

The manuscript presents a study on volatile organic compounds (VOCs) at three sites in a Chinese megacity, ~Shanghai. The concentration, composition, sources and of VOCs have been extensively studied, especially in the major cities like Shanghai. The authors claim multiple-site comparison as the main selling point. However, it is not enough to provide new insights that the authors expect. Namely, I have reservations about the novelty of this study. Moreover, I have serious concerns on the rationality of the methods and quality of the results presentation. The lots of grammatical errors also make the manuscript very difficult to be reviewed. Overall, the manuscript is well below the average of papers published in Atmospheric Chemistry and Physics, so I do not think it is worth publishing in its current state. However, I do not mind providing some specific comments for the authors' reference and reviewing a resubmitted edition after it is substantially improved.

Title:

1. The term “secondary formation potentials” is not a common expression. It is not clear what you are referring to.

Abstract:

2. Line 29: VOCs-SO₃ has not been defined before. So is SOA in line 31.

3. Line 31: “new insights”. I do not think the paper at its current state provides new insights into the accurate air quality management.

Introduction:

4. Major comment: The whole section is rather simple and most of the contents (if they are right) can be found in text book. I do not think it is necessary to elaborate them in a research article. What's worse, I am confused by the introduction of some basic knowledge. For example, line 40: RO₂ is formed following oxidation of VOCs, but VOCs are oxidized by OH, O₃ and NO₃ (NOT RO₂). Also, the oxidation does not necessarily lead to formation of secondary VOCs, although some species, e.g., formaldehyde, can be formed through photochemical reactions. Then, not all secondary VOCs can be transformed to SOA. Lines 91-96: This is not an accurate summary of the roles of VOCs in SOA formation. Lines 98-100: I do not get the point why there are strong industrial, vehicular and power plant emissions in mountainous area. Moreover, motor vehicles and power plants are significant sources of NO_x. Then, how to explain the NO_x-sensitive regime for O₃ formation?

5. Major comment: It is not clear what the authors mean by pollution characteristics, which is too general. It is also not clear what the knowledge gap is. The authors must make it clear what the manuscript adds to the current understanding of VOCs in Shanghai.

6. Inaccurate expressions and grammatical errors are everywhere throughout the manuscript. I cannot list all of them, just give some examples here: line 93: “...that declines the vapor pressure reduction”, line 94: pPM, gas-particular partition; line 95: a significantly decreased in the vapor pressure; line 98: transition ~ regime, line 100: strong emissions of industrial, vehicular, power

and biogenic, line 101: NO_x transition regime (what is it? I never saw this kind of expression), same for the “VOCs transition area” in line 104; line 106: varied photochemical reactions; line 108: “VOCs are likely to response to the pollution of $\text{PM}_{2.5}$ and O_3 ” –I am not sure if I understand correctly because of language problem; if my understanding is correct, what is the point of studying the responses of VOCs to $\text{PM}_{2.5}$ and O_3 , rather than the other way around?; line 112: pollution VOC characteristics.

7. Lines 43-45: What’s the point of emphasizing the 57 PAMS VOCs? There are a wide range of VOCs that can be the precursors of O_3 and SOA.

8. Lines 121-122: Is there any evidence proving that VOC pollution in Shanghai is more serious than ever before? It is contradictory to the statement in lines 71-71 “the VOC concentrations of China have decreased in the recent years along with the effective control strategies”.

9. Lines 122-130: It reads like pollution characteristics just means concentration, which is not true.

10. Lines 130-131: I cannot agree. In fact, sources and contributions of VOCs to O_3 and SOA have been well documented.

11. Lines 131-132: Which studies are you referring to when you say “ten years ago” – a specific time frame? At least, the studies you are referring to should be discussed.

Methodology:

Many problems in this section need to be justified or clarified.

12. Major comment: Different instruments, as well as analytical methods, were used for the analysis of VOCs at the three sites. How did the authors reconcile the data so that they can be compared? What is TD300 (line 177) that is not defined? In general, the small molecule and large VOCs are detected by FID and MSD, separately. Lines 182-184: What are the accuracies and detection limits for the minority of the species, i.e., those beyond the “95% and most VOC components”, and what is the range of the precision? Line 188: What is the SEAS site? It has never been defined before.

13. Description about this method in section 2.3 is confusing. How can the spatial heterogeneity be determined for a single site, as stated in line 190? Line 193: Does j represent site or dataset? Contradictory descriptions.

14. Major comment: There are significant deviations in the understandings of PMF. Lines 202-203: the descriptions of g_{ik} , f_{kj} and e_j are totally wrong. Lines 204-207: The function Q is introduced. However, it is not clear what the purpose of introducing it is and how the authors used it? Lines 210-212: “EF is the error faction and can be set to 0.05-0.2” – What was the EF the authors set in this study? How did the authors determine the solution with seven factors as the optimal one?

15. Lines 223-225: How did you determine the number of polluted and all trajectories in a grid, and how was the weight function W_{ij} applied?

16. Major comment: The equation (9) calculates the responses of VOC to O₃, which is opposite to the statement in lines 245-246 that “The characteristic structure and reactivity could influence the contribution of VOCs to O₃ formation”. Lines 249-252: The rationality of using 100 as a threshold of background O₃ should be justified. Why 100? Note that it is a quite high value, especially in cool seasons. It is also totally wrong to assign the VOC concentrations during the O₃-background time period as background VOC concentrations. In most cases, the patterns of O₃ and VOCs are inconsistent. For example, O₃ got lowest values at night and in early morning when VOCs are at high levels. Lines 251-252: What is the logic behind? Why is VOCs influenced by the variation of O₃, and not the other way around? Lines 254-255: The logarithmic conversion is also problematic. Equation 12 should be written as $\ln y = \ln a + \ln x$.

15. Inaccurate expressions and grammatical errors in this section include but are not limited to the followings. Lines 180-181: “The samples were condensed low-carbon (C₂-C₆) compounds and high-carbon (C₆-C₁₂) compounds ...”; Line 185: “trace instruments”; Lines 211-212: “option solution”, “greatest solution”; Line 214: “observe the back trajectories, source and direction of pollutants”; Line 218: “This study was determined the 24-h back trajectory”.

Results and discussion:

16. Line 266: “60 VOC species” is contradictory to the statement that “Totally 43 species of VOCs were observed” in line 183.

17. Lines 271-273: What’s the point of comparing the wind speed that is very spatially uneven?

18. Lines 274-288: The comparisons are rather simple. Are there same number of species, same species, same sampling season and etc.? Without discussion on these factors, the comparisons are meaningless.

19. Lines 295-312: I do not see the necessity of discussing such simple facts with too many words.

20. Lines 314-315: I am surprised to see such high levels of O₃ in the sampling period. Without any doubt, the authors made mistakes in calculation or unit conversion.

21. Lines 315-317: Readers would have no idea what the point of this discussion is. Are the dates special?

22. Lines 317-318: I do not think this was the reason for the correlation. Otherwise, did you see correlation between VOCs and O₃, where the former was also precursors of the later?

23. Lines 320-324: Why not refer to sources of PM_{2.5} and VOCs in Shanghai. Transportation as the main source of PM_{2.5} and VOCs in different cities does not necessarily mean the homology PM_{2.5} and VOCs in Shanghai.

24. Lines 325-331: First, I do not think the correlation is worth discussing. In most cases, the diurnal patterns of VOCs and O₃ are opposite. Second, the opposite patterns are mainly due to inconsistent patterns of VOC emission (e.g., emissions in morning and evening rush hours) and

O₃ formation (e.g., daytime). The discussions are far-fetched and I do not understand “, and counteraction was imposed by uncertain factors during the formation of O₃.”

Due to the obvious deficiencies and too many comments if I go on, I have to stop the review here. Please note that to save time I have not reviewed the next sections. I suppose there are also lots of problems therein. However, I would be willing to review a substantially improved manuscript.