

Atmos. Chem. Phys. Discuss., author comment AC2 https://doi.org/10.5194/acp-2022-9-AC2, 2022 © Author(s) 2022. This work is distributed under the Creative Commons Attribution 4.0 License.

Reply on RC2

Qian Liu et al.

Author comment on "Rapid reappearance of air pollution after cold air outbreaks in northern and eastern China" by Qian Liu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-9-AC2, 2022

SUMMARY:

This work focused on the rapid increasing of air pollution after cold air outbreaks. The changes of AQI after CAO events is divided into two types of events: rapid change and slow change. By comparing and analyzing one example from each of the two types of events, the authors indicated that the depth, duration and coldness of cold air masses, as well as the stability of the vertical structure, modulated the changes in air pollution after the CAO events, and also made a comparative analysis of the role of thermal and dynamic processes. In addition, the two types of events corresponded to different atmospheric circulation systems, which may become previous signals. However, a lot of work mentioned about the drop of air pollution during the CAO and the subsequent rebound. The influence of the depth and duration of the cold air mass on the rebound of air pollution is also a classic conclusion in textbooks. Therefore, I think this research is not innovative enough. In addition, when analyzing the effects of cold air mass, the authors only conducted statistical analysis based on a single case, without the verification of numerical experiments, so its credibility and persuasion are not enough. A mandatory major revision is recommended.

Response: We would like to express our sincere thanks for your review efforts. We accepted all of your comments and revised the manuscript accordingly. The high-relsolution figures can be found in the supplementary file (Reply_on_RC2.pdf).

We agree that many previous studies have noted the reappearance of air pollution after CAO, while few studies have made statistics on the reappeared air pollution. So far, the quantitative relationship between air pollution reappearance and CAO properties and relevant physical mechanisms are still unclear. In this study, we show that the reappearance of air pollution after CAO is a common phenomenon. An isentropic analysis method (Iwasaki et al., 2014) is employed to quantitatively investigate the CAO of what features and structures could lead to the rapid reappearance of air pollution. Some large-scale patterns of CAO in its initial stage are also recognized as precursors for rapid reappearance of air pollution. We describe the novelty of this study at Lines 60–65.

To gain the credibility of our results, a series of numerical experiments were added in the manuscript as you suggested (See reply to MAJOR COMMENT #5).

The detailed response and revision are given below.

MAJOR COMMENTS:

1. Line 71-72: Air pollution in winter is mainly haze, but the data used in this study is AQI. I hope to know the change of PM2.5 during and after CAO and whether it is consistent with the conclusion of AQI

Response: We agree that air pollution in winter is mainly haze, during which the primary air pollutant is $PM_{2.5}$. Following your suggestion, we check the connection between AQI and $PM_{2.5}$ concentration. Figure R1 shows the daily mean AQI and $PM_{2.5}$ concentration in winters of 2014/2015–2021/2022 (2292 days). The correlation coefficient between them is as high as 0.96. We also investigate the evolutions of $PM_{2.5}$ concentration during the two types of CAO events (Figure R2). The rapid and slow rebounds of $PM_{2.5}$ concentration still could be observed in two kinds of CAOs. Therefore, the main conclusions of AQI is thought to be consistent with the results of $PM_{2.5}$ concentration. We choose AQI as a comprehensive indicator in the manuscript as there are some cases that $PM_{2.5}$ is not the primary air pollutant. We add above result in the manuscript at Lines 89–92: "It should be noted that the daily mean values of AQI and another well-known air pollution index of $PM_{2.5}$ concentration (primary air pollutant of haze) has a high correlation coefficient of 0.96 in study period. The key results of this study are in a good agreement with results based on $PM_{2.5}$ concentration (figure not shown)."



Figure R1. Relationship between daily mean AQI and $PM_{2.5}$ concentration averaged in North China.



Figure R2. Evolutions of spatial averaged (northern and eastern China: $114^{\circ}-122^{\circ}E$, $30^{\circ}-40^{\circ}N$) cold airmass depth and PM_{2.5} concentration during CAOs. (a) and (b) are composited by the CAO_rapid events and CAO_slow events, respectively. Shading represents the 95% confidence interval of the composited mean value.

2. Lines 73-74: The data in this study is only used until 2018/2019. Why aren't the data for the last three years used? Especially in the winter of 2020/2021, many cold air events happened in North China, including record-breaking cold waves. The CAO events in the present study does not cover the recent years, lacking the timeliness of the facts.

Response: Following your suggestion, we expand the study period to

2014/2015–2021/2022. The number of CAO events increases to 52 including 33 rapid reappearance events and 19 slow reappearance events. The main results do not change after the study period is extended. We update Figures 2–6, 9, 12–13, Table 1 and relevant descriptions with new data (see revised manuscript).

3. Line 142-143: Only more than 50% of CAOs are found to show a worse AQI after reappearance. This ratio indicates that the change of AQI after CAO event is irregular, and the probability of its increase and decrease is basically the same. The phenomenon of air quality rapid rebounding after the CAO needs to be further confirmed.

Response: In this study, we find the reappearance of air pollution exists in the decaying period of all CAO events. To clarify the result of Figure 3, we would like to first explain the two indices (IA and RI) defined in this study.

The deterioration rate of air quality during the reappeared air pollution is define by the change of AQI from the period during CAO to the period after CAO,

$IA = AQI_a - AQI_d.$

To describe the index more clearly and intuitively, we add some tags in Figure R3. and

denote the maximum AQI in the periods after and before CAO, respectively denote the minimum AQI in the period during CAO.

According to above definition, **the positive value of IA indicates the reappearance of air pollution (e.g. Figure R3). Figure R4c shows that the** IA **(x-axis) in all CAO events have a positive value, which confirms the reappearance of air pollution is a common phenomenon.** We add this result in the revised manuscript at Lines 167–168: "Figure 3c shows that the IA (x-axis) in all of CAO events have a positive value, which means in AQI has experienced an increase after CAO. This result confirms the reappearance of air pollution is a common phenomenon."



Figure R3. Time series of the regional averaged cold airmass depth (blue line) and AQI (orange line) in northern and eastern China (114°–122°E, 30°–40°N). The gray dashed line denotes the threshold value of the cold airmass depth.

Another variable RI is used to compare the air quality before and after CAO,

$RI = AQI_a/AQI_b.$

The value of RI larger (smaller) than 1 indicates the reappeared air pollution after CAO has a worse (better) air quality than the air pollution before CAO. Figure R4b shows more than half of CAOs will followed by a reappeared air pollution with worse air quality.



Figure R4. (a) Statistical distribution of daily change of AQI calculated by all of days during the 8 winters (brown bars) and days during the period after the CAOs (green bars). (b) Statistical distribution of the reappearance index of air pollution in 52 CAO events. (c) The increase in the AQI (x-axis) and the change in the AQI from the period before the CAO to the period after CAO (vertical arrows) in 52 CAO events. Black dashed line denotes the standard deviation of AQI. The arrowhead and tail represent AQI_a and AQI_b, respectively. The orange (blue) arrows denote that the AQI worsens (better) after CAO.

4. Lines 146-150: This part of the description is not very clear and hard to understand. The CAOs are divided into two groups: slow reappearance and rapid reappearance. In fact, not all AQI increased after CAO events. However, using "reappearance" to describe the two groups is misleading. In addition, the title of the abscissa in Figure 3c is "Increase of AQI after CAO", but actually there have decrease of AQI after some CAO events, so it is not appropriate to use "increase". In my understanding, the difference between the value of the arrowhead and tail should equal to the value of the abscissa, but obviously there are many points that do not correspond well. Please check the correctness of Figure 3c and add to the explanation in this part.

Response: In Figure R4c the position of each arrow on X-axis is IA (AQI_a-AQI_d). As explained in the response of MAJOR COMMENT #3, we found that in all of the CAO events AQI has experienced an increase. Here, we divide 52 events into two groups according to the value of IA. A rapid (slow) reappearance of air pollution is supposed to have an IA larger (smaller) than 34.3, which is the standard deviation of the AQI daily variation during the past 8 winters.

As for Y-axis, the difference between the values of arrowhead and tile is not equal to the value of abscissa. The position of arrowhead and tail on y-axis denote the and , respectively. Thus, the arrow on y-axis has a similar meaning with RI, indicating whether the air quality after CAO will get better or worse than that before CAO. The orange up arrow (blue down arrow) represents the reappeared air pollution has a heavier (lighter) degree than the air pollution before CAO. Combine the vertical direction of arrows with their positions on X-axis, we found the CAO_rapid (CAO_slow) events usually lead to a worse (better) air quality than that before CAO.

Following your suggestion, we add some detailed descriptions in the manuscript at Lines 173–176: "In Figure 3c, the arrow on y-axis has a similar meaning with RI, indicating whether the air quality after CAO will get better or worse than that before CAO. The position of arrowhead and tail on y-axis denote the and , respectively. The orange up arrow (blue down arrow) represents the reappeared air pollution has a heavier (lighter) degree than the air pollution before CAO."

5. Lines 189-190: When analyzing the effect of the depth and NFC of cold air mass on the rapid events and slow events, only one case is used for statistical analysis respectively, and the verification of numerical experiments is lacking. I am not sure whether the conclusion drawn through the individual case apply to most of the other rapid and slow events, so I am skeptical about the applicability and credibility of the effect of the depth and NFC of cold air mass. In addition, numerical experiments should be added to confirm the conclusion in Section 4.

Response: Following your suggestion, numerical experiments are added to verify our main results (See revised manuscript at Lines 305–340). First, the CAO_rapid event and CAO_slow event selected in Section 4 are simulated using WRF-Chem version 4.3. The simulated results are consistent with the observations. Then, a series of sensitive simulations are also conducted in which the connection between cold airmass properties and diffusion conditions of air pollutant is verified. These numerical experiments confirm that the CAO properties could determine the pollutant diffusion conditions in atmospheric boundary layer and further influence the rebounding speed of air pollution. The detailed analyses are listed below.

The domain of simulations is designed with a horizontal grid spacing of 10 km covering most part of East Asia (Figure R5). The FNL data is used as the initial and lateral boundary conditions to drive the meteorological simulation. The MEIC anthropogenic emission inventories are used in the chemical simulation. The main physical and chemical parameterization schemes include the WSM6 microphysics, the MYJ PBL scheme, the RRTM for longwave and shortwave radiation, RADM2-MADE/SORGAM for gas-phase chemical and aerosol schemes.



Figure R5. WRF model domain configuration (black rectangle). The colored shading presents the temperature disturbance in NHC_C experiment at model level 9 above ground. The blue rectangle represents the northern and eastern China.

Figure R6 shows the spatial averaged cold airmass depth, boundary layer height, vertical stability and surface $PM_{2.5}$ concentration during the CAO_rapid and CAO_slow events. Here, the emissions in both two experiments are set as the values in December 2016 to investigate the impacts of meteorological conditions. In experiment of CAO_rapid event, the air pollutant increases rapidly on days 0 and +1 under the condition of the relatively low boundary layer height and strong vertical stability. Such conditions of atmospheric boundary layer are not conducive to the diffusion of air pollutant and tend to induce rapid reappearance of air pollution (Zhang et al., 2014; Liu et al., 2017). In CAO_slow experiment, however, the $PM_{2.5}$ concentration keeps in a low level due to the high boundary layer height and weak vertical stability. In addition, the temporal evolutions of these variables are highly consistent with the observations shown in Figures 7–8 and 11 in the manuscript, suggesting both the rapid and slow reappearances of air pollution can be well captured by numerical model.



Figure R6. Evolutions of spatially averaged (northern and eastern China: $114^{\circ}-122^{\circ}E$, $30^{\circ}-40^{\circ}N$) cold airmass depth, atmospheric boundary layer height, vertical stability (1000–850 hPa) and surface PM_{2.5} concentration during (a) CAO_rapid event from 14 to 17 Dec 2016 and (b) CAO_slow event from 11 to 14 Feb 2018.

To verify the connection between CAO properties and abovementioned boundary layer diffusion conditions as discussed in Sections 4.1 and 4.2, a control experiment (the CAO_rapid event in Figure 7) and additional sensitive simulations are also conducted. In sensitive experiments, temperature disturbances are artificially added in the initial field following Bai et al (2019). In NHC_C (NHC_W) experiment, the NHC of cold airmass is increased (decreased) by adding a cold (warm) bubble centered at a height of 0 km. The cold (warm) bubble had a latitudinal radius of 10 km, longitudinal radius of 5 km and a vertical radius of 2 km, with a minimum potential temperature perturbation of -8 (8) K. As shown in Figure R5, the temperature disturbance is minimized at the center and increased to 0 K following a cosine function over the horizontal and vertical radius. To increase (decrease) the cold airmass depth in DP_C (DP_W) experiment, the Cold (warm) bubble added in the initial field moves to the height of 2 km. Note that the NHC may also change with cold airmass depth in DP_C and DP_W experiments.

Table R1 shows the simulation results averaged in study area on day 0, when air pollutant has the rapid increase rate as shown by Figure R6a. In NHC_C and NHC_W experiments,

changes in NHC cannot cause an obvious variation in boundary layer height, but can lead to changes in vertical stability. In DP_C and DP_W experiments, despite the changes of NHC and vertical stability, we find that changes in cold airmass depth will result in an obvious change in boundary layer height. These sensitive experiments confirm the main results of Sections 4.1 and 4.2, that is, the properties of CAO could effectively impact the diffusion conditions in atmospheric boundary layer.

Table R1: Averaged cold airmass properties and atmospheric boundary layer conditions on day 0 from control and sensitive experiments. Upward (downward) arrows denote the value of sensitive experiment is greater (less) than the value in control experiment.

Experiment	Cold airmass depth (hPa)	Atmospheric boundary layer height (m)	Negative heat content (– K hPa)	Vertical stability)(10 ⁻³ K m ⁻¹)
Control	165.6	597.2	1898.4	5.47
NHC_C	163.9	579.9	1994.5 h	6.79 h
NHC_W	167.6	661.0	1745.9 i	4.15 i
DP_C	189.4 h	727.4 h	2447.9	5.98
DP_W	118.8 i	519.9 i	1238.4	4.74

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MINOR COMMENTS:

1. Lines 30-33: The use of tenses is confusing, alternating between the general present tense and the past tense. Suggest to check the whole text and unify tenses.

Response: Following your suggestion, we check the whole text and unify tenses.

2. Lines 28-30: What does the air pollution mainly refer to in the study? Is it haze pollution? Many articles about haze pollution are cited in the formation of air pollution in the following paragraph. Be more specific about the characteristics of air pollution.

Response: The air pollution in winter over North China is mainly haze pollution. Following your suggestion, we specify the type of air pollution in the introduction at Lines 29–30: "North China, experiences the most pollution, which mainly is haze pollution, resulting in attention from researchers."

3. Lines 40-41: impacting the variations in air pollution [on the synoptic time-scale].

Response: Revised as your suggestion. See Lines 42–43: "As emissions and topography do not vary much from day to day, meteorological conditions play an important role in impacting variations in air pollution on the synoptic time-scale."

4. Line 50: increased as much as 2.8 times than what?

Response: Following your suggestion, we revise the sentence at Lines 52–53: "During the reappearance of air pollution, the regional mean pollutant concentrations increased as much as 2.8 times than concentrations reduced by CAO."

5. Line 100: defined as the two days before onset day [to] the onset day

Response: Revised as your suggestion. See Lines 118–119: "The period before CAO is defined as the two days before onset day to the onset day (days -2 to 0)."

6. Lines 100-101: Does "the three following the period during the CAO" refer to day 1 to 4? It does not match the annotation in Figure 1.

Response: "The three days following the period during the CAO" refer to the three days after the end date of period during the CAO, when regional mean cold airmass depth falling below the threshold (166.8 hPa). It should be noted that the period after CAO varies with events, since the end date of period during the CAO is different in the selected events. Thus, in the CAO event plotted in Figure 1 (Figure R3), "the three days" is days +2 to +4. According to your comment, we add an explanation at Lines 119–122: "The period after CAO, which is also called the decay phase, is defined as the three days after cold airmass depth falling below the threshold (days +2 to +4 in CAO event plotted in Figure 1). The period after CAO varies with events, since the end date of period during the CAO is different in the selected events."

7. Lines 142-144: This view should be presented with caution. It is probably because the selection time of the period before CAO avoids the high value AOI, which may have been affected by cold air.

Response: Following your suggestion, we revised this sentence as: "Based on the definitions of periods before and after CAO in our study, more than 50% of the CAOs are found to show a worse AQI after reappearance. In some extreme events, the AQI after CAO could be twice as high as the AQI before CAO."

8. Figure 5 and 6: Add the meaning of the black box in the caption.

Response: Following your suggestion, we add the meaning of black box in the caption of Figure 5 at Line 574: "The black boxes denote the northern and eastern China"

9. Figure 10: what does the dots refer to?

Response: The dots in Figure 10 refer to the area with vertical gradient of the potential temperature $>6 \times 10^{-3}$ K m⁻¹. See caption of Figure 10 at Lines 587: "vertical gradient of the potential temperature (contour, unit: 10^{-3} K m⁻¹, dotted for $> 6 \times 10^{-3}$ K m⁻¹)"

We acknowledge your great help to improve the manuscript.

Thank you very much.

Please also note the supplement to this comment: <u>https://acp.copernicus.org/preprints/acp-2022-9/acp-2022-9-AC2-supplement.pdf</u>