Paper review #2:

Again, we wish to thank Reviewer 1 and the Co-editor for their time and comments on our manuscript. This second review is focused on clarifying the difference between the forward runs used for the attribution of biomass burning and the backrun used to generate the air history maps used to illustrate the general wind patterns of the air influencing Singapore. We also add explanations regarding the met data used and how the background concentrations were estimated.

For clarity we are attaching a pdf document highlighting the changes between the original submission and the manuscript following the two revisions. We hope that these additions, in addition to the improvements from the previous review, will accommodate the expectations of the reviewers and Co-editor.

From Co-editor:

Please take careful account of the comments of Referee 1 when preparing your revised manuscript. In addition, I have some more requests based on the review of and your responses to Referee 2.

First, I suggest that you include your assessment of UM model against observation data available in the manuscript. It was included in your response but not in the modified manuscript. Overall dispersion models are highly sensitive to their meteorological inputs and it is necessary to first evaluate these errors and discuss the uncertainties.

Second, I suggest adding a sentence or two to the manuscript on why you used inconsistent settings and resolutions for each year and if these inconsistencies have any impact on the results.

- The key thing to note is that the UM is an operational NWP model. The paragraph has been edited to the following, including the assessment of the UM:

“The Unified Model (UM) is the Met Office’s operational numerical weather forecast model. The UM is a global model based on the non-hydrostatic fully compressible deep-atmosphere equations of motion solved using at semi-implicit semi-Lagrangian approach on a regular longitude-latitude grid (Walters et al., 2017). Archived analysis meteorology from the global version of the UM was used to drive NAME. As the UM is an operational model, the dynamical core and spatial resolution have changed throughout the period, from ~40 km over ~25 km to ~17 km resolution. However, for the majority of the study the resolution is constant at 25 km. These upgrades are described in Walters et al. (2011, 2017), and the relevant changes for dispersion modelling are summarised in Table 1. These changes are not expected to have a significant impact on the results, e.g., no significant differences in the deposition are seen across the change from instantaneous precipitation and cloud to 3-hour mean data in 2013.

“Global UM model meteorological data for 2013 have been evaluated using meteorological observations available at four sites across Singapore. The UM data are interpolated in NAME to obtain wind speed and direction, temperature, and relative 20 humidity data for each location and an hourly time resolution. The results show that modelled wind speeds are higher on average than those observed during 2013 particularly during the monsoon seasons. Wind speeds are one of the most important factors affecting pollutant levels, particularly close to strong sources. Although haze in Singapore is predominantly caused by long range transport of biomass smoke, the higher wind speeds in the model may contribute to reducing modelled pollutant levels below those observed. There are some differences in wind direction between the model and observations, but the prevailing wind directions are captured well throughout the year.

“Observed ambient temperatures are slightly higher and more variable on average than the model, although there is good agreement between the model and observations. Rainfall does not appear well represented with higher hourly means and more frequent low intensity events when compared to the observations, which show less frequent high-intensity rainfall associated with the convective activity that dominates rainfall within the tropics. Modelled total monthly rainfall is higher than observed during 2013, which may decrease modelled PM levels through wet deposition and contribute to the often negative bias observed in PM10, see Sect. 3. As discussed in Redington et al. (2016) and Hertwig et al. (2015), the uncertainties from the meteorological data feed into the dispersion simulation.“

Start date	Approx. horizontal resolution	Relevant change
01/1/2010	40 km	
20/1/2010	25 km	Horizontal resolution increase
30/4/2013	25 km	Change from use of instantaneous precipitation and cloud to 3-hour mean data
15/7/2014	17 km	Horizontal resolution increase

Minor: please add the acronyms definitions to the caption for Table 1. (ASO: August-October, MJJ). Also, please reduce the number of labels on the y-axis of Fig. 4 (e.g. labels at every 200 instead of every 100). Perhaps it would be useful to improve the quality of this figure to improve readability"

- The acronym definitions have been added to the Table caption (now Table 2). The comments on the figure axes have been accommodated. Finally, the quality of the figures is significantly reduced during the upload, we would be more than happy to provide higher quality files.

From Reviewer 1:

Review of revision of Hansen et al. for Atmospheric Chemistry and Physics

General Comments

In the revised submission of "Haze in Singapore - Source Attribution of Biomass Burning from Southeast Asia", Hansen et al. have responded to some of the concerns expressed in the first round of reviews. The manuscript is strengthened somewhat, but conflicting and unclear statements remain. Additionally, the grammar still needs to be refined though the duplication of the Discussion and Conclusions has been somewhat mitigated.

To the concerns expressed that the degree of agreement between the observations and model do not constitute a case for "source apportionment", the authors only replied in agreement that the model performance was worse than they desired. If the authors think that a comparison of NAME results with observations is worth sharing with the community, they need to consider another title for the paper that does not involve "source apportionment". They have not modeled any other sources of PM10 nor have they made an effort to determine a statistically reasonable background concentration across the years.

- In the previous response to reviewers, background concentrations at the two stations were estimated based on averages of the PM_{10} concentrations during non-haze periods, this follows the method used by Kim et al, 2015:

"We estimate the smoke concentration at each site in the observations by subtracting as baseline the mean concentration for the bracketing non-burning months (June and December)."

https://doi.org/10.1016/j.atmosenv.2014.09.045

The text now reads:

"Observations of PM10 in Singapore from 2010 - 2015 show an overall background concentration during months of little or no burning of between 23 - 29 µg/m3 at the two monitoring stations. These values fit well with those determined in other studies for Singapore. For example, Hertwig et al. (2015) estimated background concentrations for PM10 to be around 30 µg/m3, based on the 2013 haze episode. In general, both background and peak concentrations vary between NTU and TP. Following the approach of Kim et al. (2015) we assume a constant background of 25 µg/m3 for the PM10 observations at both sites and subtract this value from the observation time series. "

If I understand p. 7, l. 25-7 correctly, they are simply comparing a single back trajectory model convolved with satellite-based fire emissions estimate to observations at two distinct sites, which it would be a misrepresentation to call source apportionment.

- This is not correct. For clarity: we have performed two kinds of simulations:

- Forward year-long simulations using emissions from all of the areas depicted in Figure 1, which the main results (time series and pie charts) are based on.
- Back-runs with numerous particles (also known inverse simulations), which are only used for the air history maps. These have been included as a means to provide the reader with an indication of the region of influence on the air reaching Singapore.

We had thought that this was clear in the original text, but acknowledge that this was obviously not the case. The differences between the two simulations have been highlighted in the text where relevant, e.g.:

- P4, line 9: "…biomass burning, and forwards and backwards dispersion modelling to study how…"
- P8, line 29: "In addition to the forward simulations with the GFAS biomass emissions, backward (inverse) runs were conducted with NAME using the UM…
- P12, line 14: "The timeseries and pie charts are based on results from the forward NAME simulations."

And the subsection on air history maps was revised to:

"Air history maps provide a visual indication of where air at a given location has originated from. This helps to determine the regions that influence the composition of the air arriving at this location. To construct air history maps for Singapore, backward (inverse) runs were conducted with NAME, in addition to the forward simulations with the GFAS biomass burning emissions (Sect. 2.3). Fig. 2 illustrates the air history map for Singapore for the years 2010 to 2015. For each day in the six year period from 2010 to 2015, a 10-day backrun was conducted using meteorological input from the UM global model within a domain of 90.0◦E, 140.0◦E, 15.0◦S, 23.0◦N (Fig. 2). PM10 was emitted as a tracer from a receptor site in central Singapore and model particles were released over the first 24 hours with an emission rate of 1 g/s. The resulting concentration values in the 0-2km layer were output on a 0.1◦ × 0.1◦ resolution grid and integrated backwards in time for 10 days with a timestep of 10 minutes. By summing the results from several runs, air history data can be produced for different seasons and years, as well as the total for the whole period. A higher integrated concentration indicates that more air has passed through a grid cell on route to the receptor site, compared to a grid cell with a lower concentration. For each analysis period, the multiple corresponding 10-day air concentrations were summed for each grid cell and for the total domain. A percentile value was then calculated to ascertain the proportion of air influenced by a particular grid cell vis-à-vis other areas.

Comparison between the inland site and a coastal receptor site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island when averaged over time. The results of the air history simulations helped inform the decision of domain size for the forward haze simulations. ”

Since the authors did not address the major issue raised in the first round of reviews nor all of the specific comments that were made, it is not clear that they are willing or able to do so. Accordingly, I will not recommend publication in Atmospheric Chemistry and Physics unless they are able to revisit the original comments, some of which are repeated here, and address them thoroughly.

- At the risk of repeating ourselves, we have gone through the previous review comments below, to clarify and elaborate on our responses where relevant.

Review 1:

General Comments

The major difficulty in this work seems to be representing PM10 concentrations in Singapore except when biomass burning occurs very close by the sites (e.g., Riau or the Malaysian Peninsula) (Fig. 4). The two times this occurred during the modeled period are labeled as “Atypical haze” (Sect. 3.1). As the authors note, the PM10 concentrations in Singapore, especially in the other “haze seasons”, are likely from diverse sources; however, the time series (Fig. 4) and statistical analysis of the agreement between the model and observations (Tables 1, 2) do not support the notion that “Southeast monsoon season haze” (Sect. 3.2) is represented accurately enough by the model to claim attribution of these sources to biomass burning regions. The difficulty may result from the lines of source regions not being stacked in the time series plot (Fig. 4), but the correlation statistics are fairly poor for periods other than 2013 MJJ, which supports the interpretation of the time series that the model is not representing concentrations well. One reason may be that the PM10 from biomass burning from regions not adjacent to Singapore is not well represented by the modeled processes. Another reason may be, as noted by the authors, that much of the haze comes from biomass burning sources not detected by the model. Finally, it may be that the PM10 is not from biomass burning. The first two causes would indicate that this modeling framework is not appropriate for attribution of biomass burning contributions to PM10 concentrations in Singapore. The final potential explanation could be shown by more sophisticated analysis of the background PM10 concentrations rather than using a fixed value of 25 μg/m3; if this explanation does not hold, it seems unreasonable to claim that these attributions are appropriate for episodes other than those in which the Riau and Malaysian Peninsula contributions dominate.
Given the difficulty of interpreting the results, the weakness of the Discussion and Conclusions sections seem reasonable. Specifically, the Conclusions seem to be very repetitive of the Discussion. Accordingly, I recommend this manuscript for publication in Atmospheric Chemistry and Physics once this major issue has been

addressed. Additionally, specific comments have been included below that should also be considered in the revisions of the manuscript.

We thank the reviewers for their constructive feedback and hope that our subsequent changes to the manuscript has improved the content and readability of the paper.
We acknowledge that our results are not as good as would be desired, but this work now acts as a stepping stone to improving the understanding of haze in the region and how to model haze at high temporal and spatial resolution. And we feel it is worth sharing our findings with the community. We have also highlighted uncertainties in the work, including the limitations to emissions and focussed on the four events with the best correlation between model and observations.
We have rewritten and restructured the Introduction and removed the Discussions section to make the Results and Conclusions clearer and more coherent. The figures have been updated following the reviewers' suggestions.

Specific Comments

Line Comment

p. 1, l. 8-13 The meaning of this interpretation of the results is unclear especially in the phrases "several and varying source regions", "atypical haze episodes … characterized by atypical weather conditions", and "different set of five regions dominate". Please refine.

- The paragraph has been changed to:

As should be expected, the relatively stronger Southeast monsoonal winds that coincide with increased biomass burning activities in the Maritime Continent create the main haze season from August to October (ASO), which brings particulate matter from varying source regions to Singapore. Five regions dominate as the source of pollution during recent haze seasons. In contrast, off-season haze episodes in Singapore are characterised by unusual weather conditions, ideal for biomass burning, and emissions dominated by a single source region (for each event). The two most recent off-season haze events in mid-2013 and early-2014 have different source regions, which differ to the major contributing source regions for the haze season.

p. 1, l. 14 "climate" seems inappropriate when only 5 years have been considered.

– The study covers 6 years, but more pertinently also considers the impact of El Nino and other non-weather timescale phenomena. The use of the word "climate" in this context seems appropriate and is supported by other literature.

p. 2, l. 3 "Though the popular press often attribute" - grammatical error. Also, scientific literature has supported similar conclusions (Kim et al., 2015).

– Reference added as suggested:

"Scientific studies such as Kim et al 2015 as well as the popular press often attribute peatland destruction and related haze in the region to Indonesia (Reid et al 2013), however, the haze cannot be attributed to only one region or country alone."

p. 2, l. 17-9 Please insert a comma as "approach, and source" or divide these two thoughts into separate sentences. What type of source apportionment was applied by Lee et al. (2017) and Engling et al. (2014)? Please be more specific.

– more details have been added to the text:

"Several previous studies have looked at attributing air pollution for different regions. Source attribution can be performed both through modelling and by looking at observations of air pollution in detail. For example, Heimann et al. (2015) carried out a source attribution study of UK air pollution using observations to distinguish between local and regional emissions, whereas Redington et al. (2016) estimated the sources of annual emissions of particulate matter from the UK and the EU by using the NAME model to look at threshold exceedences and episodes. Attribution studies have been performed using Eulerian models such as GEOS-chem, CMAQ, and WRF-STEM to study both Asia and the Arctic (Ikeda et al., 2017; Kim et al., 2015; Sobhani et al., 2018; Yang et al., 2017; Matsui et al., 2013) sometimes in combination with flight campaigns (Wang et al., 2011) to better constrain the emissions. Lagrangian models have also been used in combination with observations by Winiger et al. (2017). Combinations of Eulerian and Lagrangian models (Kulkarni et al., 2015) and Eulerian models and observations (Lee et al., 2017b) have been used to assess whether low visibility days were caused by fossil fuel combustion, biomass burning or a combination of the two. In Southeast Asia, Reddington et al. (2014) used an Eulerian model to study haze and estimated emissions through a bottom up approach. Source apportionment for studies of biomass burning related degradation of air quality and visibility in Southeast Asia has also been applied by Lee et al. (2017a) who used the WRF model to study the sensitivity of the results to different met data and emission inventories and Engling et al. (2014), who used observations and a chemical mass balance receptor model to compare the chemical composition of total suspended particulate matter on haze and non-haze days during a haze event in 2006."

p. 2, l. 20-2 Run-on sentence. Please correct here and throughout the manuscript (e.g., p.3, l.8-12, etc.)

– corrected:

"Haze concentrations in Singapore vary throughout the six year period from 2010 to 2015. Even though biomass burning contributes to (low) PM10 concentrations in Singapore throughout large parts of the year, some peaks in the PM10 observations can be explained by haze almost exclusively."

"In terms of biomass burning, the year in this region can be divided into seasons that relate to the monsoon seasons: FMA dominated by burning in Mainland Southeast Asia; during May, June, and July (MJJ) burning starts in northern Sumatra and traverses southward; ASO is characterised by burning in Southern Kalimantan and, in general, there is little or no burning influencing Singapore in November, December, and January (NDJ) (Campbell et al., 2013; Chew et al., 2013; Reid et al., 2012, 2013)."

p. 2, l. 26-7 Poor sentence construction leads to a lack of clarity.

- corrected:

“These events caused extremely high PM10 concentrations in Singapore.

p. 3, l. 13-4 “related haze events are linked” is redundant.
– As this section is used to link biomass burning and meteorology, we want to emphasise that the weather reports link weather and haze events.

p. 3, l. 23-4 A strong case for using dispersion modeling has not been made in the Introduction.
–more details have been added to the Introduction and significant text to make the case stronger, see comments above and the section below:
“The aim of this study is to investigate spatial variation of haze across Singapore through source attribution, including the variation in concentration and the contributing source regions across Singapore depending on the distance to source regions and the seasonal variation by looking at four recent haze events occurring during different seasons between January 2010 and December 2015. This is done by linking meteorology, biomass burning, and dispersion modelling to study how the origin of haze has varied across Singapore during this whole period. Fire radiative power and injection height from the CAMS global fire assimilation system (Kaiser et al., 2012) and higher resolution land-use data from the Centre for Remote Imaging, Sensing and Processing at the National University of Singapore have been used to calculate PM_{10} emissions from biomass burning in 29 defined source regions in Southeast Asia. Using the Met Office’s numerical weather prediction (NWP) model to drive the Numerical Atmospheric-dispersion Modelling Environment (NAME), a Lagrangian particle trajectory model, we are able to attribute the haze arriving in Singapore to its source region and study the difference between major contributing source regions at a western and an eastern monitoring station in Singapore.”

p. 4, l. 31 Please provide a reference or equation for the MNMB.
- the following reference has been included in the text:
Seigneur, C., Pun, B., Pai, P., Louis, J-F., Solomom, P., Emery, C., Morris, R., Zahniser, M., Worsnop, D., Koutrakis, P., White, W., and Tombach, I.: Guidance for the performance evaluation of three dimensional air quality modelling systems for particulate matter and visibility, J. Air Waste Manage. Assoc., 50, 588–599, 2000.

p. 4, l. 35 Please clarify here, as is stated later, that the emissions are emitted over a 24-hour period at the rate of 1 g/s. Please state how the same emissions rate results in different total emissions from a single fire (if it does).
– this emission rate only applies to the calculation of the air history maps, the text here is consistent with the biomass burning simulations. The section regarding the calculations of air history maps has been modified for clarity, see below.
Review #2: The Methods section has been divided in to subsections to add clarity to the difference between the simulations. The description of the air history maps has been revised for clarity again, please refer to the updated text above.

p. 5, l. 7 Please note the limitations of comparing a tracer with PM10.
– please refer to reply to previous reviewer comment

p. 6, l. 4-9 Please include an equation for the calculation described here. It is not clear how double counting in time is avoided given this description.
- It is unclear which aspect the reviewer is referring to, we agree that the explanation was confusing. To clarify, each 10-day back run is based on an emission over 24 hours and one run is conducted for each emission-period so there is no double counting, the text has been updated to reflect this:
"The resulting 10-day back air concentrations for each day's run were summed over the entire analysis period and a percentile value calculated to ascertain the likelihood of air originating from a particular grid cell (0.1 x 0.1) vis-à-vis other areas. The backruns shown were conducted from a receptor site in central Singapore, after comparison between a coastal receptor site and this inland site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island. This also helped inform the decision of domain size for the actual haze simulations."

Following the second review the text has been revised to:

"Air history maps provide a visual indication of where air at a given location has originated from. This helps to determine the regions that influence the composition of the air arriving at this location. To construct air history maps for Singapore, backward (inverse) runs were conducted with NAME, in addition to the forward simulations with the GFAS biomass burning emissions (Sect. 2.3). Fig. 2 illustrates the air history map for Singapore for the years 2010 to 2015. For each day in the six year period from 2010 to 2015, a 10-day backrun was conducted using meteorological input from the UM global model within a domain of 90.0◦E, 140.0◦E, 15.0◦S, 23.0◦N (Fig. 2). PM10 was emitted as a tracer from a receptor site in central Singapore and model particles were released over the first 24 hours with an emission rate of 1 g/s. The resulting concentration values in the 0-2km layer were output on a 0.1◦ × 0.1◦ resolution grid and integrated backwards in time for 10 days with a timestep of 10 minutes. By summing the results from several runs, air history data can be produced for different seasons and years, as well as the total for the whole period. A higher integrated concentration indicates that more air has passed through a grid cell on route to the receptor site, compared to a grid cell with a lower concentration. For each analysis period, the multiple corresponding 10-day air concentrations were summed for each grid cell and for the total domain. A percentile value was then calculated to ascertain the proportion of air influenced by a particular grid cell vis-à-vis other areas."

p. 16, l. 1-2 Was it expected that the "atypical and different meteorological conditions" would cause variation of the source regions when those were dominated by nearby fires? It seems unlikely, so the sentence is unexpected.
– Sentence has been reworded:

“Common for these two atypical haze events is little variation in the source regions across the monitoring stations, most likely due to the atypical and different meteorological conditions and the clear dominance of one source region.”

p. 19, l. 6 “Similarly to” should be “Similar to”.
- corrected:
“Similar to the results presented in Figure 3,”

p. 19, l. 15-7 Two statements contradict one another. The atypical haze events are said to have little variation between monitors, but then FMA 2014, one of the atypical haze events, is noted as having the largest difference in the next sentence. Please restate.
– We disagree with the reviewer here, as one sentence concerned the variation between major contributing source regions the other relates to absolute concentrations at the two monitoring stations. The section has been removed to avoid confusion.

p. 20, l. 3 Please eliminate the use of contractions here (“won’t”) and elsewhere.
- Done:
“peak concentrations will not always be captured in the model simulations”

p. 20, l. 5-6 No effort was made to show data that support this conclusion. Please show that it is true as suggested in the General Comments or remove the sentence.
– This sentence is now part of a broader paragraph in the conclusion that addresses sources of uncertainty in the results. The specific sentence has been modified to:
“Some of the varying difference between observed and modelled time series is also likely to be due to these many other sources of PM_{10} in Singapore.”

p. 20, l. 9 The grammar and sentence construction in the Conclusions section of the document require careful revision.
A significant part of the Conclusion has been reworded to improve the text, please see the revised version of the manuscript and the document highlighting the changes between the two versions of the manuscript.

Table 1 Please include the meaning of the abbreviations for the statistical correlations in the caption. – The caption now reads:
“Table 2. Statistics for PM_{10}, for both the western (NTU) and eastern (TP) monitoring stations and all years. Background concentration of 25 ug/m^3 is subtracted from the observations for all stations for all years. The metrics considered are the Pearson correlation coefficient (R), the modified normalised mean bias (MNMB), the fractional gross error (FGE), and Factor of 2 (FAC2).”

Figure 2 The caption states “air history” but the colorbar label indicates “Air Conc Percentile”, which seems to include information about emissions or concentrations rather than simply where the air has been. Please clarify.
– The meaning is the same, the air history is given in percent, as mentioned above, the text has been modified to clearly describe the air history map. If the reviewer still finds the figures unclear we will be happy to change the labelling.

Figure 4 Please specify the meaning of the colored values. Are the lines for the source regions stacked? If not, they should be in order to show how they contribute to the total observed concentration.
– The figure has been updated to show only observations and the total modelled concentration, as a stacked plot did not add clarity to the data.

Figures 6-8 The design of these figures is nice. It is not clear why only some of the contributing regions are colored. Please be consistent between the “other”, which are grey in the legend, and the grey countries in the map. Also, please order the pie chart wedges in the same order as the legend names. The colors are too similar to be able to distinguish when the pie chart is not in the same order as the regions in the legend.
– We had not spotted the issue with the grey regions, so thank you for raising it. We have modified the figures so that only major contributing source regions are coloured to highlight only relevant regions. The wedges have also been reordered in pie charts to match the legend.

Suggest reference Kim, P. S., Jacob, D. J., Mickley, L. J., Koplitz, S. N., Marlier, M. E., DeFries, R. S., Myers, S. S., Chew, B. and Mao, Y. H.: Sensitivity of population smoke exposure to fire locations in Equatorial Asia, Atmos Environ, 102, 11–17, doi:10.1016/j.atmosenv.2014.09.045 , 2015
- We thank the referee for pointing out this paper and have included it in the introduction.

Review 2:

Review of “Haze in Singapore - Source Attribution of Biomass Burning from Southeast Asia”
by Hansen et al.

This study investigates the sources of biomass burning from Southeast Asia for 6 years (2010-2015) using the Lagrangian dispersion model NAME. The tracer concentrations were evaluated using observations at two sites in Singapore. The authors also discussed the seasonal variations of emissions sources to Singapore. The topic of this paper is very interesting and important and I really appreciate the seasonal focus in this study. However, the conclusion and discussion are very confusing and repetitive. The paper lacks a coherent flow and the method section lacks significant information and clarity. I would only recommend this manuscript for publications in ACP only after substantial modifications to the manuscripts and figures. I also suggest the authors make higher quality figures for publication in ACP (Figs1, 4, …). In general, it is difficult to interpret the results and the discussion is weak.

We thank the reviewers for their constructive feedback and hope that our subsequent changes to the manuscript has improved the content and readability of the paper.
We have rewritten and restructured the Introduction and removed the Discussions section to make the Results and Conclusions clearer and more coherent. The figures have been updated following the reviewers’ suggestions.

*1- **Meteorology:***
Dispersion models are highly sensitive to their meteorological inputs. However, there is no analysis of the metrological values fed into the model. First, there is not clear statistical analysis or comparisons between modeled meteorology from UM and observations in that region. Without first evaluating the meteorological input, we cannot draw any conclusion from the Lagrangian models. For example, slight errors in modelled wind speed (and direction) and observations, makes the originating source region of the tracer very different.

- The UM is a world leading NWP model (see references in added text), and these are the data that were available to us at the time of the study. It was not feasible to conduct a thorough meteorological assessment of the UM for the whole region, but we have conducted an assessment of the UM data against observations that were available for Singapore. This is part of an internal report, that we summarise here for the reviewers, however we did not think it appropriate to add this to the final manuscript, but welcome further feedback:

“This report evaluates global UM model meteorological data, interpolated in NAME to obtain wind speed and direction, temperature and relative humidity data for a given location and time. These data are evaluated using meteorological observations available at 4 sites across Singapore.
The results show that modelled wind speeds are higher on average than those observed during 2013 particularly during the monsoon seasons. Wind speeds are one of the most important factors affecting pollutant levels, particularly close to strong sources. As such, when applying higher wind speeds in the model than observed may reduce modelled pollutant levels below those observed. There are some differences in wind direction between the model and observations but the prevailing winds appear to be captured well throughout the year.

Observed ambient temperatures are slightly higher and more variable on average than the model although there is good agreement between the model and observations. Relative humidity is higher in the model than the observations on average with the greatest variability inherent in the observations.
Rainfall does not appear well represented in NAME with higher means and more frequent low intensity events when compared to the observations which show less frequent high intensity rainfall typically associated with convective activity which dominates rainfall within the tropics. When considering the difference in total monthly rainfall between the model and observations, the former is predominantly higher during 2013 which may decrease modelled PM levels through wet deposition and contribute to the negative bias observed in both $PM_{2.5}$ and PM_{10}.

To augment the representation of the meteorology input in NAME, increasing both the temporal and spatial resolution of data for example using hourly averages is likely to improve both the modelled meteorology and pollutant levels."

Review #2: This has now been included in the revised manuscript with a table highlighting relevant changes:

"The Unified Model (UM) is the Met Office's operational numerical weather forecast model. The UM is a global model based on the non-hydrostatic fully compressible deep-atmosphere equations of motion solved using at semi-implicit semi-Lagrangian approach on a regular longitude-latitude grid (Walters et al., 2017). Archived analysis meteorology from the global version of the UM was used to drive NAME. As the UM is an operational model, the dynamical core and spatial resolution have changed throughout the period, from ~40 km over ~25 km to ~17 km resolution. However, for the majority of the study the resolution is constant at 25 km. These upgrades are described in Walters et al. (2011, 2017), and the relevant changes for dispersion modelling are summarised in Table 1. These changes are not expected to have a significant impact on the results, e.g., no significant differences in the deposition are seen across the change from instantaneous precipitation and cloud to 3-hour mean data in 2013.

"Global UM model meteorological data for 2013 have been evaluated using meteorological observations available at four sites across Singapore. The UM data are interpolated in NAME to obtain wind speed and direction, temperature, and relative 20 humidity data for each location and an hourly time resolution. The results show that modelled wind speeds are higher on average than those observed during 2013 particularly during the monsoon seasons. Wind speeds are one of the most important factors affecting pollutant levels, particularly close to strong sources. Although haze in Singapore is predominantly caused by long range transport of biomass smoke, the higher wind speeds in the model may contribute to reducing modelled pollutant levels below those observed. There are some differences in wind direction between the model and observations, but the prevailing wind directions are captured well throughout the year.

“Observed ambient temperatures are slightly higher and more variable on average than the model, although there is good agreement between the model and observations. Rainfall does not appear well represented with higher hourly means and more frequent low intensity events when compared to the observations, which show less frequent high-intensity rainfall associated with the convective activity that dominates rainfall within the tropics. Modelled total monthly rainfall is higher than observed during 2013, which may decrease modelled PM levels through wet deposition and contribute to the often negative bias observed in PM10, see Sect. 3. As discussed in Redington et al. (2016) and Hertwig et al. (2015), the uncertainties from the meteorological data feed into the dispersion simulation”

Second, P4:L1-2 mentions that the metrology runs were different (resolution and settings?) for different years. Based on previous studies, the modeled meteorological values (especially wind speed) are sensitive to the model resolution. Considering that in this manuscript, the authors compared different years with each other, I strongly recommend use of consistent settings and resolutions for the NWP runs. Else the inter-annual difference between the sources of biomass burning can easily be attributed to the difference between meteorological model differences.

- Kim et al 2015 use the same meteorology in an attribution study of biomass burning, we believe that it is better to use the highest resolution met data available. In spite of the changes in the resolution of the met data, differences between major contributing source regions (pie charts) for earlier years – 2011 and 2012 – shows results similar to 2014 and 2015 in the sense that there is significant difference between major contributing source regions at the two monitoring stations for 2011 and less so for 2012, hence, the differences in major contributing source regions at the two monitoring stations in 2014 and 2015 is not due to the changes in the NWP data resolution.

In general, there are large discrepancies between modeled and observed PM10 for all years and both stations. The authors assumed a constant 25 ug/m3 background concentration for all year. However, the emissions from various sectors (especially residential) have high seasonal variability (see Sobhani et al. 2018). Considering the same background concentration (meaning the same contribution of other sectors to your PM10) for all seasons may introduce large errors to your analysis.

– The paper the reviewer mentioned studies regions further north with stronger seasonal variation in domestic emissions than is the case for Singapore, so whilst we agree that variation in background PM occurs, there is no strong seasonal signal in Singapore.

Review #2: we have elaborated on our reasoning behind the constant background concentration and commented on the difference between attribution and apportionment:

“Subtracting a constant background from the observations does not give the exact contribution of PM10 from biomass burning alone because it does not remove all contributions from all other sources. However, it does give an indication of the periods with increased PM10 concentrations due to biomass burning. This is not an attempt to perform an *apportionment* of the observed PM10 concentrations in Singapore, as the observations, even with the subtracted background concentration, still includes contributions from sources other than biomass burning. However, the observations minus the constant background compared to the modelled time series provides an indication of the performance of the model, and through that the quality of the input used for the modelling. Using the modelled time series and the related source region information we are able to *attribute* the PM10 contribution in Singapore originating from biomass burning in Southeast Asia to the respective source regions. ”

Specific Comments:
Introduction:
1- This part lacking significant discussions and references. For example, the reference this part of P2, L2 is missing, “it is not caused by activities within Singapore, rather it is a transboundary problem caused by biomass burning across the wider region.” Maybe adding sample studies.
– References to Hertwig et al 2015, Reid et al 2013 have been added:
“Though haze occurs in Singapore (Hertwig et al., 2015; Lee et al., 2016b; Nichol, 1997, 1998; Sulong et al., 2017), it is not caused by activities within Singapore, rather it is a transboundary problem caused by biomass burning across the wider region (see Fig. 1 for a map of the region), which occurs during distinct ’burning seasons’ (Hertwig et al., 2015; Reid et al., 2013).”

2- P2, L13: I would recommend adding more discussions here. I suggest citing and/or describing some source attribution studies with the focus on other regions or bigger domains using different methods (Eulerian, Lagrangian, Observation analysis). I am not sure why the very few studies in the next lines are cited. Few examples for source appointment in different region of the world (with Eulerian methods) are: (Ikeda et al.,2017; Sobhani et al., 2018; Wang et al., 2011; Yang et al., 2017), With both Eulerian and Lagrangian methods:(Kulkarni et al., 2015) Observation Analysis + Lagrangian: (Winiger et al., 2017).
– Additional details and references have been added as suggested:
“Several previous studies have looked at attributing air pollution for different regions. Source attribution can be performed both through modelling and by looking at observations of air pollution in detail. For example, Heimann et al. (2015) carried out a source attribution study of UK air pollution using observations to distinguish between local and regional emissions, whereas Redington et al. (2016) estimated the sources of annual emissions of particulate matter from the UK and the EU by using the NAME model to look at threshold exceedences and episodes. Attribution studies have been performed using Eulerian models such as GEOS-chem, CMAQ, and WRF-STEM to study both Asia and the Arctic (Ikeda et al., 2017; Kim et al., 2015; Sobhani et al., 2018; Yang et al., 2017; Matsui et al., 2013) sometimes in combination with flight campaigns (Wang et al., 2011) to better constrain the emissions. Lagrangian models have also been used in combination with observations by Winiger et al. (2017). Combinations of Eulerian and Lagrangian models (Kulkarni et al., 2015) and Eulerian models and observations (Lee et al., 2017b) have been used to assess whether low visibility days were caused by fossil fuel combustion, biomass burning or a combination of the two. In Southeast Asia, Reddington et al. (2014) used an Eulerian model to study haze and estimated emissions through a bottom up approach. Source apportionment for studies of biomass burning related degradation of air quality and visibility in Southeast Asia has also been applied by Lee et al. (2017a) who used the WRF model to study the

sensitivity of the results to different met data and emission inventories and Engling et al. (2014), who used observations and a chemical mass balance receptor model to compare the chemical composition of total suspended particulate matter on haze and non-haze days during a haze event in 2006."

3- P2, L 22: Any reference for this sentence or is it the result of the study? If it is this result of the study please mention so.

– This can be seen from Figure 4:

In the six-year period, haze occurs almost annually during the season of August, September, and October (ASO), known as the haze season (see Fig. 4).

4- P2, L34 and P3, L3: Please add a reference for each sentence.

– Citations and a supporting figure with reference added:

"Generally, the inter-monsoon periods are characterised by light and variable winds, influenced by land and sea breezes with afternoon and early evening thunderstorms (Reid et al., 2012). The later inter-monsoon period is often wetter than the earlier inter-monsoon period (Chang et al., 2005; Reid et al., 2012). Furthermore, the inter-monsoon periods with weaker winds lead to air arriving in Singapore originating from the countries immediately west of and surrounding Singapore, see Fig A1. Previous studies have shown the importance of the ENSO in relation to reduction in convection and precipitation over the Martime Continent (MC) and corresponding increase in haze in Southeast Asia (Ashfold et al., 2017; Inness et al., 2015; Reid et al., 2012)."

5- In general, I suggest restructuring this section a bit for cohesiveness by moving few first sentences of the 5th paragraph (P2, L28) in introduction before the 4th paragraph (P2, L20). It is not clear if some the sentences in the 4th paragraph are result of this study or previous studies.

– the Introduction has been modified and sentences have been restructured for content and readability. Please refer to the updated manuscript.

6- It is not obvious why the focus of study is Singapore. Can you please add why the focus of this study Singapore?

– Singapore is the focus of this study due to the availability of observations with high spatial and temporal resolution and the interest in understanding more about the regions impacting the air quality here. We have amended the text to make this clearer.

"The Met Office (MO) and the Meteorological Service Singapore (MSS) have previously established a haze forecast system to predict haze in Singapore (Hertwig et al., 2015). This study advances the previous work to improve our understanding of haze and the underlying causes by analysing and attributing haze events of the recent past to their sources. The work focuses on Singapore due to the availability of air quality observations with high spatial and temporal resolution for recent years."
and
"The aim of this study is to investigate spatial variation of haze across Singapore through source attribution, including the variation in concentration and the contributing source regions across Singapore depending on the distance to source regions and the seasonal variation by looking at four recent haze events occurring during different seasons between January 2010 and December 2015. This is done

by linking meteorology, biomass burning, and dispersion modelling to study how the origin of haze has varied across Singapore during this whole period"

Methods:

1- This section lacks a lot of details. Can you please add some information and a paragraph describing the NAME model? How are they numerically represented in the model? What kind of aerosol processes are accounted for? Are there any know biases? Why have you used this model for this study?

– Description of the model has been added to text:

"The Numerical Atmospheric-dispersion Modelling Environment (NAME) III v6.5 (Jones et al., 2007) is a Lagrangian particle trajectory model, designed to forecast dispersion and deposition of particles and gasses on all ranges. Using the topography from the relevant met input, as NAME does not resolve buildings or terrain on scales smaller than the NWP. Emissions in the model are released as particles that contain information of one or more species, during the simulation these particles are exposed to various chemical and physical processes. NAME includes a comprehensive chemistry scheme which is not used in this study. Plume rise can also be considered, if applicable, in the model, here injection height is inferred from plume height information from the GFAS emissions. The only aerosol processes considered here are dispersion and wet and dry deposition of primary PM10. In NAME the dry deposition is parametrised using the resistance-based deposition velocity and wet deposition is based on the depletion equation. The advection is based on the winds obtained from the meteorology provided and a random component is added to represent the effects of atmospheric turbulence NAME is driven by meteorological data, which can be of various forms, in this case the Met Office's operational weather prediction model."

2- Can you please add some information and more description on the modeling setup for this study instead of just citing Hertwig et al. 2015. How are the wet and dry deposition processes calculated in the model?

– See added text above.

3- Also, can you please describe your meteorological model (UM) and why this model is used to drive NAME?

– Descriptions have been added to text:

"The Unified Model (UM) is the Met Office's operational numerical weather forecast model. The UM is a global model based on the non-hydrostatic fully compressible deep-atmosphere equations of motion solved using at semi-implicit semi-Lagrangian approach on a regular longitude-latitude grid (Walters et al., 2017). Archived meteorology from the global version of the Met Office Unified Model (UM) (Davies et al., 2005) was used to drive the NAME model"

Review #2: This has been elaborated on further in the second review, please see updated manuscript and document highlighting changes between the original submission and the revised manuscript.

4- Significant lack of clarity and explanations regarding observations: It is not clear at all where the locations of the observation sites are (maybe add them to all maps and include lat lon of the measurement sites?)? The authors should add more information on the method of

measurements in those locations. Also, tit is not clear where does these measurements come from (paper?, organization?)? Also, is this data available for public if so please include the link to the data either here or in the code and data availability section of the paper (or both).
– The location of the stations has been added as an insert to Fig 1, we thank the reviewer for pointing out that this information was missing. The observations data are from the Singapore NEA and are not publically available. The manuscript text has been extended to include details:

"Some 20 observation sites are located across Singapore, of these, one eastern and one western station have been chosen for best representation of trans-boundary PM_{10} concentrations across the main island of Singapore. In this analysis, the western station, Nanyang Technological University (NTU; 1.34505N, 103.6836E), is located relatively close to the industrial western part of Singapore and the eastern station, Temasek Polytechnic (TP; 1.34506N, 103.9304E), is placed next to the polytechnic but is also near open fields and a water reservoir, the location of the two sites in Singapore can be seen from Fig. 1. In Singapore the National Environment Agency measure hourly PM_{10} at several sites using the beta attenuation monitoring, where air is drawn through a size selective inlet down a vertically mounted heated sample tube to reduce particle bound water and to decrease the relative humidity of the sample stream to prevent condensation on the filter tape. The PM is drawn onto a glass fibre filter tape placed between a detector and a $_{14}C$ beta source. The beta beam passes upwards through the filter tape and the PM layer. The intensity of the beta beam is attenuated with the increasing mass load on the tape resulting in a reduced beta intensity measured by the detector. From a continuously integrated count rate the mass of the PM on the filter tape is calculated."

5- Air history map?? Do you mean PM10 or all aerosols (air?) lumped together? What chemical species and aerosols are considered in air history maps? Or is it only PM10 or tracer? This term is very confusing.

- Following our reply to Reviewer 1, we agree that the explanation was confusing. To clarify, each 10-day back run is based on 24 hours emissions of PM10 so there is no double counting, the text has been updated to reflect this:

"Air history maps provide an indication of where air at a given location has originated from. Fig. 2, illustrates an air history map for Singapore for the years 2010 to 2015. This helps determine the regions that influence the composition of the air arriving in Singapore. NAME backruns were conducted using the UM global Numerical Weather Prediction (NWP) model with PM_{10} as a tracer within a domain of 90.0_E, 140.0_E, 15.0_S, 23.0_N (Fig. 2). Wet and dry deposition are both turned on to simulate actual scenarios during the modelled time periods. Concentration values in the 0-2km layer were integrated at 10 minute time steps up till 10 days previous. The emission rate was set at a unit 1 g/s and emitted over 24 hours. A 10-day backrun was conducted for every single day in the six year time period from 2010 to 2015. The resulting 10-day back air concentrations for each day's run were summed over the entire analysis period and a percentile value calculated to ascertain the likelihood of air originating from a particular grid cell (0.1 x 0.1) vis-à-vis other areas. The backruns shown were conducted from a receptor site in central Singapore, after comparison between a coastal receptor site and this inland site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island. This also helped inform the decision of domain size for the actual haze simulations."

Review #2: The text describing the air history maps has been revised again, please refer to previous comments and the revised manuscript.

Results:

1- The assumption of 25 ug/m3 for both stations is problematic. Could it be because the background value from another source is higher in the western station??Also emissions from other important sectors are not accounted for which might cause the difference between the stations.

– Looking at observations for periods without haze in 2013 and 2015 we found average concentrations between 23 and 29 ug/m3 at both sites (see numbers below), therefore a background concentration of 25 seems entirely reasonable when looking at haze contributions that are of a similar order of magnitude. We do acknowledge that we are not capturing variations in local contributions to these sites and that there does appear to be a slight difference between the two sites.

2013 : except June

P09	25.99653
P28	23.66771

2013 ASO:

P09	26.92174
P28	22.79807

2015 : except ASO

P09	28.76008
P28	23.44001

Review #2: In the previous response to reviewers, background concentrations at the two stations were estimated based on averages of the PM_{10} concentrations during non-haze periods, this follows the method used by Kim et al, 2015:

"We estimate the smoke concentration at each site in the observations by subtracting as baseline the mean concentration for the bracketing non-burning months (June and December)."

https://doi.org/10.1016/j.atmosenv.2014.09.045

The text now reads:

"Observations of PM10 in Singapore from 2010 - 2015 show an overall background concentration during months of little or no burning of between 23 - 29 μg/m3 at the two monitoring stations. These values fit well with those determined in other studies for Singapore. For example, Hertwig et al. (2015) estimated background concentrations for PM10 to be around 30 μg/m3, based on the 2013 haze episode. In general, both background and peak concentrations vary between NTU and TP. Following the approach of Kim et al. (2015) we assume a constant background of 25 μg/m3 for the PM10 observations at both sites and subtract this value from the observation time series. "

2- P14, L13: Can you add some figure (maybe to SM) to show the meteorology difference for 2013 and other years. In general, this sentence is vague. What do you mean by 2013 is a unique year in terms of meteorology and burning?
– Figure A1 has been added to show air history for each season for all years (also see point 6 under Figures below), a reference to Oozeer et al 2016 has been added to P4 L10 to explain the meteorology of June 2013:
"In June 2013 a typhoon (Gaveau et al., 2014) coincided with major atmospheric emissions from peat fires in Southeast Asia (Oozeer et al., 2016)."

3- Are peaks concurrent with biomass burning incidents? Several other factors influence the peaks. For example, high residential emissions in winter in South East Asia can be attributed to the peaks.
– We are not entirely clear what the reviewer means by "winter" in this context. We are not convinced that high residential emissions in northern hemisphere winter have an impact as far south as Singapore.
By "winter", we assume that the reviewer is saying it is cold in winter, therefore people burn more for heating. Then that "winter" will be during JJASON when it is wet in Northern SEA. Besides rain being great for wet deposition, the monsoonal flow is mainly southwesterly which is unfavourable for transport from Northern SEA to Singapore (Reid et al., 2013). People in maritime SEA also don't burn more for heating as we are in the tropics. We experience a "wet" and "wetter" season for JJASON and DJFMAM respectively (Reid et al., 2013).

Reid et al., 2013. Observing and understanding the Southeast Asian aerosol system by remote sensing. Atm Res.

We see no peaks/increased background concentrations in the observations during any particular season for any year, nor ASO 2013. See previous general introductory comments on meteorology, comment 1 and 2 in this section, and corresponding replies in this response to reviewers. The increased concentrations and peaks during ASO coincides with the biomass burning season in the region, which supports the idea that the increases are due to haze caused by biomass burning.

4- There is a large redundancy between results (section 3) and discussions (section 4). I suggest merging section 4 into section 3 and conclusions.
- this has been done, please refer to the last section of the Results section and the Conclusions in the revised manuscript.

5- P 15, L30: It seems like the model did not capture the observation contrary to the claim.
- See reply to comment below and the updated Conclusions:
"For the four haze events focused on here, there is variability in the correlation between the modelled and observed time series, with the best correlations seen for haze events where the emission sources are close to Singapore. As discussed by Hertwig et al. (2015), uncertainty in these results originates from the emissions and the meteorology. For the former, the uncertainties result from the fact that the emissions used here are based on one daily snapshot of FRP and IH, and though some attempts are made to resolve issues with missing fire emissions caused by the lack of transparency of clouds the data will naturally be incomplete. At the same time, hourly emissions are calculated based on this one daily snapshot adding a

temporal resolution that the data does not provide, which also means that peak concentrations will not always be captured in the model simulations. The meteorology provides another significant source of uncertainty, as is usually the case in atmospheric modelling. When considering the resolution of the analysis meteorology used here and the size of Singapore it is clear that there will be unresolved features in both topography and in the meteorology and hence in the dispersion modelling. However, the differences we see between the two sites show that we are starting to capture this scale. Uncertainties in the NWP data such as elevated wind speeds and too frequent and too low intensity precipitation will disperse the pollutants further and wash out more than should be, resulting in lower modelled concentrations These uncertainties naturally have a larger impact over longer travel distances, which is reflected in our statistics. It should also be kept in mind that the observations are measuring all PM_{10} and we are only modelling primary PM_{10} emissions from biomass burning. Other sources of PM_{10} include sea salt, dust, secondary organic aerosol, emissions from industry, local and transboundary road traffic, as well as domestic heating, not all of which are constant throughout the year. Some of the varying difference between observed and modelled time series is also likely to be due to these many other sources of PM_{10} in Singapore. However, in spite of these uncertainties our results show that we are able to model dispersion of particulate matter from biomass burning in Southeast Asia and the resulting haze in Singapore with reasonable confidence."

6- P16, L10: The model significantly underestimates the peaks (30/125) Please explain why?
- We have taken out the peak values from the paper due to the uncertainties we are discussing in the new conclusions section. Upon reflection on the reviewers feedback, we feel that using only an hourly peak value in the text misrepresents the ability of the model to represent the broader haze event.

Figures:
1- *Does Figure 1 show the entire domain? It seems smaller that the domain mentioned in the method section. Please correct the figure include all the domain in this figure.*
– The figure has been updated to include the full domain

2- *Please add the location of Singapore to Figure1 and all other spatial figures. It is hard for someone who does not know the regions geography to find Singapore in each figure. Based on the captions description the reader might think Singapore is located in the Riau Islands.*
– An insert has been added to Figure 1 showing Singapore and the relative locations of the monitoring stations. Singapore has also been added to the subplots of Fig 5 – 8; we had not spotted that it was missing, so thank you for pointing this out.

3- Figure 2: Wrong caption. The second line of caption of this figure is not related to manuscript. Central receptor sites???? Inland and coastal sites? Are these sites discussed in this manuscript??
– The figure caption has been reduced to the relevant information and the description in the text has been extended:
"Figure 2. Air history map for 2010 - 2015, showing where air arriving in Singapore during this period originated from. Each shading shows the relative contribution of

air/PM_{10} to the central receptor site in Singapore in percent integrated over the atmospheric column from 0 to 2 km."

"The backruns shown were conducted from a receptor site in central Singapore. After comparison between a coastal receptor site and this inland site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island. This also helped inform the decision of domain size for the actual haze simulations."

4- Figure 2: Please correct the label title.
- See reply above

5- Figure 2: Can you please add more discussions about this figure to the paper? It is confusing what these figures show.
– The text has been modified to provide more information. Please refer to previous section of this reply to reviewers, e.g., the final paragraph of Methods/Section 2 and reply to Comment 5 in Methods above.
Review #2: The text in subsection 2.6 has been updated to better explain the process for creating air history maps.

6- *Figure 2: It is very nice that you included figure A2 (Figure 2 for all years to the discussion). I suggest also adding similar plots for each season. (each season averaged over the years). The season specific "Air history maps" would make it easier to understand the transport pathway in different seasons as discussed in P6, L10.*
– Thank you. We have included these maps in the supplementary material. Also see response to point 2 in Methods above

7- Figure 3: This is a good plot; however, it is difficult to compare different years because of the different scales. Also, the y axix label denote T as the unit for monthly emission which is different from the caption.
– The caption is consistent with the plot – the figure shows the monthly emissions in Tonnes which is also the tonnes emitted per month. We have decided not to change the y-axes, as too much information would be lost from using the same scale, but we have removed the units from the figure caption to avoid confusion.

8- Figure 4: This figure is very hard to read and should be modified before publications. First, it seems like hourly observations are plotted against (daily averages of model??). It is very hard to distinguish any modeled data points. Please make different plots for this figure. One way is showing monthly averages for both model and observation similar to Figure 3. Or time series of the daily observations as points overlaid on top of the modeled output. I recommend area chart for modeled value. Please include sum multiple region as "the other regions" multiple of the regions in this plot. Please only include important regions with visible high impacts. Very few of the 28 regions are visible in this plot. Maybe another scale (e.g. a log scale) is better for the purpose of this plot. Please use the same scale for all years and denote the events discuss on these plots.
The scale for all the years vary significantly. I would suggest having all PM 10 for all years on the same scale 0-700. Quick look at the plots, one might think there are higher pm 10 concentrations in 2010 compared to 2013 or 2015.

– The figure has been modified so that all years are on the same scale and only one line is plotted for the modelled time series, which is the sum of all sources (see also response to Reviewer 1 on this matter). We investigated reducing the data to daily averages as suggested, but this removes too much detail from the results so we have chosen to stick with the hourly data.

9- Figures 5-8: Please add a title with the name of event to the plot. Please add the stations (locations of NTU and TP) and denote Singapore on the plots.
– Title added and Singapore added in c) subfigures. The stations have been added to a subset image of Figure 1 as they would not be visible in these smaller figures.

10- Figure A2: What are the colored squares overlaid on the plots? Please put the figure in order that is mentioned in the paper.
– The coloured squares have been removed, and the figures in the supplementary material have been reordered.

Minor Comments and Technical Corrections:
7- P2, L3: Fig 1 is technically not related to this sentence.
– Agreed, however, a map of the region is beneficial to readers unfamiliar with Southeast Asia, an explanation has been added to this sentence:
"(see Fig. 1 for a map of the region)"

8- P2, L3: I would recommend adding reference for the second part of this sentence. (the reference for transboundary problem…)
– References have been added:
"Though haze occurs in Singapore (Hertwig et al., 2015; Lee et al., 2016b; Nichol, 1997, 1998; Sulong et al., 2017), it is not caused by activities within Singapore, rather it is a transboundary problem caused by biomass burning across the wider region (see Fig. 1 for a map of the region), which occurs during distinct 'burning seasons' (Hertwig et al., 2015; Reid et al., 2013)."

9- P2, L25: This sentence is very vague. Two events in each of June 2014-2015 and FMA 2014-2015. Or one event in June (2014 or 2015?) and one in FMA (2015?). Also, why using FMA vs June. I would recommend either using month or season. Can you use the season instead of June for consistency?
– The sentence has been removed following the rewriting of the Introduction, we hope the updated manuscript reads more easily. For clarity: the 2013 event occurred during the month of June only, whereas the 2014 event lasted throughout FMA

10- P2, L 33: Can you please elaborate what you mean by north-east monsoon and south-west monsoon seasons?
– it is outside of the scope of this paper to explain the monsoon in detail, figures have been added to the supplementary material to illustrate the seasons as defined in this study, and the manuscript has been amended to include:
"Meteorologically, the year in Singapore is split into four seasons, two monsoon seasons separated by two inter-monsoon seasons: the north-east monsoon season

is generally from December to early March and dominated by northeasterly winds; the first inter-monsoon period from late March through May; the south-west monsoon from June through September, with air in Singapore generally arriving from a southeasterly direction, and the second inter-monsoon period in October and November (Fing, 2012).”

11- P3, L 9: FMA acronym were explained last page and redundant here.
– text revised to just FMA

12- P3, L30: What does NAME stands for?
– Numerical Atmospheric-dispersion Modelling Environment, this and a model description have been added to text as described in comments above.

13- P3, L32: I recommend adding a figure to SM with the modeling domain. It seems like figure 1 does not show the complete modeling domain. (not extended 14 S or 23 N)
–Thank you for pointing this out, Figure 1 has been expanded to cover the full domain.

14- P4, L1-2: What do you mean? Is it different meteorological setup for each year??? Is the resolution of NWP runs different from 17 km to 40 km??
– As the UM is an operational model the resolution has changed over the study period, this does not seem to impact the results significantly – see reply in section on Meteorology above.
Review #2: A table has been added to the revised manuscript to provide information on the relevant model changes.

15- P4, L 14: Please add what GFED stands for.
– Global Fire Emissions Database, added to text:
“with the Global Fire Emissions Database (GFED) data set”

16- P4, L 16: Redundant, very similar sentence in the above paragraph L10….
– Sentence removed

17- P4, L24-25: Can you please point to the pie charts?
– This is a general comment referring to all figures throughout the paper. In general references are included to figures where appropriate, however reference has been added to the text to clarify:
“Annual and seasonal pie charts showing the percentage contribution from each source region at each monitoring station have been produced, to capture the spatial variation of biomass burning across the island, e.g., Figs 5c-8c.”

18- P4- L30-35: Can you please explain why did you use these metrics instead of other metrics like R2 or RMSE and many others? I suggest adding few more metrics to the tables 1 and 2 (R2 and RMSE). Please provide references or descriptions of the metrics used.
– The metrics are explained in Methods, P6 L22 – 27, and additional references have been added. These metrics have been chosen as they have been used in other related studies.

"The metrics considered are the Pearson correlation coefficient (R), i.e., the correlation between the model and observations used to get an indication of the match between patterns in the modelled and observed time series; the modified normalised mean bias (MNMB) which assesses the bias of the forecast and can have values between -2 and +2 (Seigneur et al., 2000); the fractional gross error (FGE) which gives the overall error of the model prediction and is limited between 0 and +2 (Ordóñez et al., 2010; Savage et al., 2013); and finally, Factor of 2 (FAC2) which gives an indication of the fraction of the model results that fall within a factor 2 of the observations (Hertwig et al., 2015)."

19- P4, L35: I strongly suggest using daily averages instead of hourly averages.
– This has been investigated, but not implemented as it does not add any clarity to the visualisation or data analysis.

20- P5, L7: Please add what NWP stands for.
– "Numerical Weather Prediction", added to text

21- P5-6: Air history vs air conc. percentile? Please clarify?
– This has been clarified in the figure caption and the text. Figure caption now reads:
"Air history map for 2010 - 2015, showing where air arriving in Singapore during this period originated from. Each shading shows the relative contribution of air/PM_{10} to the central receptor site in Singapore in percent integrated over the atmospheric column from 0 to 2 km."

22- P7, L20: I would highly suggest including emission maps for each year.
– The source regions and relative emissions are clear from Fig 3 and subfigures c) of Figs 5-8 highlight the major contributing source region(s) for each of the haze events. Figure limitations also prevent extra maps being added.

23- P7, L29: What is the reference for this sentence?
–the figures in the text are based on analysis of the observations, see Results comment 1 above.

24- P7, L32: Why did you assume constant 25 ug/m3 for background concentration? Is there any reference for that?
– This value was also used by Hertwig et al 2015, but we have also calculated that this is appropriate - see comments to Results comment 1 and comment above.
Review #2: This has been elaborated on following the second review, please refer to P11, line19 onwards in the updated manuscript.

25- P8, L3-5: I suggest including the values for clarity and readability.
- the values have been included:
"For years like 2013, which was dominated by one extreme haze event, the correlation between the modelled time series and the observations is very high (0.79

and 0.80 at NTU and TP, respectively, see Table 3). To some extent, this is also the case for the 2014 and 2015 events (0.27, 0.35 and 0.44, 0.43 for 2014 and 2015, respectively)."

26- P9, L4: Please add the name of the western monitoring station here and throughout the manuscript
– this has been included:
"When comparing concentrations between the two stations it can be seen that the concentrations are higher at the western monitoring station (NTU) most of the time. The opposite, concentrations at the eastern monitoring (TP) stations being higher than those at the western station (NTU),"

27- P14, L15-16: A sentence without a paragraph. remove the unnecessary line break.
- the linebreak has been removed

28- P14, L 16-18: This sentence is very confusing. Please rephrase it.
- the sentence has been rewritten to:
"Though 2013 was generally a year with weak winds and average burning, the month of June was very unique, both in terms of meteorology and burning (Fig. 5). The June 2013 haze event was caused by a typhoon coinciding with intense burning in Riau (Fig. 3)."

29- P14: In general, adding the locations of the sites to the maps would make reading the paper much easier.
– The locations of the monitoring sites have been added to Figure 1

30- P14, L24: It is not obvious if the maximum observed and modeled are concurrent or the values indicate maximum observation and maximum modeled value occurring at different times?
- The values are not concurrent so have been removed to avoid confusion

31- P15, L25: Would you discuss FMA 2014 or February 2014 only. Earlier in the text you mentioned June 2013 and February 2014 as the haze events but discuss FMA 2014 as the haze event here. In general, there is a lack of consistency between using months and seasons.
– this has been corrected to FMA, June is used on occasion as the 2013 event lasted less than a month which is not the case for the other events considered here

32- P15, L30: Can you explain the reason why concentrations at TP is double of NTU?
We have not looked into this in great detail, but the difference highlights the importance of local scale meteorology on results (both observations and model) and the importance of using higher-resolution data and Langrangian (and/or very high resolution Eulerian) models for interpretation at this spatial scale. As noted above the peak values this refers to have been removed from the text (see previous comment).

33- P16, L1-2: Very unclear and vague sentence. Different meteorology for events or between the monitoring stations? The sentence implies that in spite of the clear dominance of one source region, there is a little variation in the source regions across the monitoring stations???

– text updated for clarity:

"Common for these two atypical haze events is little variation in the source regions across the monitoring stations, most likely due to the atypical and different meteorological conditions and the clear dominance of one source region."

34- P16, L3: I suggest adding ASO to the title of this section. The inconsistency between using southwest monsoon haze and ASO makes it confusing.

– title now reads:

"3.2 ASO - southeast monsoon season haze"

35- P16, L4: Please remove the extra line break.

- linebreak removed

36- In general, not clear when the events are. I would highly suggest making a table including all the events discussed (and their corresponding figure) and also denoting each event on the time series plot.

– Thank you for this suggestion. A table has been added to beginning of results section, see Table 1.

Review #2: Following the addition of a table listing changes to the met data, this is now Table 2.

37- P 19, L1-3: This sentence is extremely confusing. For ASO or for the seasons with the most significant haze events including MJJ, FMA, and ASO?

– Sentence moved to results section and reworded to read:

"Of the seasons with the most significant haze events (e.g., MJJ 2013, FMA 2014, ASO 2014, and ASO 2015) in Singapore, the air history maps show that the region of influence for Singapore generally covers the largest area during ASO when air is coming from southeasterly directions"

38- Please check the sentence constructions of discussions and conclusions sections carefully.

– as mentioned above (Results comment 4) these sections have been restructured and reworded. Please see revised manuscript.

Review #2: Please refer to the attached pdf document highlighting changes between the original and the revised manuscripts.

39- P19- 20: The discussions seems like an extended conclusion section and there is a lot of redundancy between results (section3), discussions (section 4) and conclusions (section 5), which decrease the readability of paper.

– as above (Results comment 4) these sections have been restructured and reworded, please refer to the manuscript for updated text.

40- P20, L30: Please restate this sentence. It is very confusing.

- the sentences now read:

"Looking at emissions during ASO for the four years with the most variation across the island (2011, 2012, 2014, and 2015), the largest emissions were seen from Central Kalimantan, South Sumatra, Jambi, and also West Kalimantan. For events during FMA Cambodia, East Kalimantan, Myanmar, Thailand, and Vietnam showed larger emissions during FMA."

41- Code and data availability: Please include the link to the observations used for this study.

– This is not possible as the observations are not publicly available.

Specific Comments

The third sentence of the introduction is a run-on sentence. Additionally, the second sentence of the second paragraph in the introduction is a run-on sentence. I will not note other grammatical errors, but someone must correct these and others before this article is suitable for publication.

- These sentences read/have been edited to read:

1. Clearing forest for plantations by burning is a quick and easy way to open up and fertilise the soil, however, it is also a process that is difficult to control. The emissions from these fires can have massive and detrimental impacts far from where the original fires were lit.
2. Scientific studies such as Kim et al. (2015), as well as the popular press, often attribute peatland destruction and related haze in the region to Indonesia (Reid et al., 2013). However, the haze cannot be attributed to only one region or country alone.

Throughout the document sentences have been shortened, please refer to the document detailing differences between the original submission and the revised manuscript.

Acronyms including but not limited to NAME, GFAS, and CAMS are introduced after being used previously. Please ensure that every acronym is introduced upon first usage.

- This has been corrected.

"Sec" is not an appropriate abbreviation for "Section". Please replace all occurrences.

- Following the "Manuscript preparation guidelines for authors" (see excerpt below) "Sec." has been replaced with "Sect.":

Sections: The headings of all sections, including introduction, results, discussions or summary must be numbered. Three levels of sectioning are allowed, e.g. 3, 3.1, and 3.1.1. The abbreviation "Sect." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence.

"Air history" is an inaccurate term to refer to the convolution of emissions and back trajectory information. Please revise throughout.

- The back trajectories do not consider emissions, please see response to the general comments above.

Line Comment

p. 4, l. 16 “validated” should be “evaluated” here and elsewhere (e.g., p.7, l. 4). Please change all occurrences when speaking of a comparison of measurements and models. Both have errors, which indicates that neither is sufficient for validating the other.

- “validated” has been replaced by “evaluated” throughout the manuscript.

p. 7, l. 25-7 This statement conflicts with the last sentence of the abstract, which states that “variation in local meteorology can impact concentrations of particulate matter significantly”. If that were true, it would not be sufficient to use a central meteorological site. Please resolve by removing one of the statements. If the abstract statement is not changed, then the entire study needs to be presented for only one Singapore site. If a single receptor site was used for the back trajectories, it is not clear how the modeled concentrations in Figure 4 would be distinct as they appear to be or how these distinctions were investigated as indicated on p. 21, l. 1-3.

- This only applies to the air history maps where the concentrations are averaged over time (seasons or years) and biomass burning emissions are not considered.

We feel that the original (multi-coloured) time series plot made it easier for the reader to distinguish the contributions and relate the time series and pie charts.

Haze in Singapore - Source Attribution of Biomass Burning PM_{10} from Southeast Asia

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Abstract. This paper presents a study of haze in Singapore caused by biomass burning in Southeast Asia over the six year period from 2010 to 2015, using the Lagrangian dispersion model ~~, NAME.~~

the Numerical Atmospheric-dispersion Modelling Environment (NAME). The major contributing source regions ~~are shown to be Riau, Peninsular Malaysia, South Sumatra, and Central and West Kalimantan. However, we see differences in haze concentrations and variation in the relative contributions from the various source regions between monitoring stations across Singapore, as well as on an inter-annual timescale. These results challenge the current popular assumption that haze in Singapore is dominated by emissions/burning from only Indonesia. It is shown that Peninsular Malaysia is a large source for the Maritime Continent off-season biomass burning impact on Singapore.~~ to the haze are identified using forwards and backwards model simulations of particulate matter.

~~As should be expected, the relatively stronger southeast monsoonal winds that coincide~~ The coincidence of relatively strong Southeast monsoonal winds with increased biomass burning activities in the Maritime Continent create the main Singapore haze season from August to October (ASO), which brings particulate matter from ~~several and~~ varying source regions to Singapore. Five regions are identified as the dominating sources of pollution during recent haze seasons: Riau, Peninsular Malaysia, South Sumatra, and Central and West Kalimantan. In contrast, ~~atypical~~ off-season haze episodes in Singapore are characterised by ~~atypical~~ unusual weather conditions, ideal for biomass burning, and ~~emissions~~ contributions dominated by a single source region (different for each event). The two most recent ~~atypical~~ off-season haze events in ~~mid 2013 and early 2014~~ mid-2013 and early-2014 have different source regions, ~~whereas a different set of five regions dominate as~~ which differ from the major contributing source regions for ~~most of the recent ASO haze seasons.~~ the haze season. These results challenge the current popular assumption that haze in Singapore is dominated by emissions/burning from only Indonesia. For example, it is shown that Peninsular Malaysia is a large source for the Maritime Continent off-season biomass burning impact on Singapore.

~~Haze~~ The results demonstrate that haze in Singapore varies across year, season, and location ~~it~~ and is influenced by local and regional weather, climate, and regional burning. ~~The~~ Differences in haze concentrations and variation in the relative contributions from the various source regions are seen between monitoring stations across Singapore, on a seasonal as well as on an inter-annual timescale. This study shows that even across small scales, such as in Singapore, variation in local meteo-

rology can impact concentrations of particulate matter significantly, and emphasises the importance of the scale of modelling both spatially and temporally.

1 Introduction

~~Biomass burning is a global phenomenon, it is~~ Haze caused by biomass burning is a significant issue throughout Southeast Asia. Biomass burning occurs naturally across the world, but is being accelerated by human activities and interests. Clearing forest for plantations by burning is a quick and easy way to open up and fertilise the soil, however, it is also a process that is difficult to control. The emissions from these fires can have massive and detrimental impacts far from where the original fires were lit. Biomass burning is a global phenomenon. It is an ancient practice ~~of human occupation and~~ as well as a natural process which modifies ~~Earth surface characteristics~~ the Earth's surface (Pereira et al., 2016). The haze from biomass burning impacts human health (Crippa et al., 2016; Sigsgaard et al., 2015; Youssouf et al., 2014; Reddington et al., 2015), crops, climate, bio-diversity, tourism, and agricultural production (Jones, 2006), and also aviation and marine navigation through visibility degradation (Crippa et al., 2016; Lee et al., 2016b). Over recent decades the impacts of biomass burning have been felt in increasing degree in Southeast Asia and in Singapore (Oozeer et al., 2016).

Though haze occurs in Singapore (Hertwig et al., 2015; Lee et al., 2016b; Nichol, 1997, 1998; Sulong et al., 2017), it is not caused by activities within Singapore~~, rather~~. Rather it is a transboundary problem caused by biomass burning across the wider region (see Fig. 1 ~~), which~~ for a map of the region), which typically occurs during distinct ~~'burning seasons'. Though~~ burning seasons (Hertwig et al., 2015; Reid et al., 2013). Scientific studies such as Kim et al. (2015), as well as the popular press, often attribute peatland destruction and related haze in the region to Indonesia ~~(Reid et al., 2013)~~(Reid et al., 2013). However, the haze cannot be attributed to ~~one region~~ only one region or country alone. To mitigate this, the Association of Southeast Asian Nations (ASEAN) Haze Agreement has been formed between the Southeast Asian nations to reduce haze and mitigate the related impacts using a scientific approach (Nazeer and Furuoka, 2017; Lee et al., 2016a).

~~Considerable inter-annual variation in biomass burning and related emissions of particulate matter (PM) in Southeast Asia is due to a combination of variation in El Niño Southern Oscillation (ENSO) (Fing, 2012) and anthropogenic land-use changes (Field et al., 2009; Shi and Yamaguchi, 2014).~~

Through the ASEAN, science-based mitigation has been attempted, but many lives are still lost every year due to haze caused by biomass burning (Lee et al., 2018). The Met Office (MO) and the Meteorological Service Singapore (MSS) have previously established a haze forecast system to predict haze in Singapore (Hertwig et al., 2015). This study advances the previous work to improve our understanding of haze and the underlying causes by analysing and attributing haze events of the recent past to their sources. ~~Several previous studies have looked at attributing air pollution across the world. Source attribution can be performed both through modelling and by looking at observations of air pollution in detail. For example, Heimann et al. (2015) carried out a source attribution study of UK air pollution using observations to distinguish between local and regional emissions, whereas, Redington et al. (2016) estimated the source of annual emissions of particulate matter from the UK and EU, by using the NAME model to look at threshold exceedences and episodes. In Southeast Asia, Reddington et al. (2014) used an Eulerian~~

~~model to study haze and estimated emissions through a bottom up approach and source apportionment has been applied by Lee et al. (2017a) and Engling et al. (2014) for studies of biomass burning related degradation of air quality and visibility.~~

~~Haze concentrations in Singapore vary throughout the six year period from 2010 to 2015 and even though biomass burning contributes to (low) PM_{10} concentrations in Singapore throughout large parts of the year, some peaks in the PM_{10} observations can be explained by haze almost exclusively. In the six year period, haze occurs almost annually during the season of August, September, and October (ASO), known as the haze season. Haze events occurring during other periods of the year are referred to as atypical or off-season haze. In 2013 and 2014 two unique atypical haze events occurred in June and in February, March, April (FMA), respectively (Hertwig et al., 2015; Gaveau et al., 2014; Duc et al., 2016). These events caused extremely high PM_{10} concentrations in Singapore, and raise the question of whether high concentrations of, or long term exposure to, PM which has the most significant health impacts~~The work focuses on Singapore due to the availability of air quality observations with high spatial and temporal resolution for recent years.

The weather and climate in Singapore and hence the transport of smoke from biomass burning is dominated by monsoon periods and influenced by the variations of the El Niño Southern Oscillation (ENSO), ~~such as the El Niño Modoki which influences~~ which modifies temperatures in the central equatorial pacific (Ashok et al., 2007; Yeh et al., 2009; Reid et al., 2012; Yuan and Yang, 2012). Meteorologically, the year in Singapore is split into four seasons, with two monsoon seasons separated by two inter-monsoon seasons~~: the~~ . The north-east monsoon season is generally from December to early March ~~, the~~ and dominated by northeasterly winds. The first inter-monsoon period follows from late March through May, then the south-west monsoon is from June through September, ~~and the~~ with air in Singapore generally arriving from a southeastern direction. The second inter-monsoon period ~~in~~ is October and November (Fing, 2012). Between years, there is large variability in the onset of the monsoon over Mainland Southeast Asia (Zhang et al., 2002). Generally, the inter-monsoon periods are characterised by light and variable winds, influenced by land and sea breezes with afternoon and early evening thunderstorms (Reid et al., 2012). The later inter-monsoon period is often wetter than the earlier inter-monsoon period ~~. Furthermore,~~ (Chang et al., 2005; Reid et al., 2012). The weaker winds during the inter-monsoon periods ~~with weaker winds~~ lead to air arriving in Singapore originating from the countries immediately west of and surrounding Singapore ~~.~~ (Fig A1).

Previous studies have shown the importance of ~~the~~ ENSO in relation to reduction in convection and precipitation over the Martime Continent (MC) and corresponding increase in haze in Southeast Asia (Ashfold et al., 2017; Inness et al., 2015; Reid et al., 2012). The ENSO conditions have varied significantly during the six year period of our study (2010 - 2015). During 2010, the conditions transitioned from a moderate El Niño to a moderate La Niña lasting through 2011. From 2012 to 2014 the ENSO conditions were neutral transitioning to very strong El Niño conditions in 2015, which lasted into 2016 (NOAA, 2017).

~~In terms of biomass burning, the year in this~~

The combination of variation in ENSO (Fing, 2012) and anthropogenic land-use changes (Field et al., 2009; Shi and Yamaguchi, 2014) leads to considerable inter-annual variation in biomass burning and related emissions of particulate matter (PM) in Southeast Asia. Biomass burning in the region can be divided into seasons that relate to the monsoon ~~seasons~~periods: February, March, and April (FMA) ~~is~~ are dominated by burning in Mainland Southeast Asia~~,~~ ; during May, June, and July (MJJ) burning starts in northern Sumatra and traverses southward~~, ASO~~ ; August, September, and October (ASO) is characterised by burning in

~~Southern~~ Kalimantan and, in general, there is little ~~or no burning influencing Singapore~~ burning in November, December, and January (NDJ) (Campbell et al., 2013; Chew et al., 2013; Reid et al., 2012, 2013).

From annual weather reports by MSS ~~(NEA, 2015)~~(NEA, 2015; NEA, 2017), unusual weather events from 2010 to 2015 and related haze events are linked. In 2010 a prolonged Madden-Julien Oscillation (MJO) dry phase caused a dry October, creating ideal conditions for biomass burning in the region and related haze in Singapore. 2011 began as an ENSO neutral year transitioning to La Niña, with dry conditions in early September and prevailing low level winds bringing PM_{10} to Singapore from biomass burning in central and southern Sumatra. During the Southwest monsoon of 2012, an MJO dry phase created dry and ideal haze conditions in September. In June 2013 a typhoon (Gaveau et al., 2014) coincided with major atmospheric emissions from peat fires in Southeast Asia ~~.~~ (Oozeer et al., 2016). In 2014 Singapore experienced haze during another intense MJO dry phase and drought, described by Mcbride et al. (2015). 2015 was the joint warmest year (with 1997 and 1998) and second driest year on record. ASO 2015 saw the worst haze in recent history in Singapore (Huijnen et al., 2016; Crippa et al., 2016; Koplitz et al., 2016), caused by ~~southwest~~southwesterly/southeasterly winds and fires in Southern and Central Sumatra and Southern Kalimantan. Fire carbon emissions over maritime South-East Asia in 2015 were the largest since 1997 (Huijnen et al., 2016). ~~This paper links~~

Haze concentrations in Singapore vary throughout the six year period from 2010 to 2015. Even though biomass burning contributes to (low) PM_{10} concentrations in Singapore throughout large parts of the year, some peaks in the PM_{10} observations can be explained by haze almost exclusively. In the six year period, haze occurs almost annually during the season of ASO, known as the haze season (see Fig. 4). Haze events occurring during other periods of the year are referred to as off-season or atypical haze. In 2013 and 2014 two unique atypical haze events occurred in June and in FMA, respectively (Hertwig et al., 2015; Gaveau et al., 2014; Duc et al., 2016). These events caused extremely high PM_{10} concentrations in Singapore.

Several previous studies have looked at attributing air pollution for different regions. Source attribution can be performed both through modelling and by looking at observations of air pollution in detail. For example, Heimann et al. (2015) carried out a source attribution study of air pollution in the United Kingdom (UK) using observations to distinguish between local and regional emissions, whereas Redington et al. (2016) estimated the sources of annual emissions of particulate matter from the UK and the European Union (EU) by using the Numerical Atmospheric-dispersion Modelling Environment (NAME) model to look at threshold exceedences and episodes. Attribution studies have been performed using Eulerian models such as the Goddard Earth Observing System atmospheric chemistry model (GEOS-chem), the Community Multiscale Air Quality Modeling System (CMAQ), and the Weather Research Forecasting System - Sulphur Transport and dEposition Model (WRF-STEM) to study both Asia and the Arctic (Ikeda et al., 2017; Kim et al., 2015; Sobhani et al., 2018; Yang et al., 2017; Matsui et al., 2013) sometimes in combination with flight campaigns (Wang et al., 2011) to better constrain the emissions. Lagrangian models have also been used in combination with observations by Winiger et al. (2017). Combinations of Eulerian and Lagrangian models (Kulkarni et al., 2015) and Eulerian models and observations (Lee et al., 2017b) have been used to assess whether low visibility days were caused by fossil fuel combustion, biomass burning, or a combination of the two. In Southeast Asia, Reddington et al. (2014) used an Eulerian model to study haze and estimated emissions through a bottom up approach. Source attribution for studies of biomass burning related degradation of air quality and visibility in Southeast Asia has also been applied by Lee et al. (2017a) who used the WRF model to study the sensitivity of the results to different met data and emission inventories. Engling et al. (2014) also used observations and a chemical mass balance receptor model to compare the chemical composition of total suspended particulate matter on haze and non-haze days during a haze event in 2006.

The aim of this study is to investigate the spatial variation of haze across Singapore through source attribution. This includes the variation in concentration and the contributing source regions at different sites across Singapore. This has been achieved by linking meteorology, biomass burning ~~, and~~ emissions, and forwards and backwards dispersion modelling to study how the origin of haze ~~have~~ has varied across Singapore during ~~this whole period.~~

~~The~~ 2010 - 2015. Fire radiative power and injection height from the Copernicus Atmosphere Monitoring Service (CAMS) global fire assimilation system (GFAS, (Kaiser et al., 2012)) and higher resolution land-use data from the Centre for Remote Imaging, Sensing and Processing at the National University of Singapore have been used to calculate Particulate Matter with diameter of 10 μm or less (PM_{10}) emissions from biomass burning in 29 defined source regions in Southeast Asia (Fig. 1). Using the Met Office's numerical weather prediction (NWP) model to drive the Numerical Atmospheric-dispersion Modelling Environment (NAME), a Lagrangian particle trajectory model, we are able to attribute the haze arriving in Singapore to its source region and study the difference between major contributing source regions at a western and an eastern monitoring station in Singapore. The model output is evaluated against PM_{10} observations from the two monitoring stations. The paper is composed as follows: ~~Sec. 2 ,~~ Sect. 2 describes the methods used in the study~~and Sec~~; Sect. 3 presents ~~an overview of emissions, air history, and validation~~the results and evaluation, along with a more detailed study of ~~atypical haze eventsin Sec. 3.1 and 3.2.~~ four recent haze events. The results and related implications are discussed in ~~Sec. ?? and the paper is concluded by Sec~~ Sect. 4.

2 Methods

This section describes ~~how the model was~~ the model used, the set up and ~~the~~ input used for the simulations~~. Individual simulations using the Lagrangian dispersion model , NAMEIII~~ , as well as the methods used to evaluate the results.

2.1 The Numerical Atmospheric-dispersion Modelling Environment

We use a Lagrangian model because of its ability to track emissions and provide detailed information on source regions at any given location in the modelling domain. The Numerical Atmospheric-dispersion Modelling Environment (NAME) III v6.5 (Jones et al., 2007) ~~were performed for each of the years from 2010 to 2015 for PM_{10} in a setup similar to that of the haze forecast described in Hertwig et al. (2015) including~~ is a Lagrangian particle trajectory model, designed to forecast dispersion and deposition of particles and gases on all ranges. NAME uses the topography from the relevant meteorological input and does not resolve buildings or terrain on scales smaller than the NWP. Emissions in the model are released as particles that contain information on one or more species. During the simulation these particles are exposed to various chemical and physical processes. NAME includes a comprehensive chemistry scheme, but this is not used in this study, as we are interested only in primary PM. The only aerosol processes considered here are dispersion and wet and dry deposition ~~. The domain considered~~

~~covers 14°S - 23°N and 90°E - 131°E. Archived meteorology from the global version of~~ of primary PM_{10}. In NAME the dry deposition is parametrised using the resistance-based deposition velocity and wet deposition is based on the depletion equation (Webster and Thomson, 2014). The advection is based on the winds obtained from the meteorology provided and a random component is added to represent the effects of atmospheric turbulence. NAME is driven by meteorological data, in this case the Met Office's operational weather prediction model (Davies et al., 2005), described below.

2.2 The Unified Model

The Unified Model (UM) ~~(Davies et al., 2005)~~ is the Met Office's operational numerical weather forecast model. The UM is a global model based on the non-hydrostatic fully compressible deep-atmosphere equations of motion solved using at semi-implicit semi-Lagrangian approach on a regular longitude-latitude grid (Walters et al., 2017). Archived analysis meteorology from the global version of the UM was used to drive ~~the NAME. Throughout the period~~ NAME. As the UM is an operational model, the dynamical core and spatial resolution ~~of the UM have changed , however, always resolving Singapore as a part of the Malaysian Peninsula~~have changed throughout the period, from ~40 km over ~25 km to ~17 km resolution. ~~, some of those~~ However, for the majority of the study the resolution is constant at 25 km. These upgrades are described in Walters et al. (2011, 2017)~~.~~, and the relevant changes for dispersion modelling are summarised in Table 1. These changes are not expected to have a significant impact on the results, e.g., no significant differences in the deposition are seen across the change from instantaneous precipitation and cloud to 3-hour mean data in 2013.

Global UM model meteorological data for 2013 have been evaluated using meteorological observations available at four sites across Singapore. The UM data are interpolated in NAME to obtain wind speed and direction, temperature, and relative humidity data for each location and an hourly time resolution. The results show that modelled wind speeds are higher on average than those observed during 2013 particularly during the monsoon seasons. Wind speeds are one of the most important factors affecting pollutant levels, particularly close to strong sources. Although haze in Singapore is predominantly caused by long range transport of biomass smoke, the higher wind speeds in the model may contribute to reducing modelled pollutant levels below those observed. There are some differences in wind direction between the model and observations, but the prevailing wind directions are captured well throughout the year.

Observed ambient temperatures are slightly higher and more variable on average than the model, although there is good agreement between the model and observations. Rainfall does not appear well represented with higher hourly means and more frequent low intensity events when compared to the observations, which show less frequent high-intensity rainfall associated with the convective activity that dominates rainfall within the tropics. Modelled total monthly rainfall is higher than observed during 2013, which may decrease modelled PM levels through wet deposition and contribute to the often negative bias observed in PM_{10}, see Sect. 3. As discussed in Redington et al. (2016) and Hertwig et al. (2015), the uncertainties from the meteorological data feed into the dispersion simulation. ~~The emissions used~~

2.3 NAME forward model simulations

Table 1. Summary of the changes in the global UM data over the period of this study, relevant to dispersion modelling.

Start date	Approx. horizontal resolution	Relevant change
1/1/2010	40 km	
20/1/2010	25 km	Horizontal resolution increase
30/4/2013	25 km	Change from use of instantaneous precipitation and cloud to 3-hour mean data
15/7/2014	17 km	Horizontal resolution increase

For the attribution forward NAME runs were conducted using the haze forecast set-up designed by Hertwig et al. (2015) and extending it to year-long haze simulations. Individual forward simulations were performed for each of the years from 2010 to 2015 for PM_{10} for a domain covering 14°S - 23°N and 90°E - 131°E using the GFAS PM_{10} biomass burning emissions described in Sect 2.3.1. Each run was initialised on the 1st of January and the simulation ran until the 31st of December of the same year. A maximum of 200 million model particles were emitted during the simulation and any particles leaving the domain were lost. The simulations used no boundary conditions and so there was no inflow of particles from the domain edges. From these simulations, modelled time series for the two monitoring sites described in Sect. 2.3.2 were produced.

2.3.1 Emissions and Source Regions

The PM_{10} emissions used in this study were calculated from the Global Fire Assimilation System (GFAS, Kaiser et al. (2012)) v1.2 daily gridded fire radiative power (FRP) and injection height (IH) products, integrated with high resolution land-use data and emission factors in an approach aimed at combining the benefits of the MSS and GFAS v1.2 source approaches described in Hertwig et al. (2015). Additionally, the land cover map used has been updated to the 2015 version by Miettinen et al. (2016b), which now covers the entire Southeast Asia region, as compared to the earlier 2010 version (Miettinen et al., 2016a). The horizontal dimensions of the emissions were dx=dy=0.1°, and ~~were~~ the material was released at varying heights based on the GFAS injection height information. Using the Lagrangian nature of the model, all emissions are tagged with source information to allow for assessment of contributing source regions and relative contributions. The choice of the GFAS data set as the basis for the source calculation was based on the need for daily emissions, as in the operational setup of Hertwig et al. (2015), and the good agreement of this with observations and consistency with the ~~GFED~~ Global Fire Emission Database (GFED) data set documented previously, e.g., Kaiser et al. (2012) and Rémy et al. (2017).

For this study, 29 source regions have been defined to ~~better~~ distinguish where the PM_{10} from biomass burning originated from (see Fig 1). ~~Given the~~ The Lagrangian nature of the ~~NAME model , it is possible to label and follow each emitted pollutant with its source location. This in turn~~ model enables us to attribute the PM_{10} concentrations at specific locations in Singapore to the individual source regions.

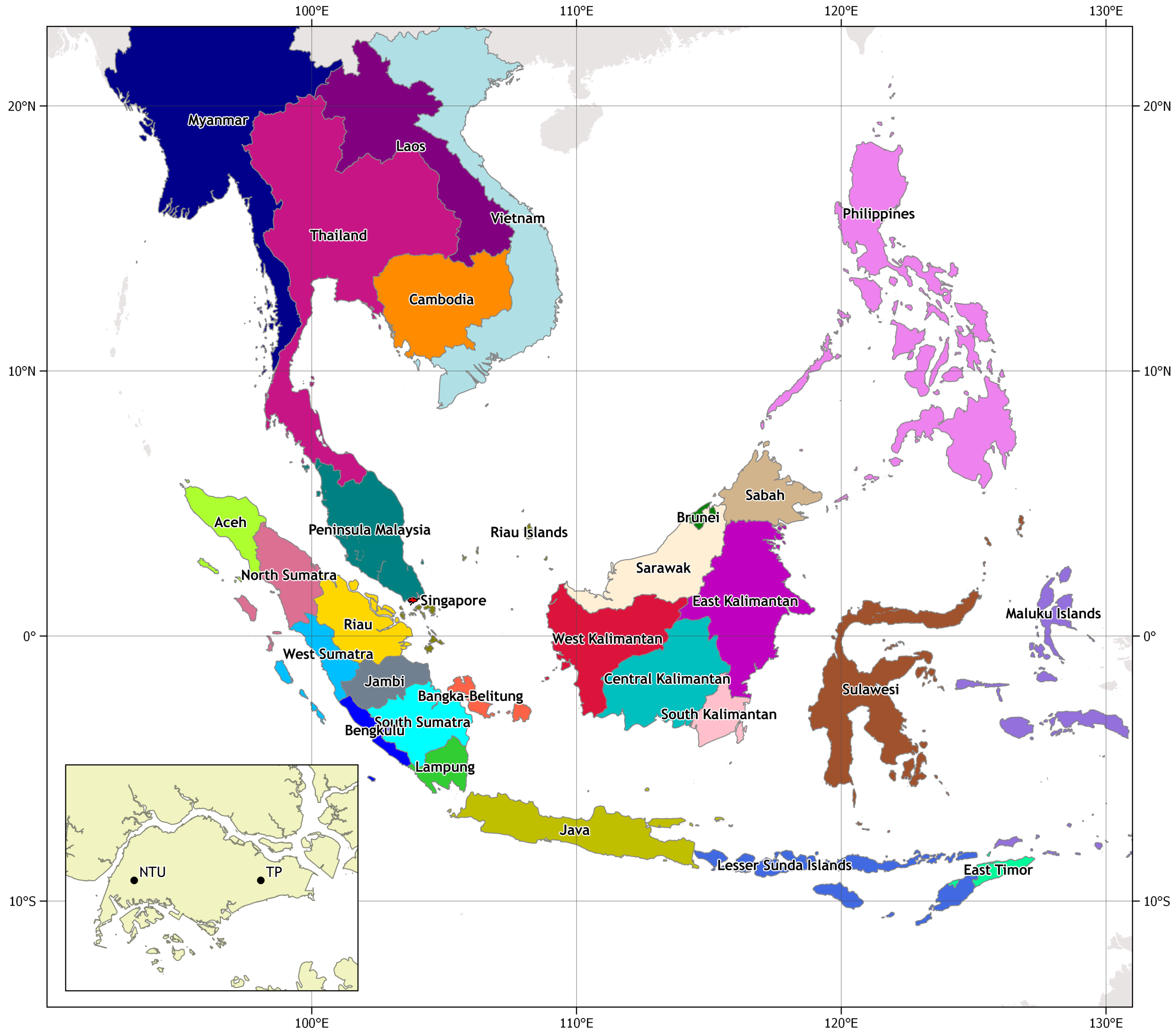

Figure 1. Locations and colour codes used for each of the 29 biomass burning source region within the domain from 10°S - 20°N and 90°E - 130°E considered in this study. Singapore is located south of Peninsular Malaysia and East of Riau. The insert in the bottom lefthand corner shows the relative location of the two monitoring stations in Singapore.

2.3.2 Observations and Performance Metrics

Some 20 air quality observation sites are located across Singapore~~, of~~. Of these, one eastern and one western station have been chosen ~~for best representation of~~ to explore trans-boundary PM_{10} concentrations across the main island of Singapore. In this analysis, the western station, Nanyang Technological University (NTU; 1.34505°N, 103.6836°E), is located relatively close to the industrial western part of Singapore~~and the~~. The eastern station, Temasek Polytechnic (TP; 1.34506°N, 103.9304°E), is placed next to the polytechnic but is also near open fields and a water reservoir. The location of the two sites in Singapore can be seen in the insert of Fig. 1. The National Environment Agency of Singapore measures hourly PM_{10} at these and other sites using beta attenuation monitoring. In this technique air is drawn through a size selective inlet down a vertically mounted heated sample tube to reduce particle bound water and to decrease the relative humidity of the sample stream to prevent condensation on the filter tape. The PM_{10} is drawn onto a glass fibre filter tape placed between a detector and a ^{14}C beta source. The beta beam passes upwards through the filter tape and the PM_{10} layer. The intensity of the beta beam is attenuated with the increasing mass load on the tape resulting in a reduced beta intensity measured by the detector. From a continuously integrated count rate the mass of the PM_{10} on the filter tape is calculated.

The following analysis is based on hourly PM_{10} observations and modelled time series at the two selected monitoring stations. Annual and seasonal pie charts showing the percentage contribution from each source region at each monitoring station have been produced ~~,~~ to capture the spatial variation of biomass burning across the island~~.~~, e.g., Figs 5c - 8c. During the period considered, several haze events occurred in Singapore.

~~To validate~~ To evaluate the model results, four performance metrics have been calculated. These evaluate ~~each species~~ the model performance at the two monitoring stations, for each year and ~~select~~ selected seasons in each of the six years with available observations. ~~The observations used are hourly PM_{10} measurements from the National Environment Agency, Singapore. The~~ The metrics considered are the Pearson correlation coefficient (R), i.e., the correlation between the model and observations used to get an indication of the match between patterns in the modelled and observed time series; the modified normalised mean bias (MNMB) which assesses the bias of the forecast and can have values between -2 and +2 (Seigneur et al., 2000); the fractional gross error (FGE) which gives the overall error of the model prediction and is limited between 0 and +2 (Ordóñez et al., 2010; Savage et al., 2013); and finally, Factor of 2 (FAC2) which gives an indication of the fraction of the model results that fall within a factor 2 of the observations (Hertwig et al., 2015). Because the emissions used are at a daily resolution ~~as~~ compared to the hourly observations of PM_{10}, a ~~possible~~ gap or mismatch in the timing of peak concentrations between modelled results and observation time series is possible. ~~Bias~~ Biases between modelled time series and the observations are expected as some fires will be missed due to the fact that they are too small for the satellites to register and the extent and/or duration of the other fires are over or under estimated due to cloud cover (Kaiser et al., 2012; Reid et al., 2013; Campbell et al., 2016).

2.4 Air History Maps

Air history maps ~~are able to provide an~~ provide a visual indication of where air at a given location has originated from. ~~Fig. 2, illustrates an air history map for Singapore for 2010 - 2015. This helps~~ This helps to determine the regions that influence the composition of the air ~~in Singapore. NAME backruns were conducted using~~ arriving at this location. To construct air history maps for Singapore, backward (inverse) runs were conducted with NAME, in addition to the forward simulations with the GFAS biomass burning emissions (Sect. 2.3). Fig. 2 illustrates the air history map for Singapore for the years 2010 to 2015. For each day in the six year period from 2010 to 2015, a 10-day backrun was conducted using meteorological input from the

Table 2. Overview of the four haze events studied in detail below. FMA: February, March, April; MJJ: May, June, July; ASO: August, September, October.

Year	2013	2014	2014	2015
Season	MJJ	FMA	ASO	ASO
Section	3.1	3.1	3.2	3.2
Figure	5	6	7	8

UM global ~~NWP model , with PM_{10} as a tracer~~ model within a domain of 90.0°E, 140.0°E, 15.0°S, 23.0°N (Fig. 2). ~~Wet and dry deposition are both turned on to simulate actual scenarios during the modelled time periods. Concentration~~ PM_{10} was emitted as a tracer from a receptor site in central Singapore and model particles were released over the first 24 hours with an emission rate of 1 g/s. The resulting concentration values in the 0-2km layer were ~~integrated at~~ output on a 0.1° × 0.1° resolution grid and integrated backwards in time for 10 ~~minute time steps up till 10~~ days ~~previous. The emission rate was set at a unit 1 g/s and emitted over 24 hours. A 10-day backrun was conducted for every single day in the six year time period from 2010 to 2015. The resulting10-day back air concentrationfor each day's run was summed for~~ with a timestep of 10 minutes. A higher integrated concentration indicates that more air has passed through a grid cell on route to the receptor site, compared to a grid cell with a lower concentration. By summing the results from multiple runs, air history data can be produced for different seasons and years, as well as the total for the whole period. For each analysis period~~and a percentile value~~, the multiple corresponding 10-day air concentrations were summed for each grid cell and for the total domain. A percentile value was then calculated to ascertain the ~~likelihood of air originating from a particular geographical area~~ proportion of air influenced by a particular grid cell vis-à-vis other areas.

~~The percentile is derived by taking a fractional contribution of each grid point (0.1° × 0.1°) concentration value as compared to the total concentration present in the entire model domain. The fractional concentration contribution of all the gridpoints were then arranged in ascending order and cumulatively summed and each 10 % band is shown in Fig. 2.~~

Comparison between the inland site and a coastal receptor site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island when averaged over time. The results of the air history simulations helped inform the decision of domain size for the forward haze simulations.

3 Results

This section presents the results based on the modeling setup described in Sect. 2 above. Air history maps show where the air arriving in Singapore has travelled through and looking at the emissions provides information on when and where the largest emissions in the region occur. Using hourly PM_{10} observations we evaluate our model output before using the results to address the research questions posed in Sect. 1. Four events are studied in more detail in the final subsections of this section, these are outlined in Table 2.

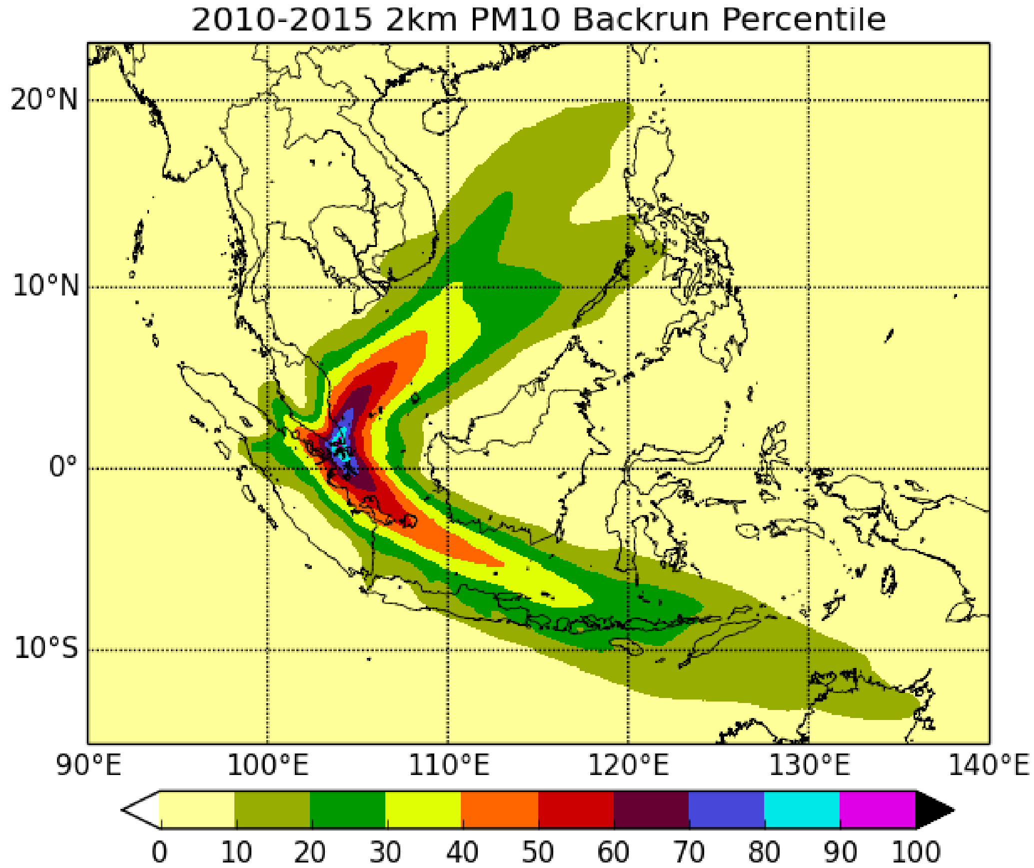

Figure 2. Air history map for 2010 - 2015, showing where air arriving in Singapore during this period originated from. Each shading shows the relative contribution of air to the central receptor site in Singapore in percent integrated over the atmospheric column from 0 to 2 km.

The air history map in Fig. 2 shows that most air arriving in Singapore has travelled from either ~~the northeast or southeast~~ northeastern or southeastern directions, illustrating the two monsoon seasons experienced in Singapore ~~.~~ (see Fig. A1 for air history maps summed over the period for each of the individual seasons). The northeastern component of the bifurcation in the wind pattern is representative of the northeast monsoon in FMA (Fig. A1a), and the southeastern "fork" shows the southeast monsoon period during ASO ~~.~~ (Fig. A1c). During the six years represented by the figure, significant variation occurs during the individual years (~~see~~ Fig. A2). In 2010 winds were quite weak and the air arriving in Singapore mainly came from a north-easterly direction and did not show the expected "fork" from the two monsoon seasons ~~.~~ (Fig. A2a). This means that the air impacting Singapore that year mainly traversed through countries and regions very near to or east of Singapore, e.g., the Philippines, Peninsular Malaysia, Riau, and Riau Islands. The air history map for 2011 (Fig. A2b) shows a clear bifurcation, with air arriving from northeast and southeast, as expected from the two monsoon seasons. The air arriving in Singapore is therefore likely to have originated from Vietnam, Cambodia, all areas of Kalimantan, Java, and the island of Sumatra including

Riau. During 2012 the northeasterly wind component was significantly weaker than average~~, also~~ . Also, a small northwesterly component is visible in the air history map (~~not shown here~~Fig. A2c). This means that air was mainly coming from the expected directions given the monsoons in the region with a small additional northwesterly component, so most air arriving in Singapore ~~will have~~ travelled through Peninsular Malaysia, or the island of Sumatra including Riau. During 2013, the same general pattern as 2012 is seen but with stronger northeasterly and westerly components and somewhat weaker southeasterly component ~~. The~~ when the air history maps show a very small region of influence for the MJJ season of 2013. The majority of air arriving in Singapore had travelled ~~through~~ only over Peninsular Malaysia or Riau. ~~During the event of June 2013, a typhoon northeast of Singapore pulled air from westerly directions over Singapore (Gaveau et al., 2014; Hertwig et al., 2015). During~~ During other seasons of this year the air in Singapore arrived from as far away as Vietnam and the Philippines ~~.~~ (Fig. A2d). 2014 was characterised by strong northeasterly and southeasterly components, both of which were stronger than those for 2013 and stronger southeasterly component compared to ~~2012.~~ 2012 (Fig. A2e). The air history map for 2015 (Fig. A2f), shows a strong northeasterly component and the strongest southeasterly component of all six years, these winds brought air from Peninsular Malaysia, Riau and Islands, Sumatra, Kalimantan, Sulawesi, Java, and the Lesser Sunda Islands to Singapore.

Analysis of the annual biomass burning PM_{10} emissions ~~, see~~ (Fig. 3~~,~~) shows that there is a ~~very similar~~ bimodal pattern in the seasons/months with significant burning and also in the dominant source regions~~,~~ . This finding is similar to that of Reddington et al. (2014)~~but hugely varying and with different~~ , though we see differences in the contributing source regions and temporal distribution. The most significant difference between the six years is in the magnitude of burning ~~,~~ - note the different scales of vertical axis ~~.~~ in Fig. 3. Overall, 2015 and 2014 were the years with the highest and second highest annual ($\sim 6.7 \times 10^6$ T and $\sim 4.2 \times 10^6$ T, respectively) and monthly ($\sim 2.7 \times 10^6$ T, October 2015 and $\sim 1.1 \times 10^6$ T, March 2014) emissions~~, respectively~~. 2010 and 2011 saw the lowest annual emissions ($\sim 2 \times 10^6$ T), though 2010 saw the third highest emissions when looking at individual months ($\sim 8.5 \times 10^5$ T, March). 2012 and 2013 saw fairly similar emissions ($\sim 2.5 \times 10^6$ T), which supports the fact that emissions are lower during La Niña and ENSO neutral conditions.

Over the six years, the highest emissions were generally seen during El Niño years and the drought of 2014. This makes sense as the majority of the fires are expected to be anthropogenic, and dry weather provides ideal conditions for initiating and maintaining burning ~~(Reid et al., 2012, 2013)~~(Reid et al., 2012, 2013; Oozeer et al., 2016). Lee et al. (2016b) looked at fire seasons and saw that there is anti-correlation between seasonal variation of fire emissions and that of rainfall, which is likely to be because underground peatland burning may not be immediately extinguished by precipitation. This also supports other papers, e.g., Reddington et al. (2014) who looked at fire/smoke seasons during the period 2004-2009 and found burning peaked from June to October and February to March, with the most burning during September - October.

Observations of PM_{10} in Singapore from 2010 - 2015 show an overall background concentration ~~of approximately 20~~ during months of little or no burning of between 23 - ~~30~~ 29 $\mu g/m^3$ at the two monitoring stations. These values fit well with those determined in other studies for Singapore~~, for example~~. For example, Hertwig et al. (2015) estimated background concentrations for PM_{10} to be around 30 $\mu g/m^3$, based on the 2013 haze episode. In general, both background and peak concentrations vary between NTU and TP. ~~Here~~ Following the approach of Kim et al. (2015) we assume a constant background of 25 $\mu g/m^3$ for the PM_{10} observations at both sites ~~, but because we are~~ and subtract this value from the observation time series.

Subtracting a constant background from the observations does not give the exact contribution of PM_{10} from biomass burning alone because it does not remove all contributions from all other sources. However, it does give an indication of the periods with increased PM_{10} concentrations due to biomass burning. This is not an attempt to perform an *apportionment* of the observed PM_{10} concentrations in Singapore, as the observations, even with the subtracted background concentration, still includes contributions from sources other than biomass burning. However, the observations minus the constant background compared to the modelled time series provides an indication of the performance of the model, and through that the quality of the input used for the modelling. Using the modelled time series and the related source region information we are able to *attribute* the PM_{10} contribution in Singapore originating from biomass burning in Southeast Asia to the respective source regions.

Because we are intentionally leaving out sources of PM_{10} other than biomass burning and there is uncertainty in the biomass burning emissions, we cannot expect perfect scores from the valuation metrics presented in Tables 3 and 4. In the present study a significant haze event has been defined as any period lasting more than one week with modelled hourly PM_{10} concentrations from biomass burning reaching 50 $\mu g/m^3$ or above at ~~at least~~ least at one of the two monitoring stations. Concentrations below 10 $\mu g/m^3$ are considered negligible in terms of haze events.

For years like 2013, ~~that are~~ which was dominated by one extreme haze event, the correlation between the modelled time series and the observations is very high (~~Table 4). To some extent, this is also the case for the 2014~~ 0.79 and ~~2015 events.~~ 0.80 at NTU and TP, respectively, see Table 3). Whereas the correlations for 2010, 2011, and 2012 are very low, which is likely to be due to the low biomass burning PM_{10} emissions and few haze events. In general it can be seen from the MNMB that the model under predicts, even when taking a constant background value of 25 $\mu g/m^3$ into account. This makes sense as the background in reality cannot be assumed to be constant~~, we~~ . We know that we are not capturing all fires, which will lead to a negative bias, and there are further uncertainties in emissions, and the NWP and dispersion models. It should be expected that not all model results fall within a factor of 2 of the observations and it is not surprising that the fractional gross error is around 40 %. It is worth noticing that the FAC2 for all years is high (between 0.76 and 0.87), and in general the FAC2 values for the individual events are also very good. When comparing the scores to other studies such as Chang and Hanna (2004) ~~and Rea et al. (2016)~~ (R = 0.91 - 0.95, FAC2 = 0.24 - 0.89) and Rea et al. (2016) (R = -0.33 - 0.92), it is important to keep in mind that even though the scores presented in Tables 3 and 4 are relatively lower (specifically R) ~~these~~ our statistics are calculated for a three month period and ~~compared studies of periods covering a couple of daysor 1 - 2 weeks, respectively, also the FAC2 is mostly better~~ other studies are for shorter periods focused only on air quality and haze days. Also, for the results presented here the FAC2 values are mostly better than those of Chang and Hanna (2004); Rea et al. (2016). In the ~~discussions of the~~ results below, the estimated background value of 25 $\mu g/m^3$ has been subtracted from all observations. The timeseries and pie charts are based on results from the forward NAME simulations.

Looking at PM_{10} concentrations at the two monitoring sites based on the forward simulations (Fig. 4), five years (all but 2013) have haze during ASO and three years (2011, 2013, and 2014) have some haze in FMA. 2013 is the only year with

Table 3. Statistics for PM_{10}, for both the western (NTU) and eastern (TP) monitoring stations and all years. Background concentration of 25 $\mu g/m^3$ is subtracted from the observations for all stations for all years. The metrics considered are the Pearson correlation coefficient (R), the modified normalised mean bias (MNMB), the fractional gross error (FGE), and Factor of 2 (FAC2).

	2010		2011		2012		2013		2014		2015	
	NTU	TP	NTU	TP	NTU	TP	NTU	TP	NTU	TP	NTU	TP
R	0.12	0.12	0.08	0.13	0.17	0.18	0.79	0.80	0.27	0.35	0.44	0.43
MNMB	0.14	0.17	0.10	0.11	0.09	0.05	0.04	0.12	-0.09	0.07	-0.19	0.01
FGE	0.39	0.45	0.37	0.35	0.39	0.37	0.37	0.38	0.36	0.36	0.44	0.43
FAC2	0.83	0.76	0.85	0.86	0.83	0.85	0.84	0.83	0.86	0.87	0.78	0.79

Table 4. Statistics for PM_{10}, for both the western (NTU) and eastern (TP) monitoring stations, for selected 3 months haze seasons. Background concentration of 25 $\mu g/m^3$ is subtracted from the observations for all stations for all seasons. The metrics considered are the Pearson correlation coefficient (R), the modified normalised mean bias (MNMB), the fractional gross error (FGE), and Factor of 2 (FAC2).

	2010		2011		2012		2013		2014		2014		2015	
	ASO		ASO		ASO		MJJ		FMA		ASO		ASO	
	NTU	TP	NTU	TP	NTU	TP	NTU	TP	NTU	TP	NTU	TP	NTU	TP
R	0.15	0.14	0.08	0.15	0.14	0.14	0.81	0.83	0.30	0.42	0.29	0.40	0.35	0.32
MNMB	0.12	0.13	-0.07	-0.01	-0.24	-0.22	-0.14	0.03	-0.13	-0.07	-0.31	-0.06	-0.65	-0.47
FGE	0.41	0.49	0.38	0.34	0.40	0.38	0.40	0.43	0.29	0.30	0.35	0.36	0.71	0.61
FAC2	0.80	0.72	0.86	0.87	0.83	0.84	0.82	0.78	0.93	0.93	0.76	0.86	0.49	0.60

significant haze in June, although the years from 2012 to 2015 all experience some additional PM_{10} from biomass burning in June. When comparing concentrations between the two stations it can be seen that the concentrations are higher at the western monitoring station (NTU) most of the time~~. The opposite, concentrations at the eastern monitoring stations being higher than at the western station, was the case during March in~~. Exceptions to this occurred during March 2011 and 2014. Of the haze events that occurred from 2010 through 2015, some were insignificant (e.g., during FMA 2010, 2012, 2013, and 2015, and MJJ 2012 and 2014), ~~some~~ i.e., lasting less than a week and with biomass burning PM_{10} concentrations below 50 $\mu g/m^3$. Some were significant but showed very little variation between monitoring stations (ASO 2010, MJJ 2013, FMA 2011 and 2014) (~~See~~Sect. 3.1)~~the~~. The remaining four events (ASO 2011, 2012, 2014, and 2015) (~~See~~Sect. 3.2), were significant events ~~, though,~~ with variation in the main contributing source regions at the two monitoring stations. Common for all four events is that they occurred during the haze season in ASO during the southeast monsoon, when the winds are the strongest for the region and the air history maps show the largest region of influence for air arriving in Singapore. Not all peaks in the observations coincide with biomass burning due to real PM levels also containing anthropogenic and other biogenic species~~,~~

~~however,~~ . However, most peaks in the modelled time series coincide with peaks in observations ~~.~~ indicating that the highest PM_{10} concentrations are due to biomass burning.

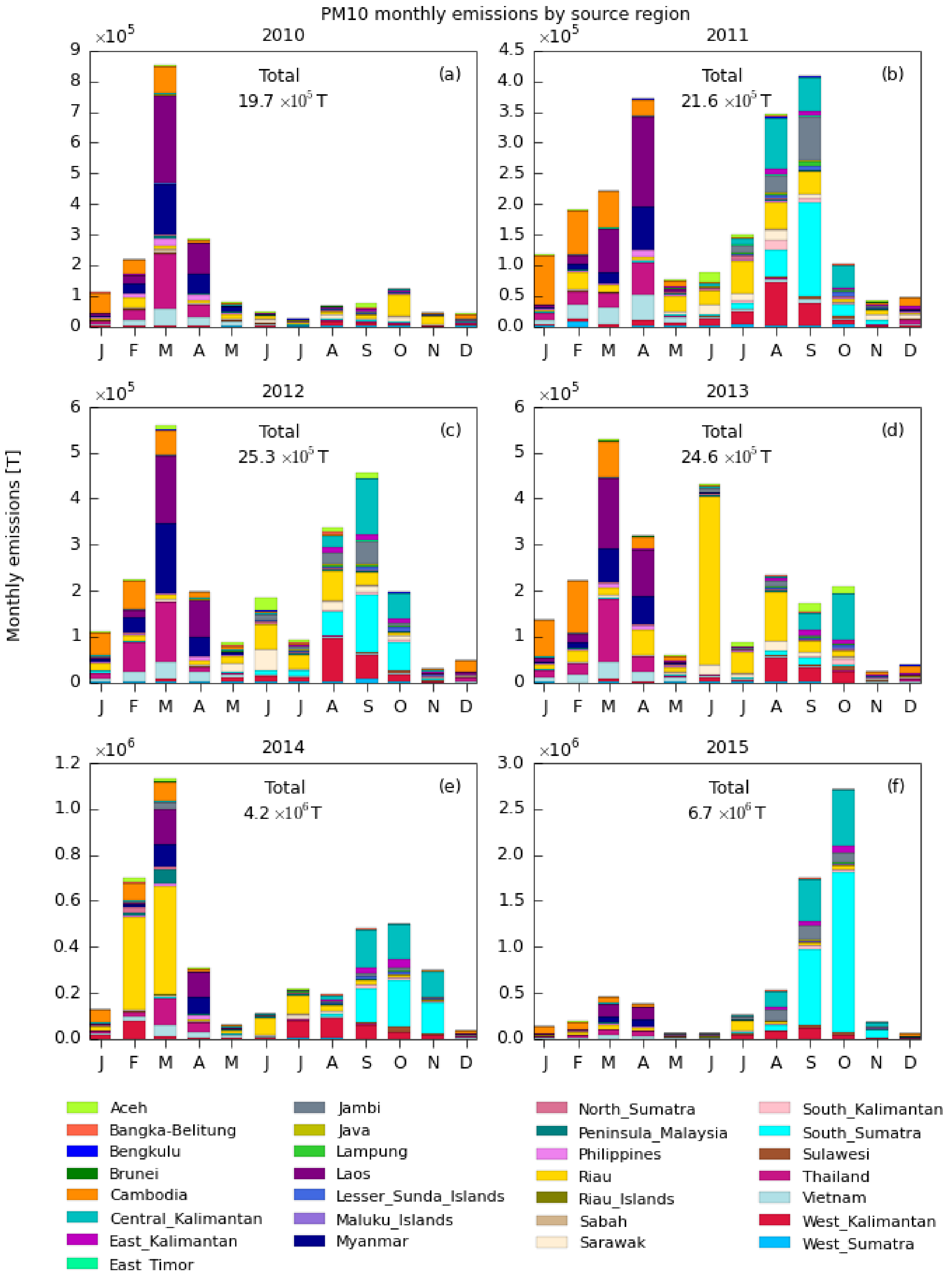

Figure 3. Caption on next page

Figure 3 (Continued). Regional PM_{10} biomass burning emissions, calculated based on GFAS fire radiative power and injection height and emission factors described in Sect. 2, for each of the six years from 2010 to 2015, summed over each month. Colours for each source region for all years are listed below the plots. Note the different scales on the y-axis, units: tonnes emitted per month.

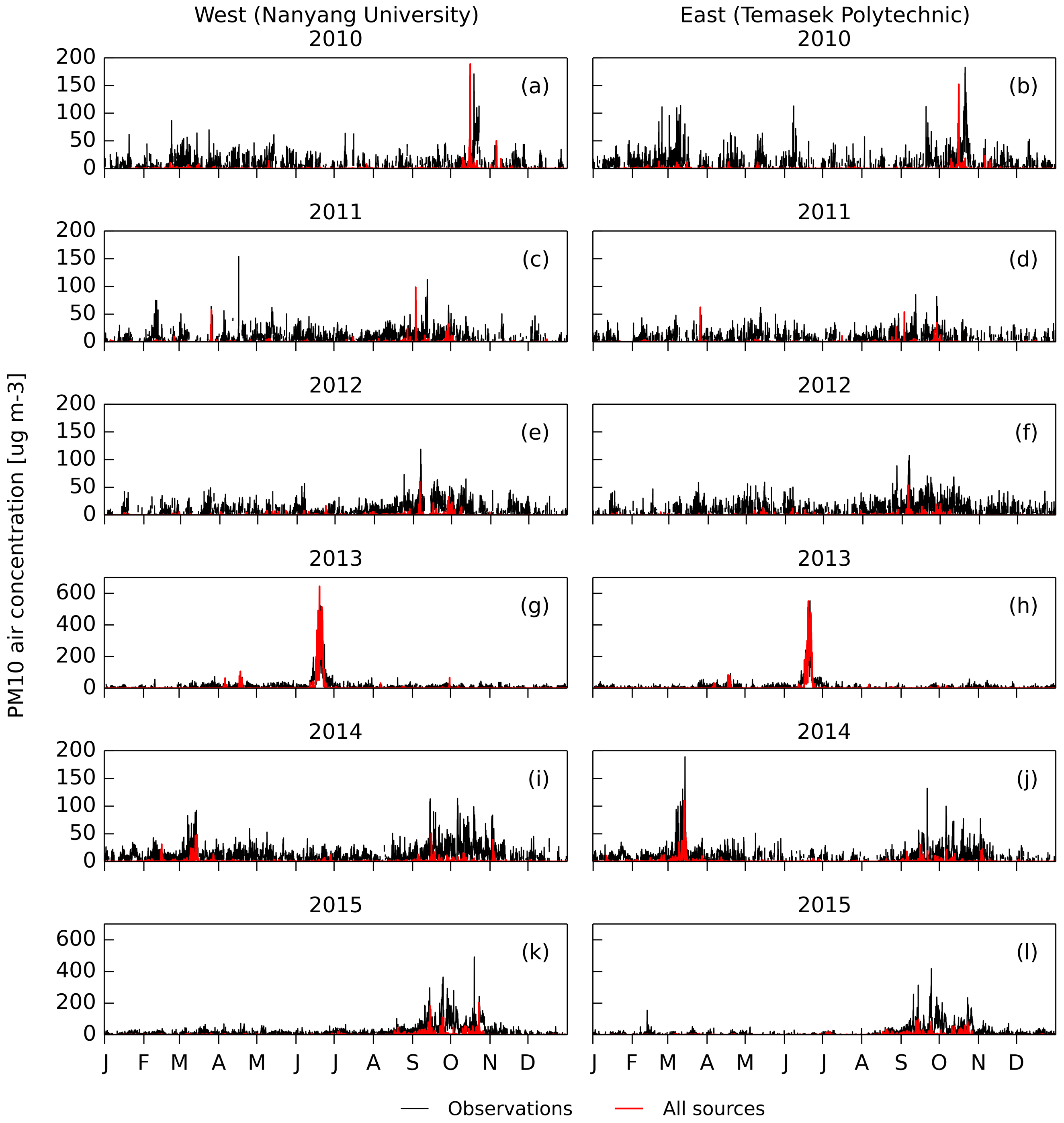

Figure 4. Modelled PM_{10} time series (red line) with observations (black line) at each of the two monitoring stations West (NTU, left) and East (TP, right) for the six years with observations available, 2010 (top row) - 2015 (bottom row). A constant background concentration of $25\mu g/m^3$ has been subtracted from the observations and any resulting negative values have been removed.

3.1 Atypical haze

During the six years, the most notable atypical haze events occurred in June 2013 and February, March, April 2014. Though 2013 was ~~a very unique year~~generally a year with weak winds and average burning, the month of June was very unique, both in terms of meteorology and burning (Fig. 5)~~, the event of~~. The June 2013 haze event was caused by a typhoon coinciding with intense burning in Riau ~~during June~~ (Fig. 3)~~, in what was generally a year with weak winds and average burning~~. The air history map for MJJ in Fig. 5 shows that, during this weather event, there was a small source region with air arriving in Singapore from Peninsular Malaysia, Riau Islands, and Sumatra including Riau. This is the only year of this six year period with significant burning in June, though in general the annual emissions are neither especially high nor low. In June about 98 % of the modelled PM_{10} emissions reaching the two monitoring stations in Singapore were from Riau. ~~The maximum modelled/observed concentrations in June reached 640/525 and 550/550~~ ~~$\mu g/m^3$~~ ~~at NTU and TP, respectively, as is seen from the time series in Fig 4.~~ Although the peak concentrations observed at NTU were lower than those of the modelled time series, overall the concentrations are fairly similar during the event.

In early 2014, a drought coincided with air arriving in Singapore from a northeasterly direction and intense burning in the whole region giving the second highest emissions of the six year period. This resulted in unexpected haze in Singapore in FMA (Fig. 6). The months with the largest emissions were March and February which were dominated by emissions from Riau, Laos, Myanmar, Thailand, Cambodia, Peninsular Malaysia, and West Kalimantan (Fig. 3d). In general the region of influence for 2014 covered an area reaching far to the northeast and slightly south-east of Singapore and was much larger than for MJJ ~~2013.~~ 2013 (Fig. 5). During FMA the winds brought air from Peninsular Malaysia, Riau, Riau Islands, and the Philippines to Singapore. ~~The event saw modelled and observed~~ ~~PM_{10}~~ ~~concentrations of up to 50/100~~ ~~$\mu g/m^3$~~ ~~and 110/200~~ ~~$\mu g/m^3$~~ ~~at NTU and TP, respectively, i.e., concentrations at TP are about double of those at NTU for both the modelled time series and the observations~~In spite of the larger emissions from Riau, Laos, Myanmar, Thailand, and Cambodia, the mainly northerly wind direction resulted in the haze in Singapore being caused mainly by emissions from Peninsula Malaysia. The event lasted for about 3 months total, and was dominated by emissions from Peninsular Malaysia, which contributed over 90 % of the haze at both monitoring stations, with smaller contributions from Riau, Cambodia, Vietnam, ~~Cambodia, Vietnam,~~ and Riau Islands.

Common for these two atypical haze events is little variation in the source regions across the monitoring stations, ~~in spite of~~ most likely due to the atypical and different meteorological conditions ~~,~~ and the clear dominance of one source region.

3.2 ~~Southeast~~ ASO - southeast monsoon season haze

~~The~~ As mentioned previously, the southeast monsoon season occurs during ASO and coincides with almost annual haze episodes. The two most recent episodes with highest concentrations were in 2014 and 2015. In addition to the haze event in FMA 2014 discussed above, another haze event occurred in 2014 during ASO (Fig. 7). This season saw the largest south-easterly region of influence for air arriving in Singapore during the six year period, with air and PM_{10} from biomass burning pollution arriving in Singapore from Peninsular Malaysia, Riau, Riau Islands, Kalimantan, Java, and the Lesser Sunda Islands, during a period of average biomass burning emissions. ~~In September-October the~~ During the two months of September and

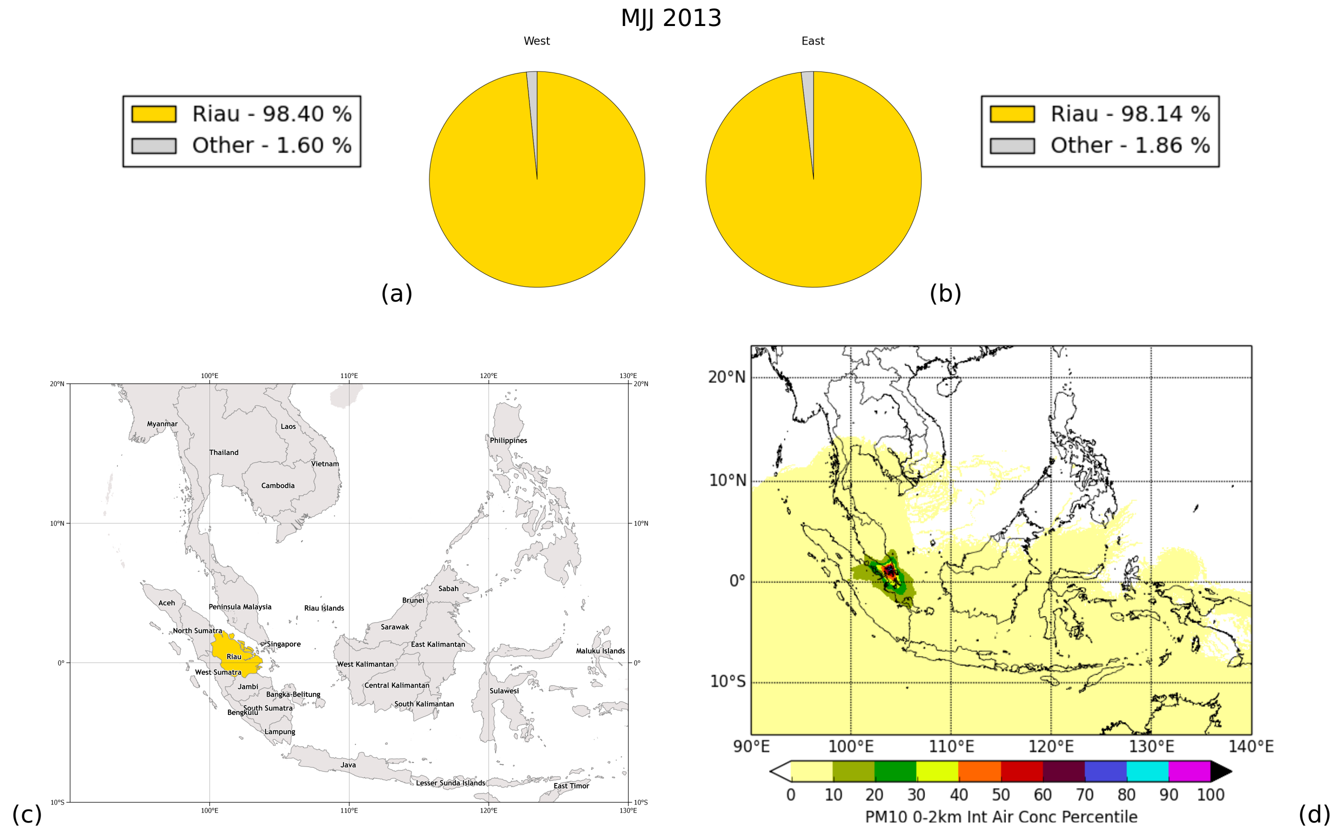

Figure 5. This figure shows results for PM$_{10}$ for MJJ 2013: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting only the major contributing source region, and the air history map (d) showing where the air arriving in Singapore originated from in MJJ 2013. The 'Other' category in the pie charts is from sources which individually contribute less than 1 %.

October the major contributing source regions to PM$_{10}$ concentrations in Singapore were Central Kalimantan, South Sumatra, and West Kalimantan ~~. The event lasted about two months, and reached peak modelled and observed concentrations of about 50/120 $\mu g/m^3$ and 30/125 $\mu g/m^3$ at NTU and TP, respectively.~~ (Fig. 3e). ASO is the expected haze season, however, this is also one of the seasons with the highest number of significant contributing source regions: South Sumatra, Central Kalimantan, West Kalimantan, Bangka-Belitung, Riau, Riau ~~, Riau~~ Islands, and the Lesser Sunda Islands (~~approx.~~ up to 2000 km from Singapore). In spite of the large annual variation (Fig. A3) in the major contributing source regions between the two monitoring stations, the difference between the relative contributions at the two stations for ASO 2014 is insignificant.

The ~~plots~~ results for ASO 2015 ~~,~~ (Fig. 8~~,~~) show a large, though seasonally "normal" region of influence, ~~and this~~ which coincided with extreme emissions. In ASO the southeasterly monsoon winds brought air from Peninsular Malaysia, Riau Islands, Sumatra including Riau, Kalimantan, Sulawesi, Java, and the Lesser Sunda Islands. During this season the largest

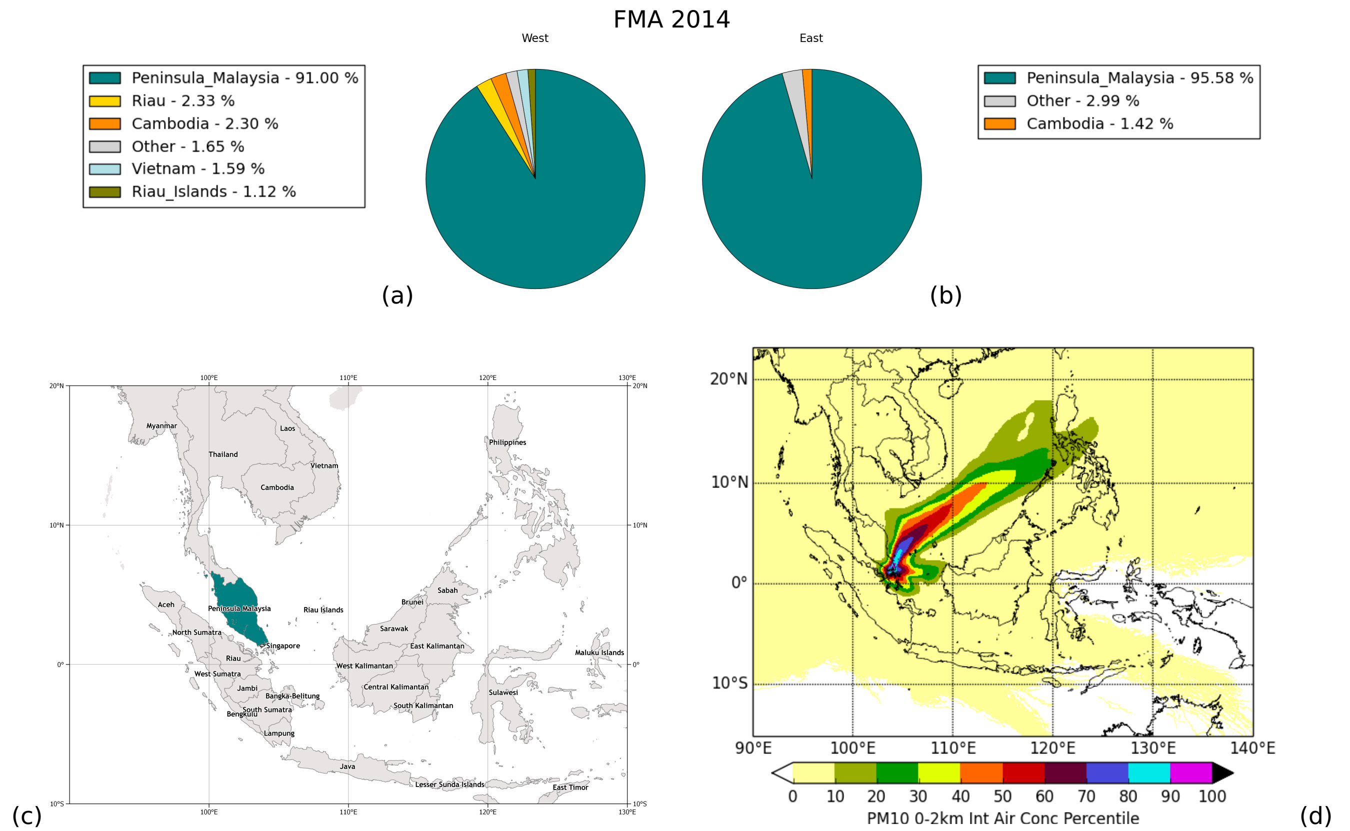

Figure 6. This figure shows results for PM_{10} for FMA 2014: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting only the major contributing source region, and the air history map (d) showing where the air arriving in Singapore originated from in FMA 2014. The 'Other' category in the pie charts is from sources which individually contribute less than 1 %.

contributing regions were Central Kalimantan, South Sumatra, and West Kalimantan. The event lasted approximately 2.5 months ~~, and peak modelled/observed concentrations reached over 200/500~~ $\mu g/m^3$ ~~at NTU - the modelled concentrations up to twice as high as those at TP, where concentrations reached 100/425~~ $\mu g/m^3$~~. During~~ in ASO 2015, during which the biggest variation between the two monitoring stations ~~of the year and~~ was seen both for 2015 and for any season with significant burning~~was seen. By monitoring station the~~. The most significant source regions at the western and eastern monitoring stations (NTU, TP) were South Sumatra (38.22 %, 21.82 %), Central Kalimantan (31.19 %, 41.45 %), Bangka-Belitung (11.32 %, 13.64 %), West Kalimantan (6.64 %, 9.41 %), and Jambi (6.53 %, 5.98 %).

~~Common for~~ Common for both ASO 2014 and ASO 2015 ~~, and also~~ are the relatively large regions influencing PM_{10} concentrations in Singapore and the variation in major contributing source regions at the two monitoring stations. This is also the case for other years with burning and related haze during this season (e.g., 2011 and 2012)~~, are the relatively large regions~~

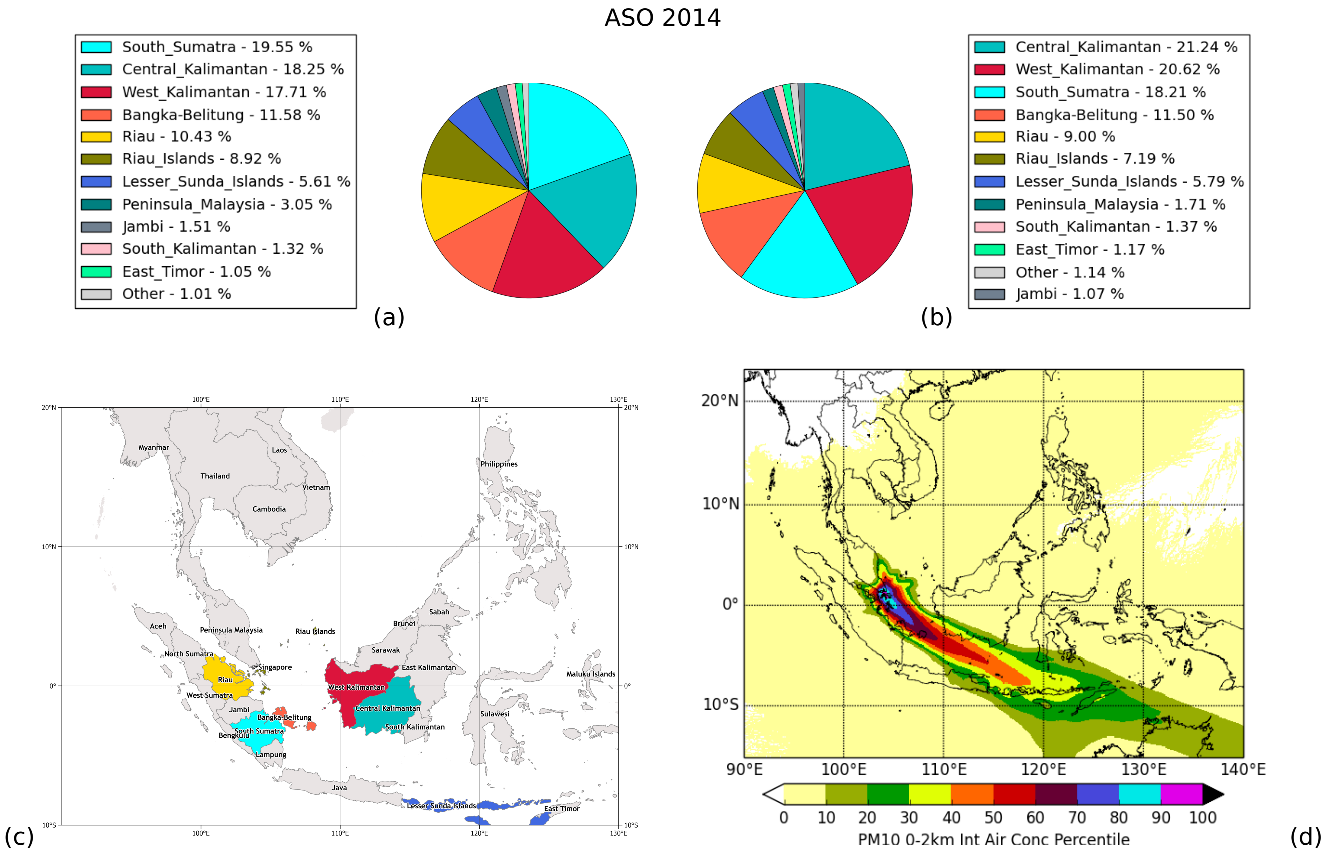

Figure 7. This figure shows results for PM_{10} for ASO 2014: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting only the major contributing source regions, and the air history map (d) showing where the air arriving in Singapore originated from in ASO 2014. The 'Other' category in the pie charts is from sources which individually contribute less than 1 %.

~~influencing PM_{10} concentrations in Singapore and the significant variation between monitoring stations~~.

In addition to the four events discussed in detail above, events also occurred during the expected haze ~~season~~ seasons in ASO 2010, 2011, and 2012, as well as during FMA 2011. The ASO event in 2010 was, except for significantly lower magnitude, fairly similar to the MJJ event of 2013, with an unusually small source region for the season and at least 90 % of PM_{10} concentrations arriving at both monitoring stations in Singapore originating from Riau. The other two ASO events, in 2011 and 2012, were fairly similar to the events of 2014 and 2015 with contributions from the expected southeast monsoon region~~and~~, a high number of contributing source regions at the two monitoring stations, and variations in major contributing source region between the two stations. The remaining event of the period was during FMA 2011, with Riau, Peninsular Malaysia, and Cambodia as major contributing source regions.

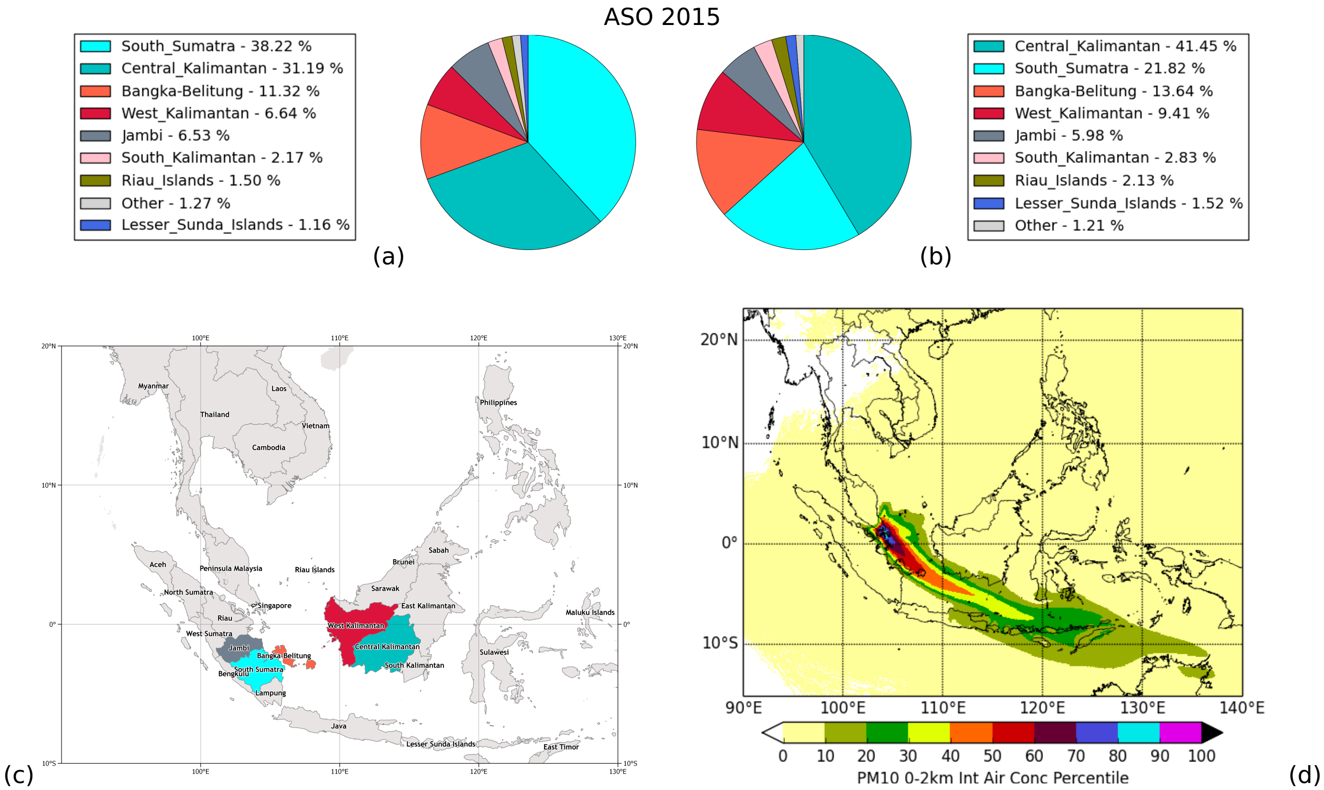

Figure 8. This figure shows results for PM_{10} for ASO 2015: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting only the major contributing source regions, and the air history map (d) showing where the air arriving in Singapore originated from in ASO 2015. The 'Other' category in the pie charts is from sources which individually contribute less than 1 %.

4 ~~Discussions~~

~~For~~ Of the seasons with the most significant haze events (e.g., MJJ 2013, FMA 2014, ASO 2014, and ASO 2015) in Singapore, the air history maps show that the region of influence for Singapore generally covers the largest area during ASO ~~with air~~ when air is coming from southeasterly directions. Of the four years (2011, 2012, 2014, 2015) with haze events during ASO, 2014 saw the largest region of influence. Of the two years with events during FMA (2011 and 2014) the winds were generally from a northeasterly direction and 2014 was, again, the year influenced by the largest source region. For seasons with southeasterly winds, but not during ASO, e.g., 2012 MJJ, the region of influence is relatively small compared to that of ASO. ~~Similarly to the results~~Our results, presented in Figure 3, ~~Lee et al. (2016b)~~ confirm the findings of other studies such as Lee et al. (2016b) who determined the source region for Singapore to be mainly Sumatra and Borneo (i.e., Kalimantan, Sarawak, Sabah, and Brunei)~~, and~~ . Shi and Yamaguchi (2014) also saw that the biggest emitters include South Sumatra and South Kalimantan, showing that spring emissions mainly originate from Cambodia, Laos, Myanmar, Thailand, Vietnam, and on occasion Peninsular Malaysia, whereas, autumn burning is seen in Central Kalimantan, Jambi, South Sumatra, West Kalimantan, and to a lesser extent Aceh and East Kalimantan. Emissions from Riau vary significantly throughout the years and individual months, though there are emissions from Riau in most months during most years, which is consistent with the emissions shown in Fig. 3.

~~When comparing all years, the results show large variability in~~

4 Conclusions

In this study we have used the atmospheric dispersion model, NAME, to attribute PM_{10} concentrations ~~and major contributing source regions between years and ASO events. Common for the atypical haze events is little variation across monitoring stations in spite of the unique and different meteorological conditions. The biggest difference between both modelled and observed concentrations at the two stations were seen during FMA 2014 and ASO 2015, with highest concentrations at TP and NTU, respectively. Common for ASO 2014 and ASO 2015, and also for some of the other years with burning and related haze during this season, are PM~~in Singapore caused by biomass burning to their source region. In order to gain a deeper understanding of the causes of haze in Singapore we have compared air history maps, showing where air arriving in Singapore originates from, with modelled and observed PM_{10} ~~contributions from a large area arriving in Singapore and related significant variation between monitoring stations. The results show that low emissions often lead to low concentrations that still affect the air quality in Singapore , and that a larger area of influence brings more variation between major contributing source regions at the~~ concentrations at two monitoring stations ~~. The largest differences between monitoring stations are seen in the annual comparison of the major contributing source regions, the variation during ASO is likely due to the larger region of influence seen during the south-easterly monsoon, which brings air and~~ ~~PM_{10}~~ ~~from biomass burning from further away. In general there were often bigger differences in contributing source regions between the~~ located at a western and an eastern location, respectively. For those two monitoring stations ~~when no significant haze events occurred in Singapore. Atypical events are often dominated by one and the same source region at both monitoring stations, whereas there is more difference between dominating source regionsat the two monitoring stations for haze events during the expected haze seasons.~~we have also compared the difference between relative contributions from all of the source regions.

The yearly and seasonal variations in emissions of PM_{10} from biomass burning from the region are not always correlated with PM_{10} concentrations in Singapore~~, which shows that haze~~. Yet the modelled results confirm that the highest PM_{10} concentrations in Singapore coincide with haze caused by biomass burning. The results show that haze in Singapore is impacted by (1) burning emissions under human influence (e.g., Fig. 3), (2) the weather through the monsoon and related winds (Fig. A2), and (3) climate, especially the variations in ENSO, ~~this~~ which is also in line with the findings by Reid et al. (2012, 2013). In previous similar studies it has been assumed that the same emission inventory can be used for different years (Kulkarni et al., 2015; Sobhani et al., 2018), and some attribution studies even used the same meteorology when studying different years (Kim et al., 2015). Our findings demonstrate that this is not sensible for biomass burning due to the inter-annual variability of both meteorology and emissions, which can be extremely high both spatially and temporally (Kelly et al., 2018).

For the four haze events focused on here, there is variability in the correlation between the modelled and observed time series, with the best correlations seen for haze events where the emission sources are close to Singapore. As discussed by Hertwig et al. (2015), ~~sources of~~ uncertainty in these results ~~originate~~ originates from the emissions and the meteorology. For the former, the uncertainties result from the fact that the emissions used here are based on one daily snapshot of FRP and IH, and though some attempts are made to ~~solve issues~~ resolve issues with missing fire emissions caused by the lack of transparency of clouds the data will naturally be incomplete. At the same time, hourly emissions are calculated based on this one daily snapshot adding a temporal resolution that the data does not provide, which also means that peak concentrations ~~won't~~ will not always be captured in the model simulations. ~~One~~ The meteorology provides another significant source of uncertainty, as is usually the case in atmospheric modelling~~is the meteorology~~. When considering the resolution of the analysis meteorology used here and the size of Singapore it is clear that there will be unresolved features in both topography and in the meteorology and hence in the dispersion modelling. ~~The~~ However, the differences we see between the two sites show that we are starting to capture this scale. Uncertainties in the NWP data such as elevated wind speeds and too frequent and too low intensity precipitation will disperse the pollutants further and wash out more than should be, resulting in lower modelled concentrations. These uncertainties naturally have a larger impact over longer travel distances, which is reflected in our statistics. It should also be kept in mind that the observations are measuring all PM_{10} and we are only modelling primary PM_{10} emissions from biomass burning. Other sources of PM_{10} include sea salt, dust, secondary organic aerosol, emissions from industry, local and transboundary road traffic, as well as domestic heating, not all of which are constant throughout the year. Some of the varying difference between observed and modelled time series is ~~due to the~~ also likely to be due to these many other sources of PM_{10} in Singapore. However, in spite of these uncertainties our results show that we are able to model dispersion of particulate matter from biomass burning in Southeast Asia and the resulting haze in Singapore with reasonable confidence.

~~In this study we have used the atmospheric dispersion model, NAME, to attribute PM_{10} concentrations in Singapore caused by biomass burning to their originating source region. In order to gain a deeper understanding of the causes of haze in Singapore we have compared air history maps, showing where air arriving in Singapore originates from, with modelled and observed PM_{10} concentrations at two monitoring stations located at a western and an eastern location, respectively. For those two monitoring stations we have also compared the difference between relative contributions from all of the source regions.~~

~~The concentrations and major contributing source regions at the two monitoring stations vary significantly both on a yearly and seasonal basis. The results show that haze caused by off-season/atypical burning often occurs during periods of low wind, which results in little variation in both source regions and in the relative contributions across the Singapore. However, the southeasterly monsoon wind creates ideal conditions for variation in contributing source regions and concentrations across Singapore, i.e., the larger region of influence during the "expected" biomass burning/haze periods means that the air arriving in Singapore originates from several regions with biomass burning, however it is important to note that the region with highest emissions isn't necessarily the major contributing source region in Singapore. Smaller contributions of PM_{10} from biomass burning arrive in Singapore throughout the year, most years, and in addition larger events occur on an approximately yearly basis. The variation between monitoring stations is often caused by smaller events with varying contribution across Singapore.~~

~~These simulations only consider emissions from biomass burning, no background concentrations are taken into consideration, which explains some of the difference between the modelled and observed concentrations.~~

Emissions from many regions contribute to the concentrations of PM_{10} in Singapore~~, the~~. The biggest contributors for the period 2010 - 2015 are Riau, Peninsular Malaysia, and South Sumatra, with smaller yet significant contributions from Jambi, Cambodia, Bangka-Belitung, Riau Islands, Central Kalimantan, and the Philippines. ~~Seeing as~~ As Riau and Peninsular Malaysia are the nearest neighbours to Singapore and given the local wind pattern this ~~makes sense~~could be expected. Looking at emissions during ASO for the four years with ~~largest~~ the most variation across the island (2011, 2012, 2014, and 2015), ~~large~~ the largest emissions were seen from Central Kalimantan, South Sumatra, Jambi, and also West Kalimantan~~, whereas~~. For events during FMA Cambodia, East Kalimantan, Myanmar, Thailand, and Vietnam showed larger emissions~~during FMA.~~ .

~~Difference in magnitude between monitoring stations and higher PM_{10} concentrations does not necessarily impact the ratio of~~ We investigated the spatial variation of haze across Singapore and found that variation in major contributing source regions ~~between the monitoring stations, over the six year period the biggest annual difference between stations was seen in 2011, 2014, and 2015. The highest concentrations during periods with contributions from a large region of influence was seen in 2014 all year and ASO 2015 and with smaller regionof influence during 2010 and 2013. Generally, a larger southeasterly region of influence is seen during ASO for all years except 2010, whereas the northeasterly winds that dominate FMA, and were seen in 2011 and 2014, generally are weaker than the southeast monsoon winds. The two 2014 events have the largest region of influence~~ across Singapore is dependent on distance to source regions: generally a shorter distance to the source region will mean less variation in the major contributing source region(s). We have also studied the seasonal variation by looking at four recent events occurring during different seasons and saw that air arriving from a larger geographical area often brings more variation in major contributing source regions. PM_{10} concentrations at the two monitoring stations vary significantly in time, both in the observed and modelled time series; from the modelled data it is possible to attribute the major contributing source regions. These show that for the two haze events not occurring during the ASO haze season, the sources are dominated by the same source region at both sites, though a different site for the two ~~seasons over the six year period. Southeasterly winds not during ASO (e. g., MJJ 2012) are relatively weaker. The air history map for MJJ 2013 shows a small region of influence for air arriving in Singaporeduring the atypical event where a typhoon was dragging air from Riau over Singapore.~~ events. For the two ASO haze events the major contributing source regions at the two monitoring sites are mainly the same but their relative contribution differ significantly. These variations are also correlated with the distance to the source regions and the season of the haze events.

The NAME model is able to provide insight into variations in major contributing source regions at a relatively smaller scale than has been done in previous studies due to its tracking capabilities and the Lagrangian nature of the model. Although the results struggle to capture the magnitude of the haze from burning further from Singapore, due to errors and uncertainties in the GFAS data and the meteorological input, they show the potential for gaining a better understanding by using higher spatial resolution. This work is a first step towards high resolution air quality forecasting for Singapore. Whilst a chemical transport model would be expected to fully capture anthropogenic and secondary particulate contributions, the inability of this study to capture the magnitude of the biomass burning concentrations shows that there is a bigger issue with emissions and potentially also modelled meteorology. Prior to investing in a full chemical transport model it is important to understand these individual components in the simulation. This work contributes towards a better understanding of the biomass burning and air quality in the region and shows that biomass burning emissions from many different source regions across Southeast Asia can reach Singapore. Accurately capturing these is essential for future air quality modelling.

In conclusion, we saw that haze events occur during seasons with both small and large regions of influence, however, most often during ASO, coinciding with a larger region of influence and often when higher emissions/increased burning occurs, resulting in variation in relative contributions from major contributing source regions across Singapore. The results emphasise the inter-annual variation between haze events and major contributing source regions, and show that Peninsular Malaysia is a dominant source of particulate matter from biomass burning for the maritime continent off-season burning impact on Singapore ~~, see Figure A4~~(Figure A4). For haze to occur in Singapore, burning is required, but so is dry weather and wind in the "right" direction. Haze comes from burning across Southeast Asia, making it a transboundary issue for the whole region. Considering that the distance from, e.g., Kalimantan to Singapore is over 500 km, this study emphasises the long-range nature of the problem.

As ~~it~~ an extension of the current study it would be interesting to gain insight into the seasonality and the relative magnitude of PM_{10} from other contributors such as industry, traffic, and domestic heating in Singapore. Further, as it is known that biomass burning varies on ~~time~~ sub-daily timescales (Reid et al., 2013), and this study has used daily GFAS FRP and IH (Kaiser et al., 2012) for source calculation, in the future it would be interesting to study the impact of sources based on higher than daily resolution. One could also use post fire inventories based on burnt area or conduct an inversion study, running NAME backwards from detection sites to estimate the emissions in certain areas corresponding to concentrations observed in Singapore and other locations in Southeast Asia. These results could also be compared to inventories based on satellite observations to help quantify how much burning is missing in such inventories.

Code and data availability. The NAME model and data are available by request to the Met Office, GFAS data available through the Copernicus Atmospheric Monitoring Service (CAMS).

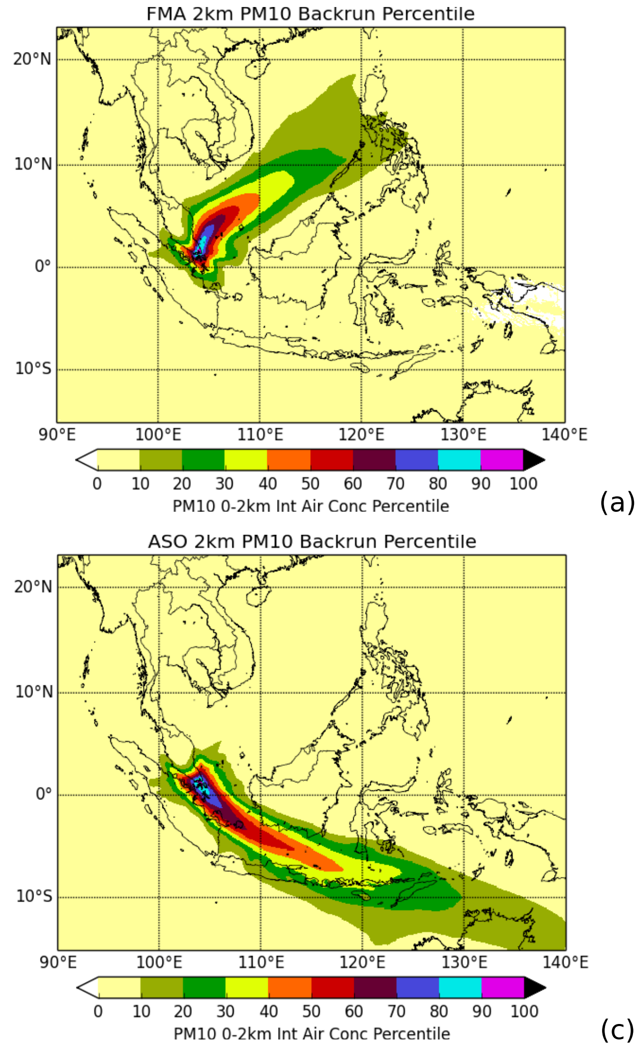

Figure A1. Air history maps for each of the four seasons (a) FMA, (b) MJJ, (c) ASO, and (d) NDJ, averaged over the years 2010 to 2015, showing where air arriving in Singapore during each season originated from. The backruns shown were conducted from a receptor site in central Singapore.

Author contributions. ABH performed most of the attribution model simulations, the data analysis and wrote the paper in collaboration with CW. WMC performed the simulations for and the visualisation of the air history maps, EK performed additional attribution model simulations, and assisted with visualisation and calculation of error metrics, BNC, CG, MCH, and SYL helped design the model setup and provided feedback on the manuscript.

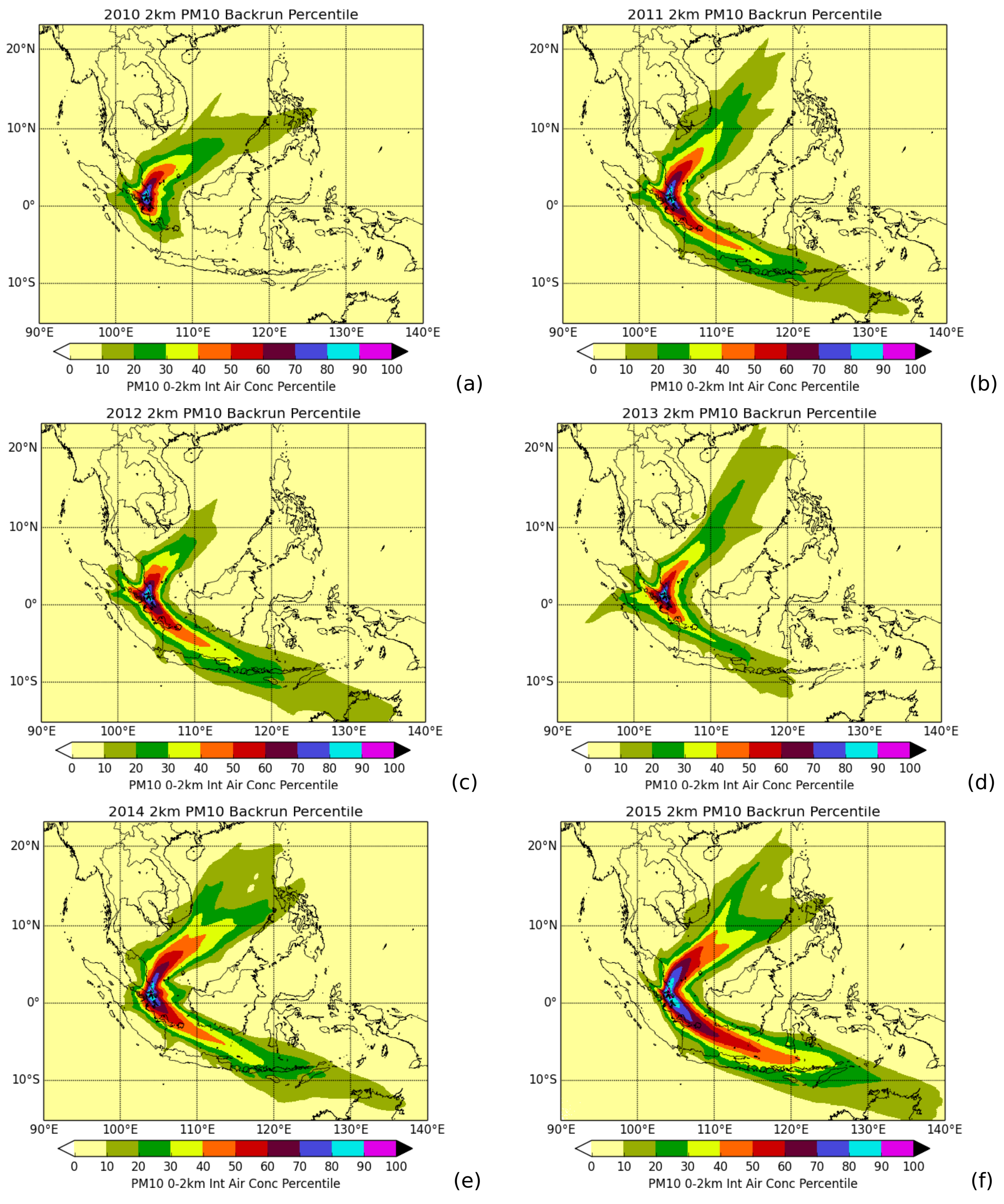

Figure A2. Air history maps for the years 2010 to 2015, showing where air arriving in Singapore during each year originated from. The backruns shown were conducted from a receptor site in central Singapore.

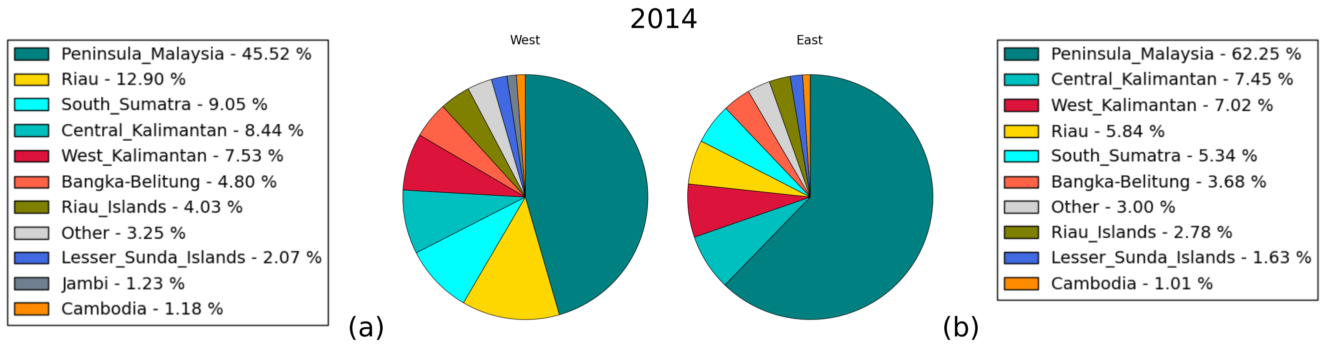

Figure A3. Attribution results for PM_{10} for 2014: major contributing source regions for the western (NTU) (left) and eastern (TP) (right) monitoring stations.

Competing interests. No competing interests are present

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~~Locations and colour codes used for each of the 29 biomass burning source region within the domain from 10°S - 20°N and 90°E - 130°E considered in this study. Singapore is located south of Peninsular Malaysia and East of Riau.~~

~~Air history map for 2010 - 2015, showing where air arriving in Singapore during this period originated from. The backruns shown were conducted from a receptor site in central Singapore. Comparison between a coastal receptor site and this inland site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island.~~

~~Regional PM_{10} biomass burning emissions, calculated based on GFAS FRP and IH and emission factors described in Sec. 2, for each of the six years from 2010 to 2015, summed over each month. Colours for each source region for all years are listed below the plots. Note the different scales on the y-axis, units: tonnes emitted per month.~~

~~Modelled PM_{10} time series with observations (solid black line) at each of the two monitoring stations West (NTU, left) and East (TP, right) for the six years with observations available, 2010 (top row) - 2015 (bottom row). A constant background concentration of $25\mu g/m^3$ has been subtracted from the observations and any resulting negative values have been removed.Note the different scales on the y-axis, units: $\mu g/m^3$~~

~~This figure shows results for PM_{10} for MJJ 2013: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting the major contributing source~~

~~region, and the air history map (d) showing where the air arriving in Singapore originated from in MJJ 2013. The 'Other' category in the pie charts is the contributions from sources which individually contribute with less than 1 %.~~

~~This figure shows results for PM_{10} for FMA 2014: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting the major contributing source region, and the air history map (d) showing where the air arriving in Singapore originated from in FMA 2014. The 'Other' category in the pie charts is the contributions from sources which individually contribute with less than 1 %.~~

~~This figure shows results for PM_{10} for ASO 2014: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting the major contributing source regions, and the air history map (d) showing where the air arriving in Singapore originated from in ASO 2014. The 'Other' category in the pie charts is the contributions from sources which individually contribute with less than 1 %.~~

~~This figure shows results for PM_{10} for ASO 2015: Pie charts for the western (NTU) (a) and eastern (TP) (b) monitoring stations showing major contributing source regions, (c) shows the regional map highlighting the major contributing source regions, and the air history map (d) showing where the air arriving in Singapore originated from in ASO 2015. The 'Other' category in the pie charts is the contributions from sources which individually contribute with less than 1 %.~~

~~Statistics for PM_{10}, for both monitoring stations and all years. Background concentration of 25 $\mu g/m^3$ is subtracted from the observations for all stations for all years. 2010 2011 2012 2013 2014 2015 NTU TP NTU TP NTU TP NTU TP NTU TP NTU TP R 0.12 0.12 0.08 0.13 0.17 0.18 0.79 0.80 0.27 0.35 0.44 0.43 MNMB 0.14 0.17 0.10 0.110.09 0.05 0.04 0.12 -0.09 0.07 -0.19 0.01FGE 0.39 0.45 0.37 0.35 0.39 0.37 0.37 0.38 0.36 0.36 0.44 0.43 FAC2 0.83 0.76 0.85 0.86 0.83 0.85 0.84 0.83 0.86 0.87 0.78 0.79~~

~~Statistics for PM_{10}, for both monitoring stations, for selected haze seasons. Background concentration of 25 $\mu g/m^3$ is subtracted from the observations for all stations for all years. 2010 2011 2012 2013 2014 2014 2015 ASO ASO ASO MJJ FMA ASO ASO NTU TP NTU TP NTU TP NTU TP NTU TP NTU TP NTU TP R 0.15 0.14 0.08 0.15 0.14 0.14 0.81 0.83 0.30 0.42 0.29 0.40 0.35 0.32 MNMB 0.12 0.13 -0.07 -0.01 -0.24 -0.22 -0.14 0.03 -0.13 -0.07 -0.31 -0.06 -0.65 -0.47 FGE 0.41 0.49 0.38 0.34 0.40 0.38 0.40 0.43 0.29 0.30 0.35 0.36 0.71 0.61 FAC2 0.80 0.72 0.86 0.87 0.83 0.84 0.82 0.78 0.93 0.93 0.76 0.86 0.49 0.60~~

~~Air history maps for the years 2010 to 2015, showing where air arriving in Singapore during each year originated from. The backruns shown were conducted from a receptor site in central Singapore. Comparison between a coastal receptor site and this inland site showed insignificant variation, meaning that the central receptor site can be considered representative for the whole island.~~

~~This figure shows results for PM_{10} for 2014: major contributing source regions for the western (NTU) (left) and eastern (TP) (right) monitoring stations.~~

~~This figure shows results for PM_{10} for years 2010 - 2013 and 2015 for FMA: major contributing source regions for the western (NTU) (left) and eastern (TP) (right) monitoring stations. (For 2014 FMA, see Fig. 6.)~~

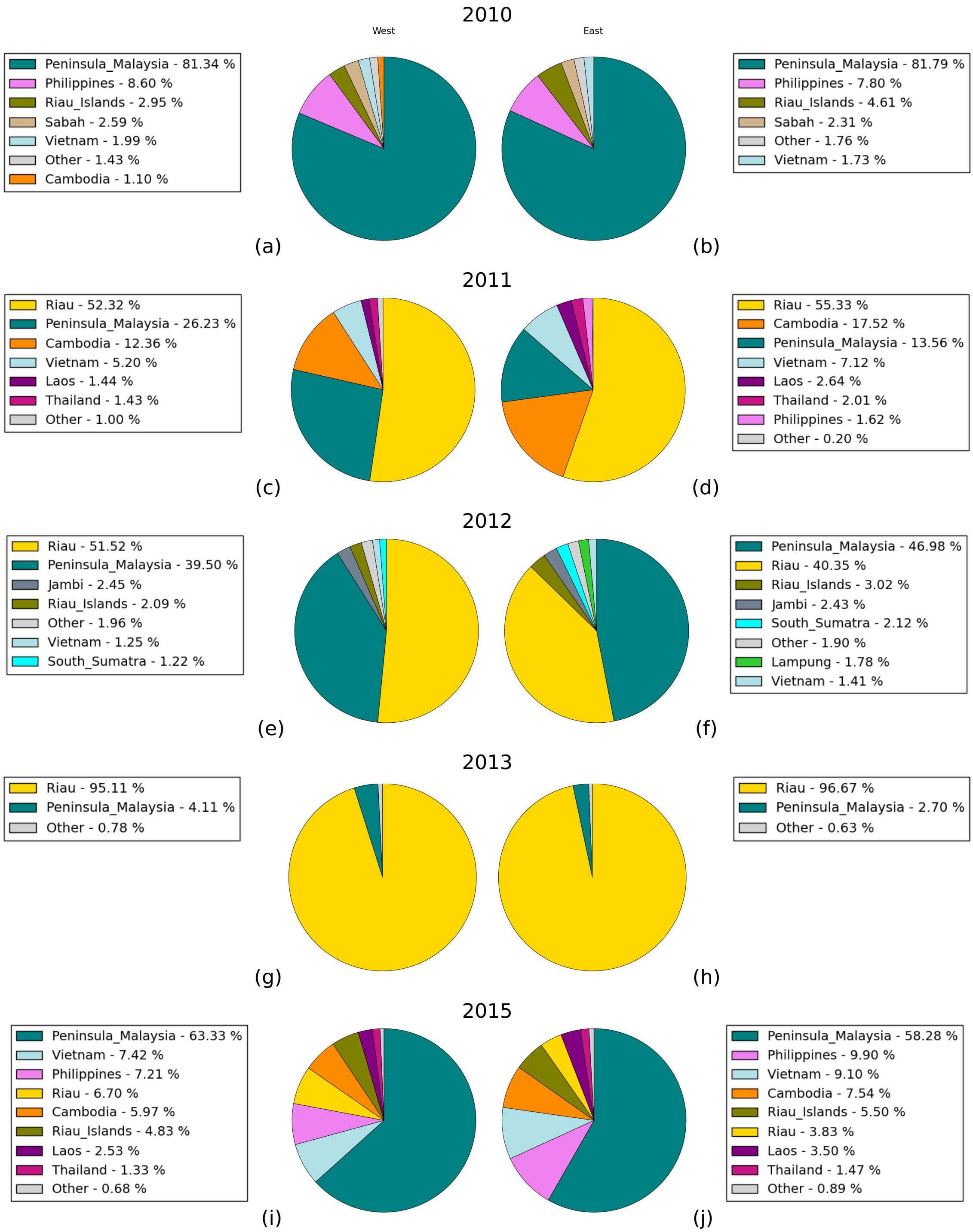

Figure A4. Attribution results for PM_{10} for FMA for years 2010 - 2013 and 2015: major contributing source regions for the western (NTU) (left) and eastern (TP) (right) monitoring stations. (For 2014 FMA, see Fig. 6.)