

Manuscript title: Characteristics, sources and reactions of nitrous acid during winter in the core city of the Central Plains Economic Region in China via high-time-resolution online measurements

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<https://doi.org/10.5194/acp-2019-916>

The paper covers the monitoring results of HONO – classified into three different pollution levels (clean, polluted days, and severe polluted days). The net HONO production rates for both homogeneous and heterogeneous reactions were determined. The observed HONO concentrations during daytime and nighttime were explained by the calculated HONO production rates. The cumulative frequency ratio distribution of HONO emission/HONO was used to elucidate the possible HONO source.

The Introduction needs a brief description of your tasks (what tasks?) to achieve your goals (specify goals). All I read is that you will monitor HONO levels along with RH, especially during the night. Rationale for your study is weak (see the following general comments),

Methodology is adequate, but you need to cover uncertainty in all measured pollutants so you have a feel about the correct use of significant figures. Results/discussion section needs rewritten to follow the logical sequence – the format is similar to that of Zhang et al., 2019.

My overall recommendations are major revision (did not read your conclusions).

General comments:

1. In general, the text can be followed. However, there are many awkward, unclear, redundant, unnecessary, ambiguous, and confusing phrases/statements (see the following tech/English comments). A professional expert must edit the text for clarity and for better flow before resubmission.
2. The rationale for your study is weak – need more elaboration. The fact that no study has been performed in Zhengzhou (L 129) does not justify the novelty of your study. You should have covered the setbacks of previous studies and state those tasks (including tables/figures) currently evaluated have not been properly addressed in previous studies. Unfortunately, I have found none. Anyway, pls emphasize the uniqueness of your study.
3. I have a hard time figuring out that the results from one single sampling site located on the rooftop of a building in Zhengzhou university (L 134) can represent the air quality in general and HONO in particular in the entire

Zhengzhou city (180 million population, L 106). This is why there are so many monitoring stations (traffic, urban and background) within a given large city to reflect the air quality within the city. Pls modify your title as well those in the text.

4. Need to clearly cover HONO sources and sinks as well as homogeneous ($\text{NO} + \bullet\text{OH}$) and heterogenous ($\text{NO}_2 + \text{H}_2\text{O}$) in the text (introduction and discussion). For example, the role of ground surface at night for HONO deposit (sink) and the reemission (source) from HONO reservoirs (e.g., soil nitrite), etc. The other sink source, albeit insignificant, is that HONO may react with others to form new compounds, as in the case of reactions with amines in forming nitrosamines. How about transport of HONO? – what is the lifetime in atmosphere (hours under in-door conditions).
5. Need to discuss the uncertainty in your results due to variations of many parameters (e.g., rate constants and OH radical values, L 250-253). This leads to the following comments about the use of significant figures.
6. Be careful about use of significant figures. Delete decimal points for RH (L 182, 185, 187, 194, Table 2, etc.), for NO_2 level (L 197-198; also, you compare with standard of 80), for $\text{PM}_{2.5}$ (Table 2), for level in $\mu\text{g m}^{-3}$ (L 198, etc.), for ratio (L 34, 304, etc.).

Use 2 significant figures for rate constants (L 17, 19, 24, 33, 258-260, 266-268, 391-394, p. 16, etc.), considering the uncertainty of all parameters used and average value of OH selected.

Use one digit after the decimal points for these values (L 271-273). Use 13.4 (L 271)

7. Be consistent with the format of unit. You use m s^{-1} (or $\mu\text{g m}^{-3}$), yet m/s and $\mu\text{g}\cdot\text{m}^{-3}$ are in Table 2 (delete centered dot). Use ppbv throughout the text, but ppbV in Tables 1 and 2 and Fig. 8 (some ppbv and one uses ppbV).
 - Fig. 2: Unit for CO is wrong – should be ppmv.
 - L 428-431: The unit for OH concentration is wrong.
 - L 441: The unit for P_{unknown} is wrong
8. Data need show the variation; use Box plots or error bars in figures and add standard deviation in tables.
9. Need proper citations for equations and rate constants, e.g., in L 247, 250

10. The comparison with others (Table 1) may not be useful and must be made with care since the studied year is different (some in 2012 – may not have adequate end-of-the pipe treatment), nature of sampling sites is different (some in urban, suburban and even rural sites) and atmospheric dynamics in these regions are far different.
11. Why no discussion of OH production rate as a function of O₃ levels?? In other words, is any HONO information related to O₃ pollution level?? It is relevant since •OH radical generated from HONO is in turn used for O₃ production.

Specific comments:

1. Title is misleading.
2. L 15: All of a sudden, the phrase “three sources” pops up. Need to clearly state what they are.
3. Why not place centered dot symbol for OH radical? or •OH
4. Use a proper term in lieu of rate which refers to time
5. L 45: Why only these two? What about others including HONO itself and acetone?
6. L 49: what reaction? Should be more than one reaction.
7. Citation:
 - a. Provide adequate spacing between citations, or Hou et al., 2016; Michoud et.... (need a spacing before the 2nd citation)
 - b. Make sure all cited references are listed and vice versa. For example, citations in L 55 (L 60) and Table 1 (Elshorbany et al., 2012) are not listed.
 - c. Delete redundant citations. No need to cite twice in the beginning and at the end of the sentence. Pls change throughout the text (L 75, 82, 88, 93, 156, 157, 246, etc.) by delete the second citation.
 - d. L 95; et al. (2013); delete the extra comma
 - e. Avoid excessive self-citations (one is enough, e.g., L 115, 116, etc.)
 - f. For the same last name, use the format of Jiang et al., 2018c, 2018e; Liu.... (L 115). Pls change throughout the text.
 - g. L 140: why cite this? Need year for this reference.
 - h. L 181: why cite this one? Delete
 - i. L 313: Acker et al. (2005) reported...
8. L 60: what is 1:1??
9. L 79: There is no connection between these two sentences. Need a sentence (such as revised R3 reaction) leading to the following sentence.
10. L 123: These two measurements (PM_{2.5} and HONO) cannot clarify the sources, sinks and reactions. Pls reword.

11. L 173: what is uncertainty of AIM? How about MDL for other gases??
12. L 190: Table 1 must come before Table 2. Rearrange the table number.
13. Interesting. You cover all these parameters (L 199-204) shown in Fig. 2, yet Fig. 2 is mentioned several sentences later (L205). The same illogical sequence is found in L 403 which mentions P_{unknown} and P_{emi} , but the equation for these is shown much later (L 436). Also need to cover these rates for estimating daytime HONO budget.
14. L 240: why??
15. L 252: why in reference to Beijing?
16. L 253: Can you calculate OH radical concentration from those discussed later in L 418?
17. P. 11: You are talking about night and mentioned no pollution source near the site. Why all these calculations related to traffic?? Unclear.
18. Pls explain the contradictory statements: important pathway (L 321) and unimportant pathway (L 20) for heterogeneous reaction for HONO formation.
19. Use the stack format for equations in a separate line (L 389); use the solidus format for those within the line (L 402).
20. L 431: Why the same values for both PD and SPD? That means you treat PD and SPD the same.
21. Fig. 1: How could one tell the wind direction?? There is no shade area of black and red color (caption says so). Change in Fig. 2 too.
22. Fig. 6d and RH effect: Was the phenomenon also observed by others?? Who is to say that 77% is the inflection point? Just say "reach a certain high level, HONO...."
23. References:
 - Be consistent!
Use lowercases for journals (e.g., L 502, 515, etc.)
 - Need periods after all journal abbreviations (e.g., L 502, 515, etc.)
 - Adequate subscript/superscript (e.g., L 535)
 - Use correct journal, or Phy. Chem. Chem Phy., (L 536)
 - Use lowercases for articles (e.g., L 505)
 - L 546 ref is not cited.

Tech/English comments:

1. L 13, 178, 179: Use polluted (not pollution)
2. L 33: Use was (not should be)
3. Be consistent about the use of verb tense; some use past tense in the first part of the sentence with present tense used in the latter part.
4. L 32, 279: Delete the redundant abatement (first one)
5. L 44: delete rate
6. L 54: UV/Vis
7. L 56: result comparison
8. L 67: have discovered

9. L 68-69: How does ratio “account for”? Use “is” 0.1-0.8%
10. Reword awkward L 81, word “radiation” (L 93), confusing (L 108-110).
11. L 99: Use pathway of...: mechanism is the same, but pathway is different.
12. L 107: “Food Production and Modern Agriculture” specified by (not published)
13. L 112: delete “is” and insert “is” ahead of selected
14. L 114: Not Zhengzhou chemical characteristics. Should have written as:
chemical characteristics of PM in Zhengzhou.
15. L 121: PM_{2.5} is not chemical
16. L 124: How systematic?
17. L 137: western Fourth-Ring Expressway
18. L 151: Use chemicals (in lieu of substances)
19. L 153: O₂ and N₂ (not O and N)
20. L 160-161: Clearly state which instrument is for which compound, e.g., 48i for CO measurement.
21. L 167-168: The manual uses the term “should be”. But in your statement, you should use “was changed””was calibrated”.
24. L 171: A space after “≥” sign
25. L 181: specify that it is daily average
26. Delete the first unit (L 182, 185, 188, 206, 301, 317, 360, etc.), first two units (L 301, 449, etc.)
27. L 189: .. north with high WS ... Also, how high is high? > 3 m/s, or > 4 m/s? Be specific!
28. L 190: effect of pollutant removal
29. L 202: mean values of what? of all pollutant concentrations?
30. L 209: Is your sampling site in urban area? You mentioned on the university campus. If so, the comparison is not valid.
31. L 214: No logic about sunrise – previous max HONO values (8-10 am) are way past sunrise. Should have written after 10 am.....
32. L 215: reword the incomplete phrase.
33. L 219: delete again
34. L 220: and the concentration remained the same (Is it true??) until sunrise.
35. L 222: you meant NO₂ diffusion?
36. L 228: How is atmospheric migration? Should be “migration of atmospheric airmass”.
37. L 248: should be Eq. (1); Eq. (2) is in L 296.
38. L 249, 337, 338, 349, 389, etc.: A space b/a the = sign
39. L 254: Adequate subscript in k_{OH+bar}
40. L 274: reword
41. L 279: Use rate (not level)
42. L 280: delete the extra spacing b/a the “/” sign
43. L 307: on the campus
44. L 324: medium of what??
45. L 349-350: wording? cannot see “indicating ...of what most cases”? Why?

46. L 361: sedimentation of what? Do you mean deposit?
47. L 367: Delete "study of the"
48. L 374: decreases after...
49. L 383: in the (not then)
50. L 396: How is rate improvement? Use increase
51. L 402: The expression of represents
52. L 411: delete an
53. Table 1: A space before (%), or HONO/NOX (%); before year, or Jun. 2012,
Also add SD
54. Fig. 3: Need errors bars. Use the present tense, "represent". Show one
example in Box plots so one has an idea about the magnitude of variation.
55. Fig. 4: Need error bars
56. Fig. 6: A space after HONO