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Interactive comment on "One-year observations of carbonaceous and nitrogenous components and major ions in the aerosols from subtropical Okinawa Island, an outflow region of Asian dusts" *by* B. Kunwar and K. Kawamura

Anonymous Referee #1

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The paper of Kunwar and Kawamura presents and discusses a one year observation period of complete characterisation of total suspended particulate matter in the outflow region of Asian dust. Scientific significance of the paper is rather weak, but given appropriately used methods and thoroughly discussed results can be considered for publication in Atmospheric Chemistry and Physics after addressing quite numerous comments.

The first major problem with the paper is the undefined size of the particulate matter

(TSP). It is important to size segregate for air quality studies while the fine fraction has important climatic implications. Both implications are poorly addressed since the TSP mass is often dominated by the large particles having short residence time in the atmosphere while at the same it is impossible to tell whether obtained results apply to fine particles.

The second major problem is a one-year measurement campaign claiming seasonal differences and patters. It is rather obvious to everyone that any given year cannot be claimed as typical thereby reducing the significance of the obtained seasonal pattern.

A more general problem is an excessive focus on correlations without considering their significance and more importantly lacking cross-check balance with absolute concentrations considering whether they make sense with the claimed pattern. Existence or absence of a correlation is not a proof of causality, therefore, needs better context every time discussing observed correlations.

Comments in sequence of their appearance in the paper:

Abstract

The abstract should clearly specify 12 months period of observations and weekly concentrations as this information is at a core of the paper. The use of expression “growing season” should be substituted with more scientific “active biota season”.

Introduction

The second paragraph seems out context to me as it presents anything known in aerosol science, but without contextual relevance to the study.

P22062, line 6. Are the claimed 70% relevant on regional or global scale? Be more specific.

Line 20. How significant is the claim of the first time ever measurements at Cape Hedo without considering other studies in the region of similar scope: seasonal patters, chemical species etc.

Line 24. Seasonal trends based on one year only can be misleading – be careful not to over-interpret.

Methods

I am confused about TSP mass measurements which were not introduced, but the pie charts suggest they were made. OC concentrations were probably significantly overestimated without using quartz-behind-quartz double filter approach. The artefact should at least be acknowledged and discussed accordingly.

P22063, line 15. Specify procedure of the field blanks as the variety exists and may be interesting to a specialised reader.

Line 21. How carbonate carbon is considered negligible in the dust outflow? How can Ca be soluble in the samples if not CaCO3 dissolved in acidic particles? Please reconsider/reword.

P22065, line 14. Claiming "some degree of uncertainty" is not justified, more likely the results would be systematically biased high with higher than the actual minimum ratio. In would be more appropriate to use more conservative and rounded ratios of 3, 4, 5 and 3 for winter, spring, summer and autumn respectively.

Section 2.4 Good approach, but things become muddled in the text on whether non-sea-salt fraction or sea-salt fraction of the component is presented and discussed. I recommend authors split each relevant component into "nss" and "ss" fractions and redo correlation analysis with all implication re-discussed. That is especially applicable to correlation Tables. Some of the correlations are dubious to me and can be due to unspecified, not split fractions of e.g. SO4, Ca, etc.

Section 2.6 Air mass trajectories have an uncertainty of 15-30% of the distance at the start/end of it (check HYSPLIT model details on NOAA site) making their length above 120hours unrealistic. For instance, 120hour trajectory length is of the order of

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2000-3000km which introduces 300-1000km error at a backward starting point. Thus a longer trajectory of e.g 168 hours (7days) puts starting point essentially in random position. The sampling duration of 7 days does not require plotting 7day backward trajectory.

Results

P22067, line 12. I already noted a problem of aerosol mass. How was it measured? Was it a sum of analysed components? In any event it has to be clearly stated.

Line 22. My guess is that the most likely reason for Ca and TSP correlation is the dissolution of $CaCO_3$ as the particles become acidic. If the authors have other ideas please state, but provide explanation in any event.

P22068, line 17. The use of air parcels is inadequate given sample duration of 7days. Change to air masses.

P22069, line 1. I am curious why POC was highest in spring and not winter as it was primary anthropogenic. Were the emissions lower in winter? Probably not, but then the lack of precipitation in spring may have contribute to the observed maximum.

Line 10-20. When was the maximum WSON of 2.2ug/m3 observed? Summer?

P22070, line 2. Correlation coefficients are considered inconsistently in many parts of the paper. Correlation coefficient is either statistically significant or not statistically significant. If not significant then should not be even mentioned as the “null hypothesis” or spurious correlation cannot be reliably dismissed. ONLY if correlation was statistically significant can it be considered high or low. My estimate by the number of samples suggests that correlation of e.g. 0.22 is not statistically significant.

Line 3. The presence of EC must be accompanied by certain amount of OC; therefore, weak correlation does not exactly mean that sources are different, but rather the OC having at least two independent sources which distort correlation.

P22072, line 4. When claiming significance of marine carbon (it is relevant claim) the authors should consider marine carbon concentrations from literature and relate them to the observed percentages. Do things match and make sense? They may well do, but be more detailed and less speculative.

P22073, line 7-12. Consider and discuss numerous isotope studies for the presence of marine carbon in polluted air masses. Again consider absolute numbers as well, do they make sense?

Line 22. If you believe Ca coming from dissolved $CaCO_3$ in dust when picking up acidic species during transport, state it.

Section 3.4.1. NSS and SS fractions should be considered separately, otherwise discussion does not make sense. For instance, in continental air masses most of the Ca is nss while in marine air masses during summer most of it may be sea salt Ca.

P22075, line 13. Why Figure 10 presents 1:1 line when the ration in sea water is 1:1.16? 1:1 line doesn't make sense as sea water is not made of NaCl alone.

Line 19. Indicate points by circle or arrow when the chlorine depletion was highest.

Line 24. How applicable is to consider 6% contribution of biogenic $nssSO_4$ in this study when Savoie et al. (1994) study was made more than two decades ago and SO_2 concentrations decreased dramatically over those two decades at least in Europe and North America? It is likely that the percentage of biogenic SO_4 increased since the DMS source is the same while anthropogenic source decreased. Without considering SO_2 decadal trend such claim is meaningless.

P22078, line 4. Again which fractions are considered, nss or ss?

Table 4-7. Redo the tables with separate nss and ss components. At the moment lack of correlation between e.g. SO_4 and NH_4 does not make sense as Ca amount is very small to neutralise significant fraction of nss sulphate and NH_4 would more readily neutralise SO_4 than NO_3 due to acid strength.

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Figure 5. I do not understand how the pie charts were constructed when aerosol mass measurements were not introduced.

Figure 10. Change 1:1 with 1:1.16. Indicate the points with highest Cl depletion clearly.

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