Atmos. Chem. Phys. Discuss., 10, C4891–C4908, 2010
www.atmos-chem-phys-discuss.net/10/C4891/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: field observations" *by* P. Sundarambal et al.

P. Sundarambal et al.

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Received and published: 7 July 2010

We thank the reviewer for the constructive comments.

General Comments

Overall, the paper was of interest and reasonably well structured. However, I think there are many editorial improvements that could be made to improve its flow and clarity and to maximize the value to the scientific community.

As I read the manuscript, I had recurring thoughts of "Why is this included?" It wasn't until the very end that I realized that this paper was part of a pair of papers that also

included modeling. This knowledge explained why some information and statements that seemed out of context were in the paper (e.g., nutrient concentrations in sea water, sea surface roughness). That linkage ought to be noted early in the paper but, even so, some of the material remains ancillary to the main focus of this paper and could be dropped.

The authors need to be consistent in their use of terms. For example, the primary data segregation is between hazy and non-hazy days or periods, not "haze and non haze periods" as occasionally stated. The number of significant figures must also be used consistently. The mean cannot be to one decimal place and the associated standard deviation to three decimal places. The authors need to review their measurements and methodologies to ensure the proper number of significant figures is reported for each nutrient and deposition type throughout the report.

The lack of context and small size of the figures made several difficult to read and interpret.

Authors' response:

The materials which are ancillary to the main focus and the seawater concentration in Table 1 are removed from the manuscript as suggested by the reviewer. The importance of the seawater concentration is however explained in the companion paper. Also, the link between the present paper and the companion paper (Sundarambal, P., Tkalich, P., and Balasubramanian, R.: Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: Eutrophication modeling, Atmospheric Chemistry Physics Discussion, 10, 7779–7818, 2010.) is now highlighted in the introduction so that the readers of these articles can put the relevant information in the proper context.

We have revised the manuscript considerably to improve its flow and clarity.

We have now used an appropriate number of significant figures to express the concen-

tration of nutrients to be consistent with the precision of analytical measurements. The figures are redrawn to improve their clarity and presented in an appropriate context with larger size.

Specific Comments

Comment 1: In line 5 of p. 7747, iron is specified as a potentially important nutrient. However, line14 on p. 7748 only lists N and P species as the objective of the field study. The authors should document/justify why they did not analyze for iron.

Authors' response:

The present research work represents the first study of its kind focused on bringing together field-based investigations and eutrophication modeling to address atmospheric deposition of major nutrients and their impact on water quality and provide a scientific basis for more in-depth future study in this region. Using established methods, we have quantified different forms of water soluble nutrients (N and P species only) from dry atmospheric deposition (aerosol particulates) and wet atmospheric deposition (rainwater) to the tropic environment for the first time. Quantification of iron and its impact is not in within the scope of the current study, and will be studied and reported in future.

Comment 2: In lines 19 & 20 on p. 7748, is this level of precision necessary in describing the location? If so, insert "between" before the first latitude and longitude. If the latitude bounds are given, it is not necessary to specify "137km north of the equator"?

Authors' response:

Since Singapore is a small country with limited water environment, the authors thought that it is important to provide its location precisely. The sentence is now rephrased as "Singapore is a small island with total land area of 710 km^2 located at latitudes between 1˚06'N and 1˚24'N & longitudes between 103˚24'E and 104˚24'E (Fig. 2)."

Comment 3: In line 5 on p. 7749, I am not sure what was meant by the range of "maximum" wind speeds. Do the authors intend "mean daily maximum" over various seasons of the year or does the range represent the absolute maximums from the SW and NE wind directions?

Authors' response:

"Southwest (SW) and Northeast (NE) winds occur in the coastal area periodically and the maximum wind speeds range from 5m/s to 10m/s."

The above range represents the mean daily maximum wind speeds from the SW and NE wind directions. This correction is made in the revised manuscript.

Comment 4: In lines 8 and 9 on p. 7749, "N" and "E" are sufficient for describing the latitude/ longitude location.

Authors' response:

The sentence is rephrased as "This island is geographically located at 1˚ 13' 10" N and 103˚ 50' 54" E."

Comment 5: The first paragraph under Section 2.2 seems disorganized (sentences out of sequence – for example, the sentence starting with "TSP" might "fit" better at the beginning of the paragraph and the sentence starting with "The mass" might "fit" better at the end of the paragraph).

Authors' response:

The sentences are reorganized to make them coherent as per the reviewer's suggestion.

Comment 6: In line 23 on p. 7749, I assume the filters were conditioned prior to weighing rather than prior to and after sampling as stated?

Authors' response:

We agree with reviewer. The sentence is corrected as “The filters were conditioned prior to weighing”.

Comment 7: In line 11 on p. 7750, 4 deg C is warm for sample storage; how long were samples typically stored before chemical analysis?

Authors' response:

Although -20˚ C storage is optimum for long term preservation, we observed that 4˚ C is sufficient for short term storage. However, our samples are analyzed as soon as possible, within 1 or 2 days. The QA/QC protocols followed in this study are discussed in our previous publications (Karthikeyan and Balasubramanian, 2006; Karthikeyan et al., 2009a & 2009b).

Comment 8: In line 14 on p. 7750, how near was the meteorological station (NUS) to the SJI deposition site? No scale was shown in Figure 2. Presumably the precipitation rates at the two sites are similar.

Authors' response:

The distance between NUS and SJI is less than 10 km. The map in Figure 2 is replaced by a new map with an appropriate scale in the revised manuscript. The precipitation rates at the two sites are similar during storm events.

Comment 9: In line 18 on p. 7751, what temperature was the ultrasonic bath and to what “ambient” temperature was the extract cooled?

Authors' response:

The temperature of the ultrasonic bath was 60˚ C and “ambient” temperature was 27˚ C. The sentence is modified with inclusion of the exact temperature.

Comment 10: In line 18 on p. 7752, Wesley and Hicks (2000) could be listed as a reference on dry deposition (M.L. Wesley and B. Hicks, A review of the current status of knowledge on dry deposition.

Authors' response:

We thank the reviewer for this suggestion. The reference “Wesely, M., and Hicks, B.: A review of the current status of knowledge on dry deposition, Atmos. Environ., 34, 2261–2282, 2000.” is now included in line 18 on p. 7752 as well as in the reference in the revised manuscript as per reviewer's suggestion.

Comment 11: In line 5 on p. 7754, what is the basis for coarse PM having an upper diameter limit of 18 microns? My understanding is that hi-volume TSP samplers have an upper sampling size diameter of 25-30 microns.

Authors' response:

We agree that an effective cut point of 30 μm aerodynamic diameter is frequently assigned to the standard high volume sampler (Web site URL: http://www.epa.gov/ttnchie1/ap42/ch13/final/c13s02.pdf). The mistake is corrected in the revised manuscript.

Comment 12: In line 13 on p. 7754, “m” is not a rate. Insert “annual” before “precipitation” and change “m” to “m/year”.

Authors' response:

The sentence is changed to annual precipitation rate (P in m/year).

Comment 13: In lines 24 & 25 on p. 7755, the PSI and API indices are introduced. Why not present material and discuss solely from the original concentration measurements and just note in line 22 of p. 7756 that the air quality was moderate or unhealthy during October 2006 (if only used PSI data, confirm that based on TSP measurement)?

Authors' response:

As PSI and API are provided by Singapore and Malaysia, respectively on a daily basis as indicators of ambient air quality, we used those data for showing changes in air quality on a regional scale due to biomass burning. The TSP values measured in

Singapore followed a trend which was similar to those in the PSI and API data.

Comment 14: In lines 24 & 25 on p. 7756, rainfall is referred to but the figure reference is 3b rather than 3c. Because Figure 3c is missing, it is difficult to know whether the adjective of the rainfall is "intermediate" as written (vague term) or "intermittent". Figure 4 is small and difficult to read but does not appear to indicate "fire activity and intensity".

Authors' response:

We apologize for the mistake. The missing Figure 3c is given in the revised manuscript. Intermediate rainfall is replaced by rainfall distribution. We improved the quality of all figures including Fig. 4.

Comment 15: In lines 28 & 29 on p. 7756, the number of samples for the hazy (4) and non-hazy (16) days analysis are presented. The number of samples would be useful information to provide in the Abstract or Introduction to help guide the reader's decision to read and, if so, the interpretation and confidence in the results, especially for hazy days as is based on a limited number of samples.

Authors' response:

We agree with the reviewer that it would be preferable to have a larger number of atmospheric samples, especially during the hazy period. At the same time, we would like to clarify that we collected both particulate and rainwater samples using established methods as frequently as we could to study the impact of biomass burning on surface water quality through atmospheric deposition. The present study clearly indicates that atmospheric deposition is an important source of nutrients and becomes more important during biomass burning periods because of increased fluxes of nutrients across the air-water interface. We will provide the number of samples in the Introduction, as suggested by the reviewer.

Comment 16: In line 23 on p. 7757, the text refers to "larger fire/hotspot clusters" in Figure 4 but the current scale of the figure makes it difficult to see.

Authors' response:

Figures 4(a-d) are replaced by clear and larger figures.

Comment 17: In line 27 on p. 7757, also specify the "low altitude" of transport. Also, at the current scale of the figure, I cannot see that one trajectory came from the Indian Ocean (the two trajectories look similar to me. It seemed to me that the discussion did not follow the sequence of the plots in Figure 4 (i.e., plots not referenced in the same sequence that they were presented).

Authors' response:

Figure 4 a-d is replaced by clear figures with the corresponding explanation in the same sequence as presented in the revised manuscript.

Comment 18: Beginning on line 10 on p. 7758, the last sentence seems speculative and poorly worded. Can the authors provide some supporting evidence of why they believe this statement to be true and rephrase the sentence more precisely?

Authors' response:

Koe et al. (2001) is referenced to support the last sentence. The last sentence is rephrased more precisely as follows: The biomass burning-impacted air masses contained elevated levels of airborne particulate matter compared to those originated from other sources of air pollution.

Comment 19: Beginning on line 17 on p. 7758, the sentence about concentrations in seawater is out of context. The seawater concentrations in Table 1 are out of context and do not appear to be needed for this paper.

Authors' response:

The seawater concentration presented in Table 1 is removed in the revised manuscript and it is explained in the companion paper "Sundarambal, P., Tkalich, P., and Balasubramanian, R. Impact of biomass burning on surface water quality in Southeast

Asia through atmospheric deposition: Eutrophication modeling, Atmospheric Chemistry Physics Discussion, 10, 7779–7818, 2010.".

Comment 20: Beginning in line 23 on p. 7758, the nutrient concentration ratios from hazy to non-hazy days requires clarification and more discussion. What is the significance or implication of the rankings? The text appears to be referring to atmospheric concentrations but the cited Fig. 7 shows DAD fluxes. Ratio rankings are again shown at the end of the dry deposition discussion "with respect to their concentrations and fluxes." The order of the two specie ratio rankings were different and the rankings implied the difference between some specie ratios were significant. However, in Fig. 7 the TN and the NO_2+NO_3 DAD ratios are not significantly different. A careful review and rewrite of this section is needed.

Authors' response:

The purpose of the rankings of nutrients resulting from atmospheric deposition during hazy & non-hazy days is to understand the contribution of each species to changes in the water quality and thus to coastal water eutrophication**.** Figure 7 is shifted to Section 2.2.2 and the explanation associated with Fig. 7 is also moved accordingly.

The ranking of N and P species in terms of their corresponding ratios between hazy and non-hazy days is as follows: NH_4^+ >NO_3^-+NO_2^- >TN>ON and PO_4^- >OP>TP, respectively. This trend is consistent with the data presented in Figure 7.

We would like to clarify that the ranking shown at the end of the dry deposition discussion is based on DAD concentrations and fluxes, but not based on the ratio of DAD fluxes between haze and non-haze periods.

Comment 21: In line 24 on p.7758, Figure 7 is referenced before Figures 5 or 6 are referenced.

Authors' response:

The sequence of the figures is correctly presented in the revised manuscript.

Comment 22: In lines 8 & 10 on p. 7759 and in line 2 on p. 7761, the notations for reactive N and reactive P are not obvious to the lay reader.

Authors' response:

Notations for reactive N and P are replaced by nitrogen and phosphorous, respectively for clarity.

Comment 23: In lines 9 through 17 on p. 7759, some of the results have inconsistent significant figures. Also, showing the results in a table might be easier for the reader to interpret and remember.

Authors' response:

The significant figures in the text are corrected, and now have two decimal points. A table with DAD fluxes is now included in the revised manuscript in place of those results presented in the text.

Comment 24: In line 27 on p. 7760, the similarity of nutrient specie concentrations in rainwater and seawater, especially with a limited number of wet deposition samples, could be coincidence. How spatially and temporally variable are concentrations in coastal areas? More evidence and discussion is needed to support the insinuation of the runoff of pollutants after storms affecting concentrations in seawater. Perhaps this is an appropriate place to introduce the modeling effort.

Authors' response:

We also observed such a similarity of nutrient species concentrations in rainwater (24 numbers of samples) and seawater in our earlier study (Sundarambal et al., 2009). The research focus of this paper is only to quantify the atmospheric deposition of nutrients (N and P species) while the companion paper deals with the estimation of the relative contribution of atmospheric nutrient deposition to coastal water eutrophication. The details of seawater concentration and its changes due to runoff pollution are not discussed in this paper. The eutrophication model (NEUTRO) was used for the simu-

lation of spatial and temporal nutrient dynamics, and to predict the water quality trends in Singapore coastal waters. There was a significant concentration variation in coastal areas of the Singapore Strait and the Johor Strait in both space and time. The seawater concentrations (which are used as model baseline values) in Figure 6 were obtained by the statistical analysis of the data measured in the Singapore seawaters (as part of the routine water quality monitoring program by the Tropical Marine Science Institute (TMSI)). The modeling effort of the companion paper is introduced at the end of this section 3.2.2.

Comment 25: In Table 1, suggest changing "aerosol during hazy and non-hazy days" to "aerosol samples on selected days with hazy (October 2006) and non-hazy (November 2006 –January 2007) conditions". Suggest changing "Haze" to "Hazy" in the "Period" column.

Authors' response:

As per reviewer's suggestion, the sentences are amended accordingly in the revised manuscript.

Comment 26: When the table is first referenced, there is no clue as to why seawater concentrations are included. The min ON concentration on a hazy day was 5.15 in the text. Although a reference for the seawater concentrations is provided, it would be helpful to know the number of samples, location of sampling, depth of sampling, and seasonality of samples. In other words, how representative are the seawater numbers and what level of confidence can be placed in them?

Authors' response:

The seawater concentration in Table 1 is not presented in the revised manuscript. The rationale for inclusion of seawater concentration is given in the Section 3.2.2 of the revised manuscript. The seawater samples were collected every month at 3 different depths (surface, mid depth and bottom) at field monitoring stations in the Singapore

Strait as part of the routine water quality monitoring program by TMSI and the measured data were statistically analyzed to get model baseline values of seawater concentration.

A long-term field monitoring program (both time and space) is being carried out in Singapore waters by TMSI, NUS, Singapore to establish trends and also to provide data for continuous upgrading of the numerical model. These integrated measures, together with cooperation of government agencies, will allow for the continued successful management of Singapore's coastal environment.

Comment 27: In Fig. 2, it would be helpful to have a larger spatial context provided. Is it possible to show (identify) Sumatra and Kalimantan, which are referenced on p. 7755, as well as the sites mentioned in Fig. 3?

Authors' response:

Figure 2 is replaced by an updated figure with identification of all the sites including Sumatra and Kalimantan clearly.

Comment 28: In Fig. 3, Fig (c) is missing. In Fig (a), why not show all data as concentrations, rather than including less informative PSI and API numbers? In Fig (b), how were 3-h PSI values developed (e.g., BAM or TEOM or other continuous PM monitoring method)?

Authors' response:

The missing Fig3.c is now included in the revised manuscript.

We measured the TSP only at the Singapore monitoring station where the present study was conducted and those data are shown in Fig 3 (a). We did not do the measurement of the TSP at all other sites mentioned in Fig 3 (a). Instead, we obtained the PSI data from Singapore NEA (National Environment Agency, Ministry of the Environment and Water Resources, Singapore, available at: http://app2.nea.gov.sg/images/Regional PSI for 2006.pdf, 2010) and the API data from

Malaysia (DOE: Department Of Environment, Ministry of Natural Resources and Environment, Malaysia, http://www.doe.gov.my/, access: October 2006) during 2006 haze period. These indices are representative of the levels of particulate air pollution at the respective sites during biomass burning periods.

In Fig (b), the 3-h PSI values, as obtained from NEA during the October 2006 haze period, are shown; 3-h PSI data are only available during hazy days. The 3-hr PSI readings were calculated based on PM_{10} concentrations only. More details about the PSI calculation can be found at http://app2.nea.gov.sg/psi_faq.aspx.

Comment 29: Data from how many sites formed the basis of the PSI number? It might be interesting to show a plot of matched 24-hr TSP concentrations from these sites versus the SJI site.

Authors' response:

As explained before, the PSI data were obtained from Singapore NEA. Air quality measurements made by NEA at the north, south, east, west and central regions of Singapore formed the basis for the PSI index calculation (reference for more details: NEA's website http://app2.nea.gov.sg/psi_faq.aspx and http://app2.nea.gov.sg/12hnowcast.aspx and http://app2.nea.gov.sg/3hnowcast.aspx).

Since the present study is focused on the quantification of nutrients from atmospheric deposition, we collected 24-hour TSP samples using the hi-vol sampler at the coastal location only and do not have data from all other sites. However, as smoke haze is a regional air pollution problem, the levels of PM are quite within Singapore and the nearby parts of Malaysia.

Comment 30: In Fig. 4 (a-c), the geographical context of the trajectory maps is impossible to know without labeling or another map. In Fig. 4 (d), the caption should include a reference as to the source of the graphic or data from which it was generated. Even with enlargement, the "hotspot" locations are difficult to see.

Authors' response:

The geographical context of the trajectory maps is now labeled. The source of the graphic is also given. A map with the location of hotspots is included in the revised manuscript. In the new figures, the location of hotspots, the geographical context of trajectory maps and the source of the graphic are indicated clearly.

Comment 31: In Fig. 6 (a), an indication must be provided to help the reader understand the spatial extent, representativeness, and uncertainty included in the seawater measurements.

Authors' response:

Please refer the answers to the comments 24 and 26.

Editorial suggestions for "Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: field observations" by P. Sundarambal et al. (manuscript ACP-2010-153)

Editorial Comments

This manuscript should be carefully reviewed for grammatical correctness. Editorial changes to improve grammar and clarity that I suggest follow.

In line 3 on p. 7749, "millions" should be "million".

In line 9 on p. 7749, insert "air pollution" before "sources".

In line 11 on p. 7749, insert "the" before "southern".

In line 12 on p 7749, insert "on" before "regional".

In line 3 on p. 7750, "the lowest readability" could be phrased "a sensitivity".

In line 16 on p. 7751, "part" could be replaced by "portion".

In line 15 on p. 7753, a hyphen could be substituted for the space in "quasi laminar".

In line 24 on p. 7754, insert “the” before “wet”.

In line 25 on p. 7754, delete the first “rate”.

In line 7 on p. 7755, “high” is an ambiguous word choice. Perhaps you mean “substantial”. Also, suggest changing “at this time period” to “then”.

In line 8 on p. 7755, suggest adding “would occur, as evidenced” after “atmosphere”; delete “followed” and “would be observed”.

In line 16 on p. 7755, suggest changing the beginning of the sentence “The maximum TSP concentration observed in Singapore during the 2006 smoke haze event ($\sim$140 ug/m^3) was greater than that during the 1997-1998 haze events ($\sim$110 ug/m^3; Balasubramanian et al., 1999)”.

The second half of this paragraph is unnecessarily detailed and is not clear or easy to read.

Authors’ response:

The editorial comments are amended in the revised manuscript as suggested by the reviewer.

Editorial Comments

In line 11 on p. 7757, “5-days” should be “5-day”.

In line 13 on p. 7757, “continues” should be “continuous”.

In line 17 on p. 7757, “5-days” should be “5-day”.

In line 18 on p. 7757, “in” should be “at”.

In line 26 on p. 7757, drop “land origin”.

In line 27 on p. 7757, change “higher” to “high”. Also, suggest changing “others” to “another air mass travelled”. Insert “the” before “Indian” and delete “origin”.

In line 29 on p. 7757, insert “at SJI” after “concentration”.

In line 9 on p. 7758, change “mass” to “masses”.

In lines 20 & 22 on p. 7758, delete “in the order of”; a range is a definite number, not an estimate.

In line 24 on p. 7758, change “day” to “days”. Also on line 24, I suggest changing “shows” to “indicates” as ratios by themselves cannot show that smoke was a significant source of nutrients.

In line 25 on p. 7758, change “water in” to “waters of”.

In line 26 on p. 7758, I suggest changing “order of occurrences” to “ranking”.

In lines 7 & 8 on p. 7762, I suggest ending sentence as “non-hazy periods was quantified for a coastal water location in SEA.”

In line 11 on p. 7762, change “haze” to “hazy” in both locations and “increasing” to “enhanced”.

In line 12 on p. 7762, change “may” to “could”.

In line 13 on p. 7762, change the beginning of the sentence to “The ranking of the N and P specie concentration ratios comparing hazy to non-hay days was . . . ”.

In line 17 on p. 7762, change “shows” to “indicates”.

In line 20 on p. 7762, change “into” to “to”.

In lines 26 & 27 on p. 7762, I do not recall seeing where “Singapore’s coastal water quality degradation during haze episodes” was documented.

Authors’ response:

The above-mentioned Editorial Comments are amended in the revised manuscript.

As for the last comment, the sentence is corrected as “they might be an important

contributor to coastal water eutrophication during haze episodes."

Editorial Comments

In line 12 on p. 7763, change "Established monitoring sites" to "Establishing monitoring sites".

What type of monitoring sites? It seems that both coastal and interior sites would be needed to address atmospheric deposition to the watershed and that the water chemistry of both coastal streams and seawater would be needed.

Authors' response:

The suggested amendment is made in the revised manuscript.

The sentence in line 12-14 on p. 7763 is modified as "There is a need to establish a network of research stations for simultaneous air and water quality monitoring in Singapore and surrounding regions in order to address the impacts of atmospheric deposition of nutrients onto coastal water in SEA."

Editorial Comments

In line 17 on p. 7763, change "act as highly significant sources" to "acts as a highly significant source".

In line 19 on p. 7763, change "water" to "waters,".

In line 27 on p. 7763, insert "a" before "eutrophication".

In line 13 on p. 7766, change "?" to "fl".

Authors' response:

The suggested Editorial Comments are amended in the revised manuscript.

The authors gratefully acknowledge the Reviewer's efforts on carefully reviewing the manuscript and providing constructive & helpful comments to improve its quality. We

revised our manuscript according to Reviewer's comments and suggestions.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 7745, 2010.