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Comment

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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We thank the reviewer for the constructive comments.

General/recurring comments:

Comment 1: Phosphorus is generally not a limiting nutrient in coastal/marine waters. If phosphorus is limiting in the marine systems discussed here, please provide a reference to that effect.

Authors' response: On page 7747 in the introduction section (line 24-26), we have stated that “Atmospheric fluxes of nutrients could contribute a substantial fraction of

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dissolved N or P species to the euphotic zone, and enhance primary production, when nutrients are limited, leading to eutrophication.” This suggests that coastal/marine systems are either N or P limited. We include the following sentences in the revised manuscript: “Nitrogen is usually considered as the limiting nutrient in the oceans, although phosphate or even trace metals may also play a role in regulating phytoplankton growth. Gin et al. (2006) reported that Singapore waters were generally nitrogen limited; however, the variable anthropogenic inputs and a N:P ratio close to the Redfield ratio implies that nutrient limitation can easily switch to phosphorus in the Johor Strait.”

There are additional reports in the literature that refer to P limitation in the coastal/marine waters. Atmospheric inputs of bioavailable N and P reinforce the unusual N: P ratios and possible P limitation in the Southeast Mediterranean Sea (Herut et al., 1999) and phosphorous deficiency led to P limitation in the Mediterranean Sea (Thingstad et al., 1998; Krom et al., 1991). It should also be noted that as humans continue to preferentially increase nitrogen deposition (e.g. Galloway, et al., 1994), ocean ecosystems may shift from nitrogen limitation to limitation by other nutrients (e.g. phosphorus) or trace elements.

References:

Gin, K. Y. H., Holmes, M. J., Zhang, S., Lin, X.: Phytoplankton structure in the tropical port waters of Singapore, in: *The Environment in Asia Pacific Harbours*, edited by: Wolanski, E., Springer, Netherlands, 347–375, 2006.

Krom, M. D., Kress, N., Brenner, S., and Gordon, L. I.: Phosphorus limitation of primary productivity in the eastern Mediterranean Sea, *Limnology and Oceanography* 36, 424–432, 1991.

Thingstad, T. F., Zweifel, U. L., and Rassoulzadegan, F.: P limitation of heterotrophic bacteria and phytoplankton in the northwest Mediterranean, *Limnology Oceanography* 43, 88–94, 1998.

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Galloway, J. N., Levy II, H., and Kasibhatla, P. S.: Year 2020: Consequences of population growth and development on deposition of oxidized nitrogen, *Ambio*, 23, 120–123, 1994.

Comment 2: Be careful with significant figures. In several instances, more significant figures are reported that are probably warranted.

Authors' response: We provide an appropriate number of significant figures in the revised manuscript to be consistent with the precision of analytical measurements.

Comment 3: Overall, the manuscript suffers from poor clarity and is difficult to read. Suggest revising to make it easier to read.

Authors' response: The revised manuscript is prepared with improved clarity of the text and better presentation of data.

Comment 4: Superscripts: throughout the document, superscripts of chemical compounds (e.g. NO₃) are missing.

Authors' response: We apologize for this error. The missing subscripts of chemical compounds are given in the revised manuscript.

Specific comments:

Comment 1: How long were samples stored at 4 degrees C? Nutrient samples are more commonly stored frozen (-20° C) unless there is a very quick turnaround time (<24 hours) for analysis. What were the filters stored in? What were the HPDE bottles cleaned with (HCl and DI)?.

What QA/QC procedures were used for both lab and field methods? Were field blanks used to insure that samples were not contaminated (see comment below regarding P).

Authors' response:

Although -20° C storage is optimum for long term preservation, we observed that 4° C

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is still sufficient for short term storage of nutrient samples. Our environmental samples are analyzed as soon as possible, within 1 or 2 days (Karthikeyan and Balasubramanian, 2006; Karthikeyan et al., 2009a & 2009b).

The filters were folded in half lengthwise so that only surfaces with collected particulate matter are in contact and placed in the filter holder (glassine envelope). HPDE bottles (250 mL, Nalgene®) were filled with 2N HNO₃ for 3 days and subsequently with ultrapure water for 3 days. Finally, they were rinsed with ultrapure water three times and kept safely in plastic bags until use. No residual nitrate was found in the rinse water after washing.

The following QA/QC procedures were used for both lab and field methods. To ensure proper QA/QC, total sampling times, average actual volumetric flow rates, tare and gross-filter weights were checked for accuracy. Field blanks, obtained at regular intervals by placing filters in the filter holder with no air being drawn through them, were used to detect any contamination that may have occurred in the sample transport process. Likewise, laboratory blanks (unexposed filters) were also weighed and processed for quality assurance purposes. Field and laboratory filter blanks were analyzed for the nutrients species, and the values obtained for the blanks were subtracted from those of the filter samples. Both the quality assurance and quality control protocols were carried out regularly to avoid the analytical errors in the laboratory measurements. The analytical quality of the data obtained was determined using important parameters such as limits of detection, recovery, linearity, and by eliminating sampling artifacts (Karthikeyan and Balasubramanian, 2006; Karthikeyan et al., 2009a & 2009b). All filters used in this study were inspected for defects under bright illumination. All blank filters and filter samples were handled with stainless steel forceps. The filters were weighed before and after the sampling using a microbalance (Sartorius, Model MC 5) with 1 µg sensitivity to obtain the particulate mass. The balance was regularly checked with NIST-traceable standard calibrated weights. The filters were pre-equilibrated in a dry box with stabilized temperature (22–23° C) and relative humidity (30–35%) for at

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least 24 h before the actual weighing.

Comment 2: Page 7749, line 3: “millions” should be “million”

Authors’ response:

This error is corrected in the revised manuscript.

Comment 3: Page 7753, line 26: “Dominance of” should be “Dominance by”

Authors’ response:

As suggested, the correction is done in the revised manuscript.

Comment 4: The sections on DAD and WAD flux calculations are overly complicated and could be simplified, especially the WAD section.

Authors’ response:

As suggested, the WAD section is simplified in the revised manuscript.

Comment 5: It is surprising that organic P dominates the P flux, even during non hazy conditions (p. 7762, line 14 and Figures 5, 6). It is most commonly reported in the literature that either atmospherically deposited P is generally low, or that it is dominated by mineral P (i.e. dust). Do the authors have an explanation for why OP is high, even during non hazy conditions? Others have used organic P as an indicator for sample contamination (e.g. bird droppings, pollen). Without any explanation for why OP would be high, I’d be suspicious that you are seeing contamination here.

Authors’ response:

Regarding organic phosphorus, it has been reported in the literature that OP could account for about 30-80 % of the total P concentration. For example, Mahowald et al. (2008) reported the concentration of different forms of P from different sources including dust which indicated that the organic phosphate can be up to 50 % or more of the total P.

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Comment 6: It seems like the WAD flux numbers are based on a very small number of samples (p.7759, line 24), i.e. 6 samples total. I would be very careful about drawing conclusions from such a small number of samples.

Authors' response:

We agree with the reviewer that it is preferable to have a larger number of rainwater samples. However, 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. Whenever there were rain events, we collected the rainwater samples. The frequency of rain events that occurred during the biomass burning period was relatively less than that during the wet season. This study was focused on quantification of water soluble nutrients from dry atmospheric deposition (aerosol particulates) and wet atmospheric deposition (rainwater). Our research work represents the first study of its kind focused on bringing together field-based investigations to quantify atmospheric nutrient deposition and eutrophication modeling (presented in a companion paper) to investigate impact of atmospheric nutrient deposition on coastal water quality and provide a scientific basis for more in-depth future study in this region.

Comment 7: In the reference section, there are at least two journal titles which are wrong. First, "The Science of the Total Environment" is just "Science of the Total Environment." (p.7768, line 8). Second, "Environ. Interpret" is "Environment International" (p. 7769, line 22).

Authors' response:

We apologize for these errors. The correct journal titles are given in the reference section of the revised manuscript.

Comment 8: Table 1: It's not clear to me why the seawater concentrations are being shown here. I assume it is to show that the concentrations of DAD are greater than

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those in seawater to show its importance. The concentration of the aerosols aren't the critical factor to determining if it's important, it's the flux rate. If these are being shown for stagnant boundary level reasons, this will not be the determining factor in determining depositional velocity in marine systems. Suggest either removing the numbers for seawater, or better explain why this is important.

Authors' response:

The concentrations of seawater were given to indicate the baseline seawater concentration of nutrients and to show the similarity in the variation of N and P species between aerosols and seawater. We have now removed the seawater data from Table 1 to avoid the confusion. The importance of seawater concentration is explained in the companion paper "Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: eutrophication modeling".

Comment 9: Several figures (3,4,5,6) are very small and hard to read.

Authors' response:

Figures 3, 4, 5 and 6 are replaced with clear figures with larger size in the revised manuscript.

The authors gratefully acknowledge the reviewer's efforts on carefully reviewing the manuscript.

We revised our manuscript according to the reviewer's comments.

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

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