

## ***Interactive comment on “Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: eutrophication modeling” by P. Sundarambal et al.***

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AUTHORS NAME: P., Sundarambal, P., Tkalich, and R., Balasubramanian

TITLE: Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: eutrophication modeling

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Reviewer: Anonymous Referee #1

We would like to thank the reviewers for very constructive and detailed comments. We have made a number of changes and improved the overall quality of the manuscript by addressing the reviewers' comments as best as we could in the text.

**General comments** This paper focuses on the effect of Southeast Asian biomass burning on seawater nitrate levels in the Singapore Strait. The authors sampled nutrient deposition during days they designated as either influenced or not influenced by biomass burning. Based on high- and low-end nutrient deposition data they collected, they ran 20-day biogeochemical model simulations in the Singapore Strait to assess the effects of biomass burning on surface N & P concentrations. For comparison, they also sampled nutrient concentrations in the Strait during days influenced and not influenced by biomass burning.

While this is a very interesting and worthwhile topic, there were some major flaws in the study methodology. Some of the apparent flaws could stem from an incomplete description of the methods and results, and so the authors should more thoroughly describe and substantiate their methods (see specific comments below). In addition, the modeling aspects of the study were incompletely validated. If these issues can be addressed satisfactorily, then the authors can further improve their paper by comparing/contrasting their results with other relevant studies in other regions, and by paying more attention to appropriately referencing their statements. They should explicitly discuss some of the limitations of their assumptions. I would also strongly suggest that the authors get a native speaker to correct the manuscript. Additionally, the manuscript needs to be better organized, as currently portions that should be in the methods or results are in the wrong section (see technical comments). Authors' response: Thank you for the valuable comments and suggestions. The description of methods and results is revised to be in accordance with the reviewer's comments (both specific comments and technical comments) to eliminate apparent flaws in the revised manuscript.

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Using established methods, we have quantified different chemical forms of nutrients that undergo atmospheric deposition to the tropic environment for the first time. 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 inputs of nutrients across the air-water interface. This study also shows that biomass burning is an important contributor to coastal water eutrophication.

We wish to highlight that the present study aims at estimating the relative contribution of atmospheric nutrient deposition to coastal water eutrophication using a combination of the nutrients concentration data from field observations and the 3-D modelling program, NEUTRO.

For model verification, the figure below (Fig 1) showing the comparison between model predictions and field observations of water quality parameters in Singapore waters is provided in the revised manuscript. Simulations using NEUTRO model showed that the model output is able to achieve baseline levels, and is also able to reproduce correctly the general features and the observed fine patterns in the Singapore waters. More information on the model and its model validation is given by Tkalich and Sundarambal (2003), Sundarambal (2009) and Sundarambal and Tkalich (under review). Moreover, the current manuscript utilizes classical sensitivity-type investigation, assessing how the calibrated model responds to changes in the nutrients loading from the atmosphere, and compares that with the ocean fluxes.

Model results for baseline concentration simulation at a monitoring station on the South Coast of Singapore (Tkalich and Sundarambal, 2003; Sundarambal, 2009 [Sundarambal, P. Estimation of the contribution of atmospheric deposition to coastal water eutrophication. Ph.D. Thesis, Department of Chemical and Biomolecular Engineering, National University of Singapore, Singapore, 2009]) The present research work represents the first study of its kind that is focused on bringing together field-based investigations to quantify atmospheric nutrient deposition and eutrophication modeling to investigate impact of atmospheric nutrient deposition on coastal water quality. It also

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provides a scientific basis for a more in-depth future study in this region. The model results are compared or contrasted with other relevant studies in other regions and the statements made in the text are appropriately referenced in the revised manuscript.

The limitations of the modeling assumptions are briefly discussed in the revised manuscript as given below: -In order to delineate atmospheric and ocean fluxes, the model is tuned to a quasi-equilibrium state by assuming constant lateral fluxes; i.e., the boundary conditions are assumed to be constant in the vertical planes of the computational boundaries. -The water quality is run for 20 days to allow a new quasi-equilibrium state to be established (with atmospheric fluxes). The two quasi-equilibrium states are then compared. -Another assumption in this study is that the water-soluble nutrients of atmospheric origin are assumed to be deposited directly and uniformly onto the water surface and then transported through the water column by the action of tidal currents, while the nutrients undergo physical, chemical and biological processes and transformations in the water column.

The English language is improved considerably in the revised manuscript. More relevant explanations are included in the revised manuscript as and when needed to present the results obtained from the modeling study in a systematic and clear manner.

Specific comments Comment 1: The authors determined whether a day was influenced by biomass burning based on atmospheric haziness alone, which I did not find convincing. The authors should more thoroughly describe and substantiate why they used haziness as a metric for biomass burning. The PSI index was used to define haziness, but the PSI index itself was never defined. The authors should talk about the strengths and weaknesses of using this index. How do they separate pollution haze from biomass burning haze? Pollution is another very important source of atmospherically deposited N and so by using haziness as an index, they may overestimate the impacts of biomass burning. I suggest using other methods to validate their biomass burning day classification. These may include back trajectories coupled with satellite

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images of point biomass burning sources, aerosol optical thickness, FLAMBE models, etc. If they have any chemical characteristics in their samples that would help trace the degree of biomass burning influence (such as K+), that would also be ideal. Authors' response: The increased levels of nutrients in atmospheric samples during biomass burning are supported by back trajectories coupled with satellite images of point biomass burning sources and are explained in our companion paper. Also, the link between the present paper and the companion paper on field observations (Sundarambal, P., Tkalic, P., Balasubramanian, R., and He, J.: Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: field observations, *Atmospheric Chemistry Physics Discussion*, 10, pp. 7745–7778, 2010.) is now indicated in the introduction for its better understanding.

PSI stands for 'Pollutant Standards Index' which is an index developed by the United States Environmental Protection Agency (USEPA) to provide accurate, timely and easily understandable information about daily levels of air pollution. The National Environment Agency (NEA) in Singapore operates 13 air quality monitoring stations (NEA, 2006 and [http://app2.nea.gov.sg/psi\\_faq.aspx](http://app2.nea.gov.sg/psi_faq.aspx)). The air quality data are collected from 5 zones (north, south, east, west and central), and the 24 hour statistical average concentrations of the 6 criteria pollutants are used for the calculation of the PSI on a daily basis. The concentrations of major criteria pollutants (sulfur dioxide, nitrogen oxide, carbon monoxide, ozone and PM10) are measured daily. These pollutants are used in the determination of the PSI index. The US EPA advocated calculation of the index value on a daily basis for each of the five criteria pollutants and the reporting of the highest I value and identification of the pollutant responsible for the highest value. The PM10 measurements used in this study for the PSI calculation were obtained via the beta attenuation method. The air quality monitoring stations are positioned strategically in industrial, urban and sub-urban areas as well as along roadsides to monitor the ambient air quality.

The published information on the aerosol optical thickness and their chemical char-

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acteristics (such as K+) (See, S.W., Balasubramanian, R., Wang, W.: A study of the physical, chemical and optical properties of ambient aerosol particles in Southeast Asia during hazy and non hazy days, *Journal of Geophysical Research*, 111, D10S08, 2006.), FLAMBE and NAAPS models (Hyer, E. J., and Chew, B. N.: Aerosol transport model evaluation of an extreme smoke episode in Southeast Asia, *Atmospheric Environment*, 44(11), 1422-1427, 2010.) are included in the revised manuscript.

Comment 2: The authors sampled changes in concentrations of marine nutrients on hazy vs. non-hazy days, but in the Methods section, they indicated only very little about where they sampled. Based on 14 samples taken at an uncertain/poorly described location, time, and depth, a correlation between haziness and surface seawater nutrient concentrations was supposedly observed (although based on Fig. 1, I am not convinced it is a strong correlation at all). From this very small sample size, the authors draw the conclusion that on hazy days, atmospheric deposition is increasing surface seawater nutrient concentrations. However, even if there were a good correlation, a correlation does not indicate a causal relationship. Their interpretation of these data is particularly difficult to believe as a reader because the sampling area is inadequately described. The authors do not provide any indication on how surface nutrients vary normally over time, and thus how we can believe that the change observed was not just normal environmental variation. They should also provide more details about what their analytical precision/accuracy is. Finally, in their modeling results, they see a 1% increase in surface NO<sub>3</sub> due to high-end N deposition (I think, see comment #5). A 1% increase is not likely to produce observable differences in surface seawater, and this discrepancy is never addressed. Authors' response: We agree with reviewer that it is preferable to have a larger number of seawater samples. However, we collected seawater, particulate and rainwater samples at the sampling location, St. John's island, (SJI, see Fig 2 of the companion paper and it is indicated in Fig 2 of this paper in the revised manuscript) using established methods as frequently as possible to study the impact of biomass burning on ocean water quality through atmospheric deposition.

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It was found (Tkalic and Sundarambal, 2003) that the sampling point represents fairly well baseline characteristics of the Singapore Straits, and therefore could be a good representative of the large domain. This area is known to authors of having uniform vertical mixing of the water column.

The surface nutrients concentration vary uniformly over time in the Singapore Straits except Johor Straits where more fluctuations were observed depending on the monsoons and local sources of pollution (river discharges, runoff, ship spills, fish farms etc) (Gin et al., 2006). The spatial and temporal variability of the nutrients concentration in coastal waters of Singapore have been reported by Gin et al. (2000, 2006). The concentrations (which are used as model baseline values) in Table 3 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)).

Our research work represents the first study of its kind that is focused on bringing together field-based investigations to quantify atmospheric nutrient deposition (presented in a companion paper) and eutrophication modeling (this paper) to investigate impact of atmospheric nutrient deposition on coastal water quality. This study provides a scientific basis for a more in-depth future study in this region. In our future research, a local and regional long-term field monitoring program would be established to collect the representative temporal and spatial samples of dry atmospheric deposition and wet atmospheric deposition, as well as coastal water and offshore samples over the Singapore waters and Southeast Asia (SEA) region for measurement of nutrients and algal biomass based on the present research work.

The seawater sampling location is now provided in Fig 2 of the revised manuscript and also in the companion paper. In methods section, the details of atmospheric deposition and seawater sampling and analysis with relevant references are described with improved clarity in the revised manuscript. In this study, we have sampled limited samples seawater only at the surface level to establish seawater nutrients correlation with

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atmospherically deposited nutrients during hazy days. There is a moderate correlation, but not very strong, as this study has found that the contribution of atmospheric fluxes is significant, but typically limited to 4-24 %. The rest of the nutrients is supplied to the water bodies via ocean and land-based fluxes. The sensitivity study in section 3.1 (Fig. 4) describes increment of total mass of nitrate + nitrite in seawater due to atmospheric deposition of nitrite + nitrate nitrogen concentrations at different extremes. The percentage increase in total mass of nitrate + nitrite in seawater due to various atmospheric nitrite + nitrate fluxes over the seawater baseline values was in the order of 0.01 %, 0.13 %, 0.63 % and 1.26 % for nitrite + nitrate concentration of 1 mg/l, 10 mg/l, 50 mg/l and 100 mg/l, respectively (Fig. 4). Please note that the percentage increment of nitrite + nitrate nitrogen concentration in seawater (Fig. 5) was 2 to 30 % (mean ~ 15 %) during non-haze period and 5 to 111 % (mean ~ 70 %) during haze period based on conservative admixture assumption. To avoid confusion, the description of sensitivity analysis is rewritten with improved clarity.

The different chemical forms of nutrients (N and P species) were quantified using validated laboratory techniques (Sundarambal et al., 2007, 2009; Karthikeyan et al., 2009; APHA, 2005). The analytical precision/accuracy associated with the analysis of atmospheric samples is described in the companion paper (Sundarambal et al, 2010, field observation, ACPD) and for seawater as in Gin et al. (2000).

References APHA: Standard methods for the examination of water and wastewater, 21st edn, American Public Health Association/American Water Works Association/Water Environment Federation: Washington, D.C., USA, p.1368, 2005. 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. Gin, K. Y. H., Lin, X., and Zhang, S.: Dynamics and size structure of phytoplankton in the coastal waters of Singapore, *Journal of Plankton Research*, 22(8), 1465–1484. 2000. Sundarambal, P., Balasubramanian, R., Karthikeyan, S., and Tkalic, P.: Atmospheric deposition of nutrients

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and its role on coastal eutrophication in Southeast Asia, in: *Advances in Geosciences*, Vol. 9: Solid Earth, Ocean Science and Atmospheric Science, edited by Chen, Y.-T., World Scientific Publishing Company, Singapore, 149–166, 2007. Sundarambal, P., Balasubramanian, R., and Tkalich, P.: Atmospheric fluxes of nutrients onto Singapore Strait, *Water Science and Technology*, 59(11), pp. 2287-2295, 2009. Sundarambal, P., Tkalich, P., and Balasubramanian, R.: Modeling the effect of atmospheric nitrogen deposition on marine phytoplankton at the Singapore Strait, *Water Science and Technology*, 61(4), 859-867, 2010a. Sundarambal, P., Balasubramanian, R., Tkalich, P., and He, J.: Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: Field observations, *Atmospheric Chemistry Physics Discussion*, 10, 7745–7778, 2010b.

Comment 3: The details about the atmospheric and surface seawater sampling are never provided. We are referenced to many other sources for this information, e.g. on p. 7785, l. 5 they direct us to a companion paper (without reference). Without a reference, its difficult to evaluate whether the data collected were sampled well enough to be even be used in the context of the study. The paper would be better if the authors made it more of a stand-alone work. Authors' response: The details about the atmospheric sampling are given in the companion paper (Sundarambal, P., Balasubramanian, R., Tkalich, P., and He, J.: Impact of biomass burning on surface water quality in Southeast Asia through atmospheric deposition: Field observations, *Atmospheric Chemistry Physics Discussion*, 10, 7745–7778, 2010.) and those about the surface seawater sampling are provided with relevant references in the revised manuscript. We apologize for the unclear information given. The proper reference of the companion paper is given and proper link between the present paper and companion paper is discussed in detail in the introduction of the both papers. We hope this will provide clear information to the readers.

Comment 4: The model validation section needs a lot of work. For validation, the authors provide a conference proceeding abstract reference (P. 7790, l. 26). They also

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compare nutrients from a model run from one day with field observations (Fig. 3). However, we don't know the details about that those field observations. The authors only state that these samples were gathered "at a monitoring location in the East Johor Strait." We are not provided with any information about how representative these values are, nor are we told over what model domain the comparisons are averaged over for comparison purposes. We are not provided with any information about how good the hydrographic current representations are. The authors should show more model validation data. Authors' response:

Some of the concerns raised by the reviewer have already been addressed in response to the Comment 2. The focus of the present paper is only to assess the magnitude of atmospheric nutrient fluxes by allowing a quantification of the relative contribution of atmospheric and ocean fluxes in the Singapore seawater and to estimate the relative contribution of atmospheric nutrient deposition to coastal water eutrophication using the quantified atmospherically deposited nutrients data and the 3-D modeling program NEUTRO. This method is similar to a sensitivity study which emphasizes on the analysis of model response to the input variability. This method is considered to be robust, and in fact often serves as one of the important steps in the model validation.

Simulations using NEUTRO model showed that the model output is able to achieve baseline levels (see Figure shown in response to general comments) and is also able to reproduce correctly the general features in the Singapore waters by using appropriate kinetic coefficients and other parameters (Tkalich and Sundarambal, 2003; Thesis Report of Sundarambal, 2009). The model performs well and its performance is comparable with that of best-established modeling practices and standards in the region (Sundarambal and Tkalich, under review 2010).

The scope of the present paper is to demonstrate the application of the eutrophication model for atmospherically deposited nutrients and to estimate the resultant changes in water quality from the baseline values due to atmospheric deposition of nutrients only. However, the model will be run with all possible sources of nutrients in the model

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domain in our future study. Model predictions will then be compared to actual field observations of nutrient concentrations in seawater.

The information on the hydrographic current representations was provided by Chan et al. (2006) and the description of important hydrodynamic characteristics of the Singapore waters is included in the section 2.3.2 on Marine Hydrodynamic Model (TMH).

Comment 5: The authors saw at the most, a 1.36% increase in NO<sub>3</sub> “total mass” in the water column (Fig. 4) due to atmospheric N deposition. Firstly, they should explain what they mean by “total mass.” This is not a commonly used or intuitive unit (total mass of what?). Secondly, I am confused because in the abstract they say that “computations showed that atmospheric fluxes might account for up to 17–88% of total mass of nitrate nitrogen in the water column during hazy days and 4 to 24% during non-hazy days.” On p. 7797 they say that, “atmospheric fluxes might account for an increase of nitrite + nitrate nitrogen concentration in water column in the range of 1–16% (mean 9.3%) and 5–76% (mean 45%) during non-haze and haze periods, respectively.” Why are these numbers inconsistent? Because of this confusion, I have difficulty interpreting their statement in the conclusions from p. 7795, l. 5 that “Increased atmospheric nutrient fluxes, even as much as 100 times above the typical atmospheric nitrogen flux, could cause eutrophication in nearshore waters of Singapore and surrounding waters and also areas where tidal action is low”? This conclusion needs much stronger support/explanation (and the sentence itself needs to be re-written). Authors’ response: The sensitivity study in section 3.1 (Fig. 4) describes increment of total mass of nitrate + nitrite in seawater due to atmospheric deposition of nitrite + nitrate nitrogen concentrations at different extremes. In Fig. 4, “total mass” means the total mass of nitrate + nitrite nitrogen in the water column due to atmospheric N deposition and is indicated clearly in the revised manuscript.

The sentence in the abstract “computations showed that atmospheric fluxes might account for up to 17–88% of total mass of nitrate nitrogen in the water column during hazy days and 4 to 24% during non-hazy days.” shows the results obtained from the

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conservative admixture assumption in terms of total mass of nitrate + nitrite nitrogen in the water column.

The sentence on p. 7797 “atmospheric fluxes might account for an increase of nitrite + nitrate nitrogen concentration in the water column in the range of 1–16% (mean 9.3%) and 5–76% (mean 45%) during non-haze and haze periods, respectively.” shows the results obtained from the non-conservative admixture assumption (with full eutrophication kinetics) in terms of nitrate + nitrite nitrogen concentration in the water column.

Clear explanations with relevant references are given in the revised manuscript with no inconsistency of numbers so that the conclusions from p. 7795, l. 5 can be easily understood, as per reviewer’s suggestions.

Comment 6: Section 2.2. Although wet and dry deposition data were apparently sampled, it is later stated that only wet deposition was simulated (e.g. p. 7793). The authors should justify why they only simulated wet deposition and they should add something about how this may affect their results. Authors’ response: In this study, we have modeled only wet atmospheric nutrient deposition because as it was clearly observed by Sundarambal et al. (2009) and Sundarambal et al., 2010b (the companion paper) that the wet atmospheric deposition was more dominant than the dry atmospheric deposition. The justification for inclusion of only wet deposition in the modeling study is included in the revised manuscript. The details of model simulations by dry deposition alone and together with wet deposition and its effect on water quality will be presented elsewhere with a more extensive database.

Comment 7: I understand that only NO<sub>3</sub> was added to the model from deposition. However, biomass burning is a large source of water soluble organic N, as well as NH<sub>4</sub><sup>+</sup>. The authors should talk a little bit about the uncertainty of excluding these pools of potentially bioavailable nutrients may affect the interpretation of their results. Authors’ response: We quantified all potentially bioavailable nutrients (N and P species) from atmospheric deposition (mean concentrations S<sub>j</sub>WD in Table 2 and seawater base-

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line in Table 3) and subsequently incorporated them into the simulation of model with full eutrophication kinetics (section 3.3, non-conservative admixture assumption). The sentences in the manuscripts are revised to provide clear explanation.

Comment 8: The title is inappropriate for the papers' focus- while biomass burning aerosols presumably enter the study region from all over SE Asia, the oceanographic region of focus is only the Singapore Strait. Therefore, it is misleading to indicate that the paper addresses water quality in all of SE Asia. Authors' response: We agree to reviewer's suggestion. However, the model domain covers the Johor Straits in North and some part of Java Sea in South as shown in Figure 2. The companion paper deals with the impact of biomass burning on regional air quality in SE Asia based on field observations. We therefore would like to retain the current title.

Comment 9: The authors should provide more background in the introduction on the magnitudes of fluxes of nutrients from biomass burning vs. the concentrations of nutrients in the water column. a. First, how does biomass burning compare with other sources of atmospherically deposited nutrients? The authors state in P. 7781, l.25 that "Most of local knowledge regarding contamination due to forest fires (biomass burning) originates from earlier studies conducted elsewhere, at various parts of the world (e.g., The United States, Australia, Brazil, Mexico, Africa). However, the results of these studies are of little use in assessing the environmental impacts of the resulting pollution since their main objective was to quantify the flux to the atmosphere of various trace gases such as CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O from biomass burning." This is not correct, there are plenty of relevant data that they can and should cite. A more thorough literature review should be conducted and then the authors should discuss in the introduction the relevance of these studies in context of their study region. b. The authors should also provide the reader with some idea about the existing concentrations of nutrients in the Singapore Strait. They should provide more justification on why it is reasonable to believe that atmospherically deposited fluxes of biomass burning N can affect the existing nutrient pool in this region. Authors' response: a. The background information

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on nutrients from atmospheric deposition (the companion paper and its supplementary material) is given in the revised manuscript. As per reviewer's suggestion, the manuscript is revised with inclusion of relevant references and the pertinent contained in them. b. The background information on nutrients in the water column with relevant references (Sundarambal, P., Estimation of the contribution of atmospheric deposition to coastal water eutrophication, Ph.D. Thesis, Department of Chemical and Biomolecular Engineering, National University of Singapore, Singapore, 2009.) is given in the revised manuscript.

Comment 10: The study was based on a 20-day model simulation, but the impact of anthropogenic N inputs to ocean areas will probably have more important long term impacts based on longer-term accumulation rather than on episodic impacts. Why were only short-term changes investigated? The authors may consider mentioning longer-term impacts as a future issue, although not one addressed in this study. Authors' response:

The model was run for 20 days to achieve steady-state/equilibrium conditions which reflect the long-term water quality changes due to a constant pollution loading from sources; the present study is focused on atmospheric deposition only. The equilibrium conditions without atmospheric fluxes correspond to long-term monitoring observations. What might be lacking are the short-term non-equilibrium responses which could be addressed in the future study, but for this a more extensive database is required. Please kindly note that the focus of the present study is only to assess the magnitude of atmospheric nutrient fluxes by allowing a quantification of the relative contribution of atmospheric and ocean fluxes in the Singapore Strait and to estimate the relative contribution of atmospheric nutrient deposition to coastal water eutrophication using the quantified nutrients data and the 3-D modeling program NEUTRO.

Comment 11: Food webs were not really discussed, and so I think the authors should either take any mention of food webs out of their abstract or discuss food webs more in the results and discussion/conclusions sections. Authors' response: The word "food

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webs” is removed from the abstract. The model takes into account of phytoplankton and zooplankton dynamics and the sentence in the abstract is revised accordingly.

Comment 12: This study needs to be put in better context of other relevant studies (e.g. Krishnamurthy et al., 2007, 2009, 2010; Zamora et al., 2010, etc.). For example, their observation of P enrichment due to atmospheric deposition is in direct contrast to these other studies. Similar studies modeling the affect of atmospheric nutrient deposition to the ocean and coastal areas should be referenced, and compared/contrasted to the results. Authors’ response: We thank the reviewer for the above suggestion and the manuscript has been revised accordingly.

Technical comments Comment 1: As mentioned previously, there are numerous errors in the English. For example, in the abstract, it is “plankton” not “planktons.” However, these errors are too numerous to point out- I leave that responsibility to the authors of finding a native speaker to correct these errors. Authors’ response: As per reviewer’s advice, the manuscript has been revised considerably to improve the English.

Comment 2: Section 2.8, p. 7793: The authors’ state that Case I indicates the effects of physical oceanography, Case III indicates the effects of atmospheric deposition + physical oceanography, and Case II indicates the affects of atmospheric deposition. So therefore Case II=Case III-Case I. Why run Case II in the first place? Authors’ response: We agree with reviewer. Case II in the section 2.8 is run to visualize the relative contribution of atmospheric deposition flux only when compared to that of regional ocean flux.

Comment 3: P. 7782, l. 2: reference? Authors’ response: References, Sundarambal et al, 2007; 2009, are given in the revised manuscript.

Comment 4: P. 7782, l. 5: “SEA surface waters receive a large nutrient supply of which a substantial portion is of anthropogenic origin.” Reference? Is this for coastal or open ocean waters in SE Asia? Authors’ response: The sentence is rewritten for coastal waters with the relevant reference.

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Comment 5: P. 7782, l. 6 “Accelerated eutrophication and its subsequent effects such as nuisance algal blooms and reduced oxygen levels pose significant problems for coastal waters and aquatic ecosystems in SEA. Algal blooms resulting from complex coupled physical/biological processes are steadily increasing in coastal waters.” Reference? Authors’ response: The relevant reference is now included in the revised manuscript.

Comment 6: 7783, l. 1: “Besides the advection-diffusion redistribution, a series of terms for the biochemical interactions between non-conservative quantities is considered.” Redistribution of what? Authors’ response: The word “redistribution” is replaced by “transport”.

Comment 7: Section 2.1: There is a lot irrelevant information in here, the authors should take out anything not directly related to their study, and add more about the oceanographical/hydrological region of focus. For example, they should talk about the relevant currents, depth of the Strait, whether rivers impact the area, any freshwater lenses that may develop after a rain event to prevent immediate mixing of rain with the rest of water column, etc. They should add latitude/longitude to their map (Figure 2). Authors’ response: In Section 2.1, we thought is good to provide information on the study area. Any information that is not directly relevant to the present study is removed. The suggested information about the relevant currents, depth of the Strait, whether rivers impact the area, etc are reported by Chan et al. (2006) and the description of hydrodynamic characteristics of the Singapore waters is included in the section 2.3.2 on Marine Hydrodynamic Model (TMH). The additional information as per reviewer’s suggestion is now included in the revised manuscript. The latitude/longitude is added to the map in Figure 2.

Comment 8: Section 2.2: The first two sentences belong in section 2.1. Authors’ response: The sentence is moved to Section 2.1 as suggested.

Comment 9: P. 7785 l. 16- units of years should not be compared with units of days

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in the same sentence. Authors' response: We agree with reviewer. The unit of flux is corrected as suggested.

Comment 10: Section 2.2. Any discussion of results (e.g. regressions) should not be in the methods section, but rather should be in the results. Authors' response: The regression study results are moved to results and discussion.

Comment 11: P. 7788, l. 25: the printed flux equation is wrong, the correct version is:  $F = \text{settling velocity} \times \text{concentration}$ . There is no surface area component that I am aware of, and the units don't work out if you add in surface area anyways Authors' response: Thank you for pointing out this error in the calculation. The sentences related to mass flux in P. 7788, l. 24-25 are corrected accordingly.

Comment 12: Section 2.3.2., lines 7-17: this should be in the methods section site description Authors' response: As per reviewer's suggestion, the description of typical hydrodynamic characteristics of the Singapore coastal waters is moved to section 2.1 The study area and the hydrodynamic data used in this modeling study are now included in section 2.2 Data.

Comment 13: Where is the reference for the 2136 mm/yr rainfall rate? Authors' response: Reference for the 2136 mm/yr rainfall rate is provided in the revised manuscript.

Comment 14: P. 7794, l. 17-23 belongs in the methods section. The authors should state why they used the NO<sub>3</sub> concentrations indicated on line 23 and how these values are environmentally relevant, particularly with respect to biomass burning Authors' response: Thank you for the suggestion. In P. 7794, l., line 17-23 is moved to the methods-Section 2.7. The explanations pertaining to the NO<sub>3</sub> concentrations are included in the revised manuscript.

Comment 15: P. 7797 l. 25: reference to Fig. 6 is a typo. Authors' response: The typo error is corrected.

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Comment 16: P. 7798, l. 1: Put in a qualifier, algal growth in the marine environment is not always N limited. Authors' response: It is amended accordingly as per the reviewer's suggestion.

Comment 17: P. 7800, l. 9: how was NEUTRO enhanced and why is this relevant? Authors' response: The original NEUTRO model, used in our earlier studies, did not have a module to address the nutrients loading from atmospheric deposition. The model is improved with features to include atmospheric deposition of nutrients so that its impact on water quality can be studied and quantified.

Comment 18: The authors' state in their conclusions that "It was found that nutrient loading onto the coastal and estuarine ecosystems of the Singapore and surrounding countries from the atmospheric wet and dry deposition during hazy days was remarkable, the contribution being between 2 and 8 times that of non-hazy days." If they still have reason to believe this after more rigorously confirming that those days were primarily influenced by biomass burning, they should state here actual concentrations, compare those concentrations to other locations, and state that this conclusion is based off field data, not model data. Does this conclusion belong in the companion paper? Authors' response: We agree with reviewer and the conclusion belongs to the companion paper. However, we observed the similar increase in water quality as seen from the present paper. As per the reviewer's suggestion, the statements in the conclusion are rewritten in the revised manuscript.

Comment 19: P. 7800, l. 25: "The results of the present study depict that the impacts of nitrogen species through AD onto the coastal region are more significant than phosphorus species." The authors should spend more time talking about P and Fig. 7 if they want to make this point in their conclusions. Otherwise, they should take this out. Authors' response: Yes, we agree with reviewer's suggestion. P and its results are discussed in more depth in the revised manuscript to support the conclusion in P. 7800, l. 25.

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Comment 20: P. 7801, l. 1: Sewage is never mentioned. Talk about it earlier or take it out. Authors' response: Sewage is taken out from the text.

Comment 21: P. 7780, l. 24: "fixed and organic N" doesn't make sense. Organic N can be fixed N. fixed N is N that was originally N<sub>2</sub> in the atmosphere but was captured and transformed into reactive N by diazotrophs. Authors' response: Thank you for pointing out this error. We have rephrased the sentence.

Comment 22: P. 7780, l. 26- Add Mahowald et al., 2008. Authors' response: Mahowald et al., 2008 has been added in line 26, P.7780.

Comment 23: P. 7781, l. 19- El Niño Authors' response: El Nio is replaced by El Niño.

Comment 24: Table 2: seawater baseline units? Authors' response: The seawater baseline unit is now included in Table 2.

Comment 25: Figure 2: Show sampling sites on the map. Authors' response: The sampling site, SJI, is now shown on the map in Figure 2.

Comment 26: Fig. 4: "C" for concentration is easily confused with C for carbon/biomass. Use "DIN" instead. Authors' response: Fig. 4: "C" is replaced by "DIN".

Comment 27: Figs. 6,7,8: scale is difficult to read, adjust and make the font larger. Authors' response: Figs. 6,7,8: Scale are adjusted and made larger as suggested.

The manuscript is revised with proper explanations in the text to ensure that it is conveyed clearly. Also, the clarity of the presentation of the study is checked and improved where needed. The explanations of figures in the text and figure caption are also checked for their clarity. We thank the reviewer for the valuable comments and suggestions to improve the quality of the manuscript.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/10/C5138/2010/acpd-10-C5138-2010-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 7779, 2010.

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Figure 1

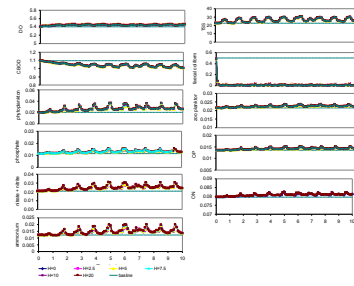


Fig. 1.

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