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Interactive comment on “Sensitivities of NO_x transformation and the effects on surface ozone and nitrate” by H. Lei and J. X. L. Wang

Anonymous Referee #1

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This paper investigates the change of surface ozone and aerosol nitrate in response to the perturbation on chemical transformation of NO_x in the atmosphere using both model simulations and ground station measurements. The authors conducted a series of experiments to examine the impact of NO_x transformation due to increase and decrease of NO_x emissions and of atmospheric temperature. Although the presented material is interesting and valuable and the topic of the paper is suitable for Atmospheric Chemistry and Physics, I believe a major revision is needed before it can be accepted for publication as explaining following.

General Remarks:

The major conclusion of the paper is that “the decrease or small increase in ozone

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concentrations over industrial regions result in the responded nitrate increasing rate staying above the increasing rate of NO_x emissions” [Abstract lines 11-13 and page 21968 lines 1-4]. I do not think this conclusion can be inferred from Figures 4 and 5. Over major industrial regions such as Northeast U.S. and Los Angeles, the authors point out that there is less active NO_x transformation in ozone chemistry due to ozone decrease in Figure 5 [page 21967 line22], so that more active NO_x could be available for nitrate formation [page 21968 lines 1-3]. However, the increase of nitrate formation over these regions is less than the NO_x emission increase rate as shown in Figure 4. Over the other contiguous states, both ozone and nitrate formations are increased with NO_x emission increase. However, the figures give no information that indicates that the significant increase of nitrate formation resulted from the more active NO_x due to the ozone increase.

The paper needs further in-depth discussion. The authors show changes of surface ozone and nitrate under the various perturbations on NO_x emission and atmospheric temperature. However, explanations of the changes are generally vague, without clear and detailed support. Many explanations are presented as hypotheses [i.e. Page 21967 lines 16-18, Page 21968 lines 11-12, Page 21968 lines 23-25, Page 21970 line 2]. The explanation is also incomplete without including the discussion of reduced nitrogen (i.e. NH₃ and NH₄) for the change of nitrate. For example, another hypothesis in explaining Figure 4 is that the availability of free atmospheric NH₃ is the dominant factor for the pattern of nitrate change in response to NO_x emission change. The authors should discuss the relative contribution to nitrate formation from ozone and NH₃ chemistries.

Specific Comments: Page 21962 lines 11-16: I do not see any support for these conclusions in the paper. The paper shows only the change of surface ozone and nitrate in response to the change of NO_x emission. There is no clear and direct evidence in the paper to demonstrate that the change of ozone concentration is the driving reason for the rate of nitrate increase staying above the rate of NO_x emission increase.

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Page 21962 lines 16-18: Atmospheric temperature impacts not only NO_y chemistry but also the partitioning of nitric acid between gas phase and aerosol phase.

Page 21963 lines 6-9: No, other studies do account for the effects on the present condition, such as the studies of HTAP activity.

Page 21964 lines 3-4: “Partitioning” is not an appropriate word to use here. Surface ozone and nitrate do not share a common source entirely.

Page 21965 lines 17-21: Please describe briefly the mechanism of nitrate simulation in the CAM-Chem model.

Page 21966 lines 24-26: CASTNET does provide the measurement of dry depositions for aerosol nitrate and gas phase nitric acid separately. Please do model-observation comparison for aerosol nitrate directly since this paper investigates the change of aerosol nitrate, not oxidized nitrogen.

Page 21967 line 22: How do you know “less active NO_x is transformed”? NO_x titration reduces ozone concentration but do not change NO_x volume mixing ratio.

Page 21968 lines 1-3: Figure 5 tells nothing about how NO_x changes due to the change of ozone.

Page 21968 lines 3-4: I do not think this conclusion can be inferred from Figures 4 and 5 as I discuss in the major comments.

Page 21968 lines 6 and Figure 7: It would be more valuable to add the monthly change rate of surface NO_x concentration in the figure.

Page 21968 lines 8-10: Why does warmer climate cause higher nitrate formation? The relationship of nitrate formation and temperature change shown in Figure 8 does not support this conclusion. Meanwhile, I do not understand how “a response of the NO_x-to-ozone pathway as mentioned above” supports the summer increase of nitrate formation.

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Page 21968 lines 25-28: The months are “January, February, and March”.

Page 21969 lines 13-15: The change of surface nitrate in response to the temperature change shown in Figure 8 is partially attributed to the impact of temperature on the partitioning of total HNO₃ between gas phase (nitric acid) and aerosol phase (nitrate). Since this paper investigates the impact on nitrate formation via perturbation on ozone chemistry, it would be valuable if the authors could explore how much of nitrate change is solely due to perturbation of NO_y and ozone chemistry by temperature change.

Page 21969 lines 18-19: How is active NO_x concentration affected by the NO_x-to-ozone transformation?

Page 21970 line 11-16 and Figure 10: What do the symbols “P-C1” and “W1-P” represent? Please change the scale of “change ratio” in the left panel so that the readers can tell the diurnal cycle of ozone change ratio.

Page 21970 lines 16-17: Again cold temperatures also favor the total HNO₃ partitioning toward aerosol nitrate.

Page 21970 lines 17-22: How do you know that the NO_y chemistry, and not the HNO₃ partitioning, is the major reason for the daytime decrease of nitrate? Please check the other relevant tracers (i.e. NO_x, gas phase nitric acid, etc) or design sensitivity experiments to confirm your conclusion.

Page 21973 line 1: Please delete “further” since the authors do not provide any other reasons before.

Technique Corrections Page 21963 line 14: Is it a typo of “processes”? The word of “progresses” makes more sense. Page 21964 lines 5-6: How about “Then, the air concentrations of ozone and nitrate aerosol and emissions of their precursors NO_x and VOCs on several ...”. Page 21966 line 6: Please spell out CCSM3. Page 21966 line 28: Please change “right” to “bottom” and “left” to “top”. Please do a similar change in the Figure 3. Figure 6: Please add “and ozone” after “Surface aerosol”. Page 21970

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line 23: Please add “For the experiment of temperature increase” before “The ozone
...”.

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