Response to comments

Reviewer: 2

Comments:

This manuscript presents a field observational study on the measurement of lead-rich particles using a single-particle aerosol mass spectrometer (SPAMS) in Beijing. The study draws interesting conclusions that contribute to deepening our understanding of the seasonal distribution, formation mechanisms, and influencing factors of toxic lead particles following the coal-to-gas conversion. Although the study is localized to Beijing, the findings have broader implications, especially in terms of human intervention and policy making related to lead pollution. The reduction of lead in the atmosphere due to the coal-to-gas conversion can serve as an important case study for other regions facing similar air quality challenges. I recommend publication of the manuscript after the authors address the following comments:

We sincerely appreciate the reviewer's thoughtful and constructive feedback on our manuscript. Their careful review and insightful comments have been invaluable in improving our work. Below, we have provided detailed responses to the specific points raised, incorporating their suggestions to enhance the quality of the manuscript.

1. The manuscript should include a clearer explanation of how the internal and external mixing states of lead particles were determined. It would be helpful to expand on the methodology used to identify and categorize these mixing states for better transparency.

Thank you for bringing up our mistake. Our results could only indicate lead are mixed with nitrate, but cannot distinguish between internal mixing and external mixing. We have added the criteria for determining the mixture of lead and nitrate in section 2.2 of the manuscript.

"The mixed states of lead, nitrate, and other ions are identified by detecting their characteristic peaks using single-particle mass spectrometry." has been added to the revised manuscript. Please refer to Lines 120–121 of the revised manuscript.

2. Some sections of the manuscript contain complex phrasing that could benefit from greater clarity. For example, certain sentences related to the comparison of Pb-rich particle types between heating and non-heating periods are dense and need to be simplified for better readability. Additionally, the explanation of the factors influencing Pb nitrate formation could be more explicit.

We agree with the comment. We have reviewed the entire manuscript and revised the complex phrasing to make it clearer. Please refer to Lines 265–274, 283–285 and 306–309 of the revised manuscript.

We have replaced complex sentences related to the types of lead-rich particles during heating and non-heating periods with simpler sentences to improve readability, as shown in the following text:

"In this study, about 60% of the Pb-rich particles are mixed with chloride and oxygen, and this proportion was similar during the heating period and non-heating period in spring, and the heating period in winter (Fig. 5). The mixing ratio of Pb-rich particles and sulfate particles during the spring heating period is about 10% higher than that during the non-heating Pb-rich particles, which may be affected by the increase in sulfur dioxide concentration during the heating period (Table S1)." have been revised as"In this study, about 60% of the Pb-rich particles are found to mixed with chloride and oxygen. This proportion remain consistent across the heating and non-heating periods in spring, as well as the heating period in winter (Fig. 5). However, the mixing ratio of Pb-rich particles with sulfate is about 10% higher during the spring heating period compared to the non-heating period. This increase is likely influenced by the sulfur dioxide concentrations (Table S1).". Please refer to Lines 229–232 of the revised manuscript.

"As shown in Fig. S9, Pb-N particles increase with the increase of particle size, with more than 85% of particles distributed between 0.5 and 1.0 μm, indicating that most of the lead nitrate comes from the secondary generation of Pb-containing particles during transportation."have been revised as "As shown in Fig. S9, the number fractions of Pb-N particles rises with increasing particle size, with over 85% of these particles found within the size range of 0.5 to 1.0 μm. This distribution suggests that the majority of lead nitrate originates from the secondary formation of Pb-containing particles during their transport." Please refer to Lines 238–241 of the revised manuscript.

Thanks for the comment. More discussion on how factors such as temperature and relative humidity affect the formation process of lead nitrate has been added to the manuscript.

"As a precursor of nitrate, NO_2 participates in the formation of lead nitrate both during the day and at night. A significant positive correlation ($r = 0.88$, $p < 0.01$) between the RH of nitrate and NO_2 was observed (Peng et al., 2020). Temperature, relative humidity, and light intensity indirectly affect the formation of lead nitrate by influencing the formation of nitrate. As suggested by previous studies, the heterogeneous hydrolysis of N_2O_5 under high RH plays a crucial role in particulate nitrate formation and significantly contributed to the elevated fine nitrate during nighttime (Zhang et al., 2017). Solar radiation affects the formation of OH radicals, and OH radicals are oxidants that transform NO_2 into HNO_3, so a high concentration of OH radicals is conducive to the formation of HNO_3. At the same time, higher temperature is also conducive to NO_2 and OH reactions (Slater et al., 2019). Specifically, the number fractions of Pb-N particles decreases with increasing T, especially when the temperature exceeds 30 ℃, which may be an indirect result of the decomposition of nitrate particles caused by high temperature (Song et al., 2019; Luo et al., 2020)." Please refer to Lines 259–274 of the revised manuscript.

Wang, H.C., Lu, K.D., Chen, X.R., Zhu, Q.D., Chen, Q., Guo, S., Jiang, M.Q., Li, X., Shang, D.J., Tan, Z.F., Wu, Y.S., Wu, Z.J., Zou, Q., Zheng, Y., Zeng, L.M., Zhu, T., Hu, M. Zhang, Y.H.: High N_2O_5 concentrations observed in urban Beijing: implications of a large nitrate formation pathway. Environ. Sci. Tech. Let. 4, 416–420, DOI: 10.1021/acs.estlett.7b00341, 2017.

3. **Lines 120-123:** “They are named potassium-sodium (K-Na), K-Na internally mixed with Fe (K-Na-Fe), K-Na internally mixed with Cu (K-Na-Cu), K-Na internally mixed with Zn (K-Na-Zn), K-Na internally mixed with elemental carbon (K-Na-EC), and potassium internally mixed with organic carbon (K-OC).”

The sentence has been revised as "They are named potassium-sodium (K-Na), K-Na mixed with Fe (K-Na-Fe), K-Na mixed with Cu (K-Na-Cu), K-Na mixed with Zn (K-Na-Zn), K-Na mixed with elemental carbon (K-Na-EC), and potassium mixed with organic carbon (K-OC).". Please refer to Lines 121–123 of the revised manuscript.

4. **Lines 124-131:** Although the method for selecting lead nitrate used in this manuscript has been employed in previous research, I wonder if this selection method could potentially underestimate the content of lead nitrate. Could this affect the conclusions of the study?

We sincerely appreciate the reviewer’s insightful comment regarding the potential underestimation of lead nitrate content due to the selection method employed in our study. We acknowledge that the method for selecting lead nitrate, while consistent with previous research, may have limitations in fully capturing the variability or total content of lead nitrate in certain scenarios.

To address this concern, we have revisited our methodology and conducted additional analyses to evaluate the potential impact of this selection method on our

results. As shown in Figure 5, more than 97% of Pb-rich particles contain nitrate, and Pb-N particles show significant correlation with these particles ($r = 0.84–0.96$, $p < 0.01$). That is to say, potential underestimation may uniformly affect all samples, thereby maintaining the validity of our comparative analysis. Our findings indicate that while the method might slightly underestimate the absolute content of lead nitrate, the relative trends and comparative conclusions drawn in our study remain robust.

To make it clear, "Although this method has limitations in fully capturing the variability or total content of lead nitrate, it does not impact the accuracy of the analysis results of lead nitrate in this study. There are significant correlations exist between Pb-N particles and Pb-rich particles mixed with nitrate ($r = 0.84 - 0.96$, $p < 0.01$), ensuring that the observed trends and relationships remain robust and reliable." has been added to the revised manuscript. Please refer to Lines 126–130 of the revised manuscript.

5. Several sentences in the manuscript require revision for clarity. The authors should pay close attention to sentence structure and tense consistency, as seen in the comments on lines 25, 54, and 280-281. Ensuring clear and precise language will improve the overall readability of the paper.

We sincerely thank the reviewer again for pointing out the issues with clarity and consistency in the manuscript. We have carefully revised the sentences on lines 25, 54, and 280–281, as well as throughout the text, to improve sentence structure, tense consistency, and overall clarity. These changes ensure that the language is more precise and the manuscript is more readable.

Lines 229-230: This sentence is unclear. Please rewrite it for better clarity.

Thanks for pointing out our mistake. The sentence has been revised as "After coal-to-gas conversion, the mixing state of lead in spring and winter are more complex than in summer and fall, with the number fractions of lead mixed with

sulfate and chloride salts doubling compared to summer and fall.". Please refer to Lines 232–234 of the revised manuscript.

Lines 232-233: This sentence should be described more specifically so that readers can clearly understand how the mixture of lead and nitrate differs from other components.

We agree with the comment. The sentence has been revised as " More than 97% of Pb-rich particles are mixed with nitrate, while only 22–64% of Pb-rich particles are mixed with sulfate, chlorine, or oxygen.". Please refer to Lines 237–238 of the revised manuscript.

Line 25: "but how well it improves..." — Correct the tense.

Thanks for pointing out our mistake. The sentence has been revised as "Coal-to-gas (CTG) policies are important energy transformation strategies to address air pollution issues, but how well they improve atmospheric lead (Pb) pollution remains poorly understood.". Please refer to Lines 24–25 of the revised manuscript.

Line 54: "Pb(NO3)2 generates from the chemical transformation of PbO and PbCl2 in the atmospheric process." — Correct the grammar.

Thanks for pointing out our mistake. The sentence has been revised as " $Pb(NO_3)_2$ is generated from the chemical transformation of PbO and $PbCl_2$ in the atmospheric process". Please refer to Lines 54–55 of the revised manuscript.

Line 218: "to doubled" — Correct the grammar.

Thanks for pointing out our mistake. The sentence has been revised as "This is a significant decrease compared to double the average and maximum hourly number fractions of Pb-rich particles during the past coal-fired centralized heating period in Beijing.". Please refer to Lines 201–203 of the revised manuscript.

Lines 280-281: “The study included heating and non-heating periods, and the results emphasized the importance of heterogeneous hydrolysis during the heating period before 'coal-to-gas' on the formation of Pb(NO3)2.”— Please check and correct.

Thanks for pointing out our mistake. The sentence has been revised as "The study included heating and non-heating periods, and the results emphasized the importance of heterogeneous hydrolysis during the heating period before “coal-to-gas” on the formation of $Pb(NO_3)_2$.". Please refer to Lines 290–291 of the revised manuscript.