Herein the authors present data collected during the BraVo campaign wherein they collected data within the HTHH plume over Brazil. The authors present data collected from balloon-borne instruments, satellite instruments, model, and ground instruments in an attempt to better understand the composition of the particles in the plume. This paper represents a massive amount of work that went into collecting, analyzing, and interpreting the data; I commend the authors for this effort.

I have to be frank, this paper was very frustrating to read. I apologize if my frustration came through in the review, but I cannot believe that any of the 24 co-authors put a serious effort into reading this paper or evaluating the methodology and conclusions prior to its submission. It is very frustrating to, as a reviewer, do the work that should have been done by co-authors. That said, I endeavored to keep the criticism constructive, if blunt at times.

In my view, this paper has a very long road to travel before it is ready for peer review. The overall structure of the paper is very disjointed and confused, it lacks focus, and does not provide sufficient evidence to accept the authors' conclusions. There are many sections that repeat information in previous sections, paragraphs containing instrument/measurement details appear in the main body of the text when they should have been in section 2, there is a lack of detail that precludes me from understanding what methodology the authors employed (much less repeat the experiment myself), there are contradictory statements and conclusions drawn about the data, etc.

Perhaps the most disconcerting aspect of this paper is the number of times the authors admit that their data do not match what is expected, based on previous work, but follow that admission a "just-so" explanation. I apologize for using that phrase, but the authors continually defend their conclusions, despite what the data actually support, by invoking phrases such as "it is possible", "could be due to", "potentially", "might be", etc., all the while failing the provide definitive evidence for their conclusions. For example, to justify the disagreement between their measured ion ratios and those published and accepted in the literature we receive statements such as (all emphases added):

1. line 340: "This suggests *potential* exposure to wildfire smoke...", but their back trajectories do not go over the burn areas.
2. line 343: "it's also *possible* that some air parcels *could have* originated from the Eastern Pacific". Possible, yes, but their back trajectories do not support this.
3. line 471: "...it's *possible* that additional sources contributed to higher concentrations of other ions..." What other sources? Where is the support?
4. lines 482-487: "Ca2+ could *potentially* be associated with gypsum...this ratio *could* arise from two primary mechanisms...the observed ratio alone cannot definitively distinguish between them."
5. lines 590-591: "The low sulfate concentrations observed in our IC analysis *could* be due to interactions of SO4 with Ca2+...*potentially* leading to sulfate loss"
6. lines 610-613: "This, in turn, suggests a shorter SO2 lifetime and, consequently, the *potential* for larger sulfate formation through enhanced coagulation. This process *likely* contributed to the lower sulfate concentrations in our analysis..."

7. lines 650-651: "Our balloon-borne sampling, utilizing filters, *may have* preferentially retained larger particles while *potentially* losing smaller..."

8. line 669: "The lack of sulfate in our samples is *likely* real but also highlights a *potential* deficiency in our method..."

9. lines 698-699: "The rapid nucleation of gypsum, driven by high calcium concentrations, *likely* led to the effective removal..." I note that the authors did not demonstrate the gypsum was formed, it is only a convenient explanation for their low sulfate ratio.

10. line 710: "...*potentially* enhancing sulfate particle formation..."

This is only a representative, not exhaustive, sample from the text, but it demonstrates the point that their observations do not match literature values for sea salt and they have no proof of their alternative explanations.

I do not necessarily disagree with the author's claim that sea water (salt water) persisted in the stratosphere for months. What else could we expect from an injection of 146 Tg of sea water? But I have to ask which is more plausible: The authors' correctly identified every *potential* reason for their data being significantly different from the accepted standards (despite the lack of scientific evidence to support their claims), or their sampling was flawed and their samples were contaminated? I find the latter more likely and I believe all of the evidence, as presented within this paper, points directly to that conclusion. Interestingly, the authors acknowledge this possibility (lines 623/635) as well, but do not embrace this possibility.

Everything in this paper points to either contaminated sampling or some very interesting atmospheric chemistry and the only support we receive from the authors are passing statement of it *could* be something else. If the former, then the conclusions that can be drawn from this work are very narrow and the paper requires a massive rescope and revision. If the latter, then the authors must provide definitive proof and the paper, again, requires a massive rescope and revision. Again, all of the IC data presented in this paper point to contamination. Therefore, I see no opportunity for either of these options playing out. It is for these reasons that I must recommend rejection without further review.

Below are specific comments regarding the text.

1. The abstract reads like an introduction. Please rewrite as an abstract.

2. line 45: I do not know what the sentence that begins with "Unlike major..." means. Please clarify.

3. line 55: is SO4 missing its charge?

4. Section 2: there is no discussion of balloon measurements.

5. line 99: "SAOD" was defined earlier.

6. line 100: Kelvin does not use the degree symbol.

7. line 100: Sentence beginning with "Given that the...", I do not understand what the authors are trying to communicate. Please revise.

8. line103: "...contrary to previous claims." This is an unsubstantiated claim and is made en passant. The authors fail to discuss the differences in calculation and the impact this may have on the results. If the authors wish to make this claim they should do so in the body of the text alongside a thorough discussion of why their method is correct.

9. Figure 1: this figure makes no sense here. If you wish to use it, please move it to an appropriate location.

10. line 114: "400 km west", west of what?

11. Figure 2: please include panel labels. Please make range of y-axes the same for each panel so the reader can better appreciate your visualization. Please make the font size for the ground-based lidar profiles larger (x/y axes). Please make the map's scale larger so it can be read. As is, the map is not useful; please zoom in to better show location in Brazil.

12. line 124: no need to redefine CALIPSO.

13. line 126: This reads better if you create a new paragraph between sentences here.

14. lines 130-135: Text better fits in section 2.

15. "sampler was coupled with a radio-controlled valve attached to the balloon's neck". Why? What did this accomplish? Please clarify why this is important.

16. Section 3.1: All subsections belong under section 2.

17. line 140: Should "OPC" be "POPC" here? If not, please explain the difference.

18. Section 3.1.1: The authors describe 2 versions of the POPC. 1 with 8 channels, the other with 30 channels. Please include the size bins for each (you use this information later in the paper) and please clarify which version you use in the following analyses/figures.

19. line 150: what is meant by "phase-sensitive detection"?

20. line 154: What is meant by scattering ratio? Is this the color ratio?

21. Figure 3: Within the legend, what is "NPOPC"?

22. Figure 3 caption: What is meant by "backscatter"? Is this the returned signal intensity; why is it greater than 1?

23. line 164: Should this be a new section or subsection? Fits better in section 2.

24. line 171: reference to "Fig. 7" is way to early (you just finished discussing Fig.3!) and is not needed anyway. Please revise for clarity or just remove.

25. line 174: Reads better if there is a paragraph break here.

26. line 180: Where in France?

27. Figure 4: Extending the plots to 3.5 hours after the landing does not make sense and seems like a waste of page space. If this is needed, please tell the reader, otherwise eliminate the unneeded portion.

28. line 189: Should this be a new section? Fits better under section 2.
29. line 189: Neither ECC nor CFH are defined in the text. Please define.
30. line 189 ff: Please include manufacturer and model for this equipment.
31. line 192: Please provide a reference for the accuracy claim. There are a plethora of papers on ECC and CFH instrument/measurements. Can the authors cite an appropriate number of them to help curious readers better understand these measurements?
32. line 195: Is it dew point or frost point? They are different; please clarify.
33. line 197: Please provide a reference for the 4% claim.
34. line 199: Can the authors provide a guide for what is meant by "middle stratosphere and tropical tropopause"? What altitude does this specifically mean for your measurement location?
35. Table 2: This table is never reference in the text. Do you need it?
36. Table 2 caption: Why the period in "Table.2"? This occurs in all table captions. Please revise as needed.
37. line 204: What "previous balloon-borne aerosol collection efforts" are you talking about? Whose? When? Where? Please provide context and references.
38. line 207: Is there a reference for the India work?
39. lines 204 - 210: This reads very similar to the earlier "Sampler" section. Please consider condensing.
40. lines 217-218: This information is also redundant with that presented in a previous section. I see no value in restating this (nearly verbatim); please revise or remove.
41. Figure 5: In section 3.1.1 you used radius, here you use diameter. Please be consistent.
42. line 239: It is unclear where the 12.8 ng number came from. Doesn't this assume a total volume of 6 mL? Please explain.
43. line 239: The authors failed to explain what is meant by filter positions #5 and #8. Please provide enough detail to allow the reader to understand (especially those that have not done IC).
44. Section 3.3: Overall, this entire section requires a thorough revision to communicate clearly. As written, I could not reproduce the method and I am unable to understand it well enough to appreciate your work.
45. lines 249-256: Much of this text is out of place and fits better in the introduction. Please revise for clarity and brevity.
46. Figure 6: label the color bar and include units

47. Figure 6 caption: "Colored circles depicting peak volcanic SO2 emissions" Really? Why does this only go to 1? Your table 1 has Pinatubo at 21 Tg. Is this scale relative to Pinatubo? Also, from the text I would have guessed this is the SO2 / SAOD ratio. Please ensure the colorbar label, the caption, and the text tell the same story.

48. Figure 6 caption: ...versus the corresponding semi-annual mean (sSAOD)..." The y-axis and title of Figure 6 show SAOD, not sSAOD. Please ensure consistency.

49. Figure 6 caption and line 274: sSAOD has different definitions in the caption and in the text. Please correct.

50. line 268: GloSSAC already defined.

51. line 278: Why compare only with the largest eruptions? Perhaps it is better to say "When compared with past eruptions of comparable VEI magnitude..."? Event then, you should note that VEI is not an indicator of ejected SO2 mass. VEI is a measure of the volume of ejected tephra, so I am unsure how the sentence in question is relevant to the paper.

52. line 298: This sentence is out of place and should be removed. Perhaps more appropriate in the introduction.

53. line 336: "only NO_2^- was present in reportable quantities at Pos #4. Due to associated error bars, other ions measured at this position were not statistically significant..." I cannot see NO2- on the bar plot of Figure 7. If NO2- is "statistically significant" and NO3 and SO4 are not, then I certainly have no idea how to interpret this figure or your results. Please clarify and correct.

54. line 341: What is meant by DeepBlue AOD? This is put in the text with no introduction. Can you provide something for the reader so he won't have to search (e.g., a brief statement of what this is, a reference, etc.).

55. line 343: "also possible..." This is pure speculation that is not supported in any way by anything you have shown. This is grasping at straws to support a predetermined conclusion. Either support this claim, or remove it from the paper.

56. line 355: In Section 3.1.1 you stated POPC has either 8 or 30 bins; here you say 10. Please clarify and/or correct.

57. line 363: Should be Figure 11.

58. lines 363-368: Here, you discuss extinction before explaining how you calculated extinction. You say the method is explained in the next section, but the next section references a non-existent section! Please ensure your argument is presented in a logical and coherent manner.

59. Figure 9: The text in this figure (axis labels, tick labels, legend text) is very small and the x-axis tick labels are crammed together so much that it is hard to read. Please consider improving the readability of this figure.

60. Section 6: Much of the content in this section should be moved to section 3.1.1.

61. line 384: "...and therefore the size of the POPC within the HTHH plume is underestimated" I think you mean the POPC underestimates particle size in the HTHH plume?

62. Figure 10: What are the bins for this instrument?
63. Figure 10: This is scattering efficiency at 90 degrees, correct?
64. Figure 10 caption: What is meant by "near 785 nm"? This is all theoretical work, so didn't you define wavelength? Can this be more precise?
65. lines 392-401: There is a lot of redundancy with previous sections. Please condense.
66. lines 396-397: COBALD and SAGE already defined.
67. line 399: "...(see Methods)..." No such section exists and the promised explanation is nowhere to be found in the text. Please correct. Must include detailed description of how you calculate extinction for POPS and COBALD.
68. last paragraph on page 19: I must disagree with your conclusions here. This does not prove sea salt was present. What it shows, is that by changing the refractive index you can get your extinction profile to be within $\pm 50\%$ of the SAGE value. You then state that the quality of the agreement is dependent on both wavelength and composition (this is contradictory to your overall conclusion). Please revise for clarity and provide additional support for your conclusions or consider removing this section entirely.
69. Figure 11: At this point in the text you have not discussed what is meant by "correction". Please consider revising the text for clarity.
70. lines 426-431: It is unclear why this is at this specific location within the paper or why it is in the paper at all. It does not seem to contribute; please revise or remove.
71. line 451: Here you state the expected Cl/Na ratio is 0.8. Earlier you stated it is 1.8 (line 337/338). Please correct.
72. line 462: Which equations are you referring to? Please provide enough information for the reader to understand what these equations do and provide the reference (Zhang et al. 2021 is not in the bibliography).
73. lines 458-481: It is difficult for me to follow your rationale in presenting this information and I have great difficulty at this point in seeing how your overall research fits together. Please revise for clarity.
74. line 472: "ss-X" is introduced and never used. Should this appear in the table?
75. line 477: "The Ca/Cl ratio was measured to be 0.8...(see Table 5)" Table 5 shows it to be 0.88, which is it?
76. Table 5: I am at a total loss for how to interpret/compare these tables. The units aren't even the same. Even assuming a density of 1 g/mL, the numbers between the top tables and the bottom are vastly different. As presented, this provides far more support for rejecting the hypothesis that sea water was present than it does for accepting the hypothesis. If there is a different interpretation the authors need to provide it.

77. line 541: "...significant deviations between our BraVo aerosol samples, collected at stratospheric altitude...and established oceanic ratios reported by *multiple studies*" (emphasis added). If the authors insist on concluding that sea water was present, then they must provide a robust argument to explain this discrepancy. The burden is on them to provide more than possibilities.

78. line 541: Did the authors collect "blank" samples on the flights? If not, then you cannot discount the possibility of contamination in flight, after landing, in transit to the lab, or during analysis.

79. line 551: "The presence of sea salt in the stratospheric plume is further supported by our analysis of POPC measurements."

    (a) First, the authors claim this is "further support", which indicates they already provided support for stratospheric sea salt, which they did not. On line 541 they stated that their measurements had "significant deviations" from established observations. This explained away by an untested and unproven hypothesis that these deviations might be caused by "fractionation" as if the assertion stands on its own.

    (b) Second, the POPC measurements do not provide support for sea salt. I can only assume they refer to the calculated extinctions; this is after they admit their agreement depends on composition and wavelength (the fact that there is a wavelength dependence indicates that your refractive indices are wrong). It is exceedingly simple to adjust the POPC-based extinction calculation to match SAGE to within $\pm 50\%$. I am not insinuating misconduct on the authors, but we know nothing of how they calculated extinction and what the sensitivities of their assumptions are. In short, this is far from convincing support.

80. lines 552-554: "Therefore, the combined evidence from our BraVo samples and *the comparison with established oceanic ratios provides compelling support* for the hypothesis that the [HTHH] eruption injected significant amounts of sea salts directly into the stratosphere." (emphasis added). What? The authors previously stated there were "significant deviations" (line541) and "...our samples consistently diverge from typical seawater composition" (line 543), now they claim "compelling support"? I am at a loss for how to reconcile this. Please clarify.

81. Page 26: Neither GEOS-Chem/MERRA-2 nor ACE-FTS appeared in Section 2. Please include them there.

82. lines 590-592: The authors present a hypothesis for why sulfate is too low in their sample (supposedly it reacted with calcium). However, if you are removing sulfate *and* Ca through this mechanism, wouldn't this result in the Ca/Na ratio being lower than expected? Your Table 6 shows this ratio to be too high, which contradicts your hypothesis.

83. line 592: The authors speculate that gypsum was produced as a loss mechanism for sulfate. Is there any support for this claim other than "sulfate was too low"?

84. Table 7: Do we really need 8-9 decimal places? Please revise to correctly communicate your level of precision.

85. Page 29: The authors introduce another field mission in which nearly every condition surrounding the eruption is different. This is very difficult to follow and I fail to see how this is germane to the original thesis. Is this presented solely to demonstrate the gypsum is formed as part of volcanic eruptions? If so, the authors fail to deliver on this point by stating "These findings imply that gypsum was present in the ground samples." They *imply*, but do not prove.

86. line 634: Here, the authors nail it! This is the far more likely issue at play. Their sampling was flawed, which biased their entire ion analysis. This should be the end of the story.

87. lines 654-655: It is unclear what the authors mean here. "...the aerosol population could have undergone significant changes leading to the observed differences in our in-situ samples." You're comparing your measurements to those made by ACE-FTS (3 days separated). Are you suggesting that it changed more in 3 days than it did in the preceding 8 months?

88. Conclusions: I did not read the conclusions of this paper. There is too much inconsistency and speculation, while ignoring real issues related to their sampling, to give any merit to any conclusions that could be drawn from this.