Urban site	31/10-4/11/2016 (Haze-I)	8.0±1.5	This study
Urban site	4-5/11/2016 (Haze-II)	25.4±5.9	This study
Mountain site	31/10-4/11/2016 (Haze-I)	10.6±2.2	This study
Mountain site	4-5/11/2016 (Haze-II)	27.9±4.7	This study
Beijing city	16-19/1/2010	4.5	Zhao et al. (2013)
Jinan city	13-23/12/2014	5.5	Chen et al. (2017)
Tianjin city	10-12/1/2013	5.3	Han et al. (2014)