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**Impacts of air pollutants from fire and non-fire emissions on the regional
air quality in Southeast Asia**

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31 **Abstract**

32 Severe haze events in Southeast Asia caused by particulate pollution have become
33 more intense and frequent in recent years. Widespread biomass burning occurrences and
34 particulate pollutants from human activities other than biomass burning both play important
35 roles in degrading air quality in Southeast Asia. In this study, numerical simulations have
36 been conducted using the Weather Research and Forecasting (WRF) model coupled with a
37 chemistry component (WRF-Chem) to quantitatively examine the contributions of aerosols
38 emitted from fire (i.e., biomass burning) versus non-fire (including fossil fuel combustion,
39 and road dust, etc.) sources to the degradation of air quality and visibility over Southeast
40 Asia. These simulations cover a time period from 2002 to 2008 and are respectively driven
41 by emissions from: (a) fossil fuel burning only, (b) biomass burning only, and (c) both fossil
42 fuel and biomass burning. The model results reveal that 39% of observed low visibility days
43 can be explained by either fossil fuel burning or biomass burning emissions alone, a further
44 20% by fossil fuel burning alone, a further 8% by biomass burning alone, and a further 5%
45 by a combination of fossil fuel burning and biomass burning. Analysis of 24-h $PM_{2.5}$ Air
46 Quality Index (AQI) indicates that the case with coexisting fire and non-fire $PM_{2.5}$ can
47 substantially increase the chance of AQI being in the moderate or unhealthy pollution level
48 from 23% to 34%. The premature mortality among major Southeast Asian cities due to
49 degradation of air quality by particulate pollutants is estimated to increase from ~4110 per
50 year in 2002 to ~6540 per year in 2008. In addition, we demonstrate the importance of
51 certain missing non-fire anthropogenic aerosol sources including anthropogenic fugitive and
52 industrial dusts in causing urban air quality degradation. An experiment of using machine
53 learning algorithms to forecast the occurrence of haze events in Singapore is also explored
54 in this study. All these results suggest that besides minimizing biomass burning activities,

55 an effective air pollution mitigation policy for Southeast Asia needs to consider controlling
56 emissions from non-fire anthropogenic sources.

57 **1 Introduction**

58 Severe haze in Southeast Asia has attracted the attention of governments and the
59 general public in recent years due to its impact on local economy, air quality, and public
60 health (Miettinen et al., 2011; Kunii et al., 2002; Frankenberg et al., 2005; Crippa et al.,
61 2016). Widespread biomass burning activities are one of the major sources of haze events in
62 Southeast Asia. Our previous study demonstrated that biomass burning aerosols contributed
63 to up to 40-60% of haze events in the major cities of Southeast Asia during 2003-2014 (Lee
64 et al., 2017). On the other hand, biomass burning in Southeast Asia could impact climate
65 through emissions of both carbon dioxide (CO₂) (van der Werf et al., 2009) and particulate
66 matter – the latter has a substantial impact specifically on regional climate features including
67 the spatiotemporal distribution of precipitation and energy budgets (Wang, 2004, 2007).

68 Regarding the impact of biomass burning aerosols on public health, a recent study based
69 on the health model in the United States (U.S.) has estimated the number of deaths resulting
70 from black carbon (BC) to be more than 13,500 in 2010 (Li et al., 2016). Considering that
71 both the ambient concentration of particulate matter and overall population in Southeast
72 Asia are higher than those of the U.S., a worse scenario in the region could thus be
73 foreseeable. In fact, a few studies quantifying the consequences of aerosols on human
74 health in Southeast Asia have already suggested taking necessary measures to reduce
75 biomass burning and deforestation in order to prevent related public health issues (Marlier et
76 al., 2013). However, as important as biomass burning pollution may be, it is not the only
77 source of particulate pollution in Southeast Asia. Indeed, aerosols emitted from fossil fuel

78 burning alongside other non-biomass burning human activities, as indicated in our previous
79 study (Lee et al., 2017), also contribute significantly to air quality degradation.

80 Particulate pollutants from human activities other than biomass burning in Southeast
81 Asia include species both locally produced and brought in from neighboring regions by
82 long-range transport. Fossil fuel emissions in Southeast Asia have increased significantly in
83 recent years, especially in areas where energy demands are growing rapidly in response to
84 economic expansion and demographic trends (IEA, 2015). Therefore, advancing our
85 understanding of the respective contributions of aerosols from fire (i.e., biomass burning)
86 versus non-fire (including fossil fuel combustion, road and industrial dust, land use, and land
87 change, etc.) activities to air quality and visibility degradation has become an urgent task for
88 developing effective air pollution mitigation policies in Southeast Asia.

89 In this study, we aim to examine and quantify the impacts of fire and non-fire aerosols
90 on air quality and visibility degradation over Southeast Asia. Three numerical simulations
91 have been conducted using the Weather Research and Forecasting (WRF) model coupled
92 with a chemistry component (WRF-Chem), which is a sophisticated regional weather-
93 chemistry model, driven respectively by aerosol emissions from: (a) fossil fuel burning only,
94 (b) biomass burning only, and (c) both fossil fuel and biomass burning. By comparing the
95 results of these experiments, we examine the corresponding impacts of fossil fuel and
96 biomass burning emissions, both separately and combined, on the air quality and visibility
97 of the region. We also use available is-situ measurements to evaluate and correct model
98 results for providing a better base for further improvement of particularly emissions over the
99 region. Beyond the traditional process models such as WRF-Chem, we also experiment
100 using machine learning algorithms to identify suitable conditions for haze based on

101 historical data and hence to forecast the likelihood of the occurrence of such events in this
102 study.

103 We firstly describe methodologies adopted in the study, followed by the results and
104 findings from our assessment of the relative contributions of fire and non-fire aerosols in
105 degrading air quality and visibility over Southeast Asia. We then discuss the uncertainty of
106 current emission inventories alongside the results from an exploratory experiment of using
107 machine learning algorithms to forecast the occurrence of haze events in several major cities
108 in Southeast Asia. The last section summarizes and concludes our work.

109 **2 Methodology**

110 **2.1 Observational data**

111 **2.1.1 Surface visibility**

112 The observational data of surface visibility from the Global Surface Summary of the
113 Day (GSOD) (Smith et al., 2011) are used in our study to identify the days with low
114 visibility due to particulate pollution, i.e., haze events. The GSOD is derived from the
115 Integrated Surface Hourly (ISH) dataset and archived at the U.S. National Climatic Data
116 Center (NCDC). The daily visibility data are available from 1973 onward.

117 **2.1.2 Particulate matter (PM₁₀)**

118 The surface concentrations of particulate matter with sizes smaller than 10 μm (PM₁₀;
119 measured in $\mu\text{g m}^{-3}$) in Malaysia are derived from the Air Quality Index (AQI; named Air
120 Pollutant Index or API in Malaysia) records obtained from the website of Ministry of
121 Natural Resources and Environment, Department of Environment, Malaysia
122 (http://apims.doe.gov.my/public_v2/home.html). When PM₁₀ is reported as the primary
123 pollutant with a maximum pollutant index, the 24-h PM₁₀ concentrations are calculated from

124 AQI based on the equations in Table S1 (Malaysia, 2000). Data from 51 AQI observation
125 stations are available in Malaysia from October 2005 onward. AQI number is reported
126 twice daily (11 AM and 5 PM local time), and the data reported at 11 AM are used in this
127 study.

128 **2.1.3 Carbon monoxide (CO) and ozone (O₃)**

129 Surface mole fractions of CO and O₃ are measured by the World Meteorological
130 Organization (WMO) Global Atmosphere Watch (GAW) station in Bukit Kototabang,
131 which is located on the island of Sumatra, Indonesia. Hourly data are archived at the World
132 Data Center for Greenhouse Gases (WDCGG) under the GAW program
133 (<http://ds.data.jma.go.jp/gmd/wdcgg/>).

134 **2.1.4 Crustal matter and residual matter**

135 The Surface PARTiculate mAtter Network (SPARTAN) is a network of ground-based
136 measurements of fine particle concentrations (<http://spartan-network.weebly.com/>)
137 (Snider et al., 2016; Snider et al., 2015). Available data in the SPARTAN network include
138 hourly PM_{2.5} concentrations and certain compositional features (Table S2). Crustal matters
139 and residual matters, which are mainly organic components, from filtered PM_{2.5} samples are
140 used in this study to fill the gap in modeled PM_{2.5} created by the missing anthropogenic
141 aerosol in emission inventory (Philip et al., 2017). The four operational SPARTAN sites in
142 Southeast Asia are Bandung (Indonesia), Hanoi (Vietnam), Manila (Philippines), and
143 Singapore (Singapore). The chemical components of PM_{2.5} in each city are presented in Fig.
144 S1.

145 2.2 The model

146 WRF-Chem version 3.6.1 is used in this study to simulate trace gases and particulates
147 interactively with the meteorological fields using several treatments for photochemistry and
148 aerosols (Grell et al., 2005). We selected the Regional Acid Deposition Model version 2
149 (RADM2) photochemical mechanism (Stockwell et al., 1997) coupled with the Modal
150 Aerosol Dynamics Model for Europe (MADE), which includes the Secondary Organic
151 Aerosol Model (SORGAM) (Ackermann et al., 1998; Schell et al., 2001), to simulate
152 anthropogenic aerosols evolution in Southeast Asia. MADE/SORGAM uses a modal
153 approach (including Aiken, accumulation, and coarse modes) to represent the aerosol size
154 distribution, and predicts mass and number for each aerosol mode. The numerical
155 simulations are employed within a model domain with a horizontal resolution of 36 km,
156 including 432×148 horizontal grid points (Fig. 1), and 31 vertically staggered layers based
157 on a terrain-following pressure coordinate system. The Mellor-Yamada-Nakanishi-Niino
158 level 2.5 (MYNN) (Nakanishi and Niino, 2009) is chosen as the planetary boundary scheme
159 in this study. By using a vertical coordinate that is stretched to have higher resolutions
160 inside the planetary boundary layer, the model has about 4-5 vertical layers inside the
161 planetary boundary layer with a vertical resolution of ~ 30 m near the surface. The domain
162 covers an area from the Indian Ocean to the Western Pacific Ocean in order to capture the
163 Madden-Julian Oscillation (MJO) pattern. The time step is 180 seconds for advection and
164 physics calculation. The physics schemes in the simulations include Morrison (2 moments)
165 microphysics scheme (Morrison et al., 2009), RRTMG longwave and shortwave radiation
166 schemes (Mlawer et al., 1997; Iacono et al., 2008), Unified Noah land-surface scheme
167 (Tewari et al., 2004), and Grell-Freitas ensemble cumulus scheme (Grell and Freitas, 2014).
168 The initial and boundary meteorological conditions are taken from the U.S. National Center

169 for Environment Prediction FiNaL (NCEP-FNL) reanalysis data (National Centers for
170 Environmental Prediction, 2000), which has a spatial resolution of 1 degree and a temporal
171 resolution of 6 hours. Sea surface temperatures are updated every 6 hours in NCEP-FNL.
172 All simulations used a four-dimensional data assimilation (FDDA) method to nudge NCEP-
173 FNL temperature, water vapor, and zonal as well as meridional wind speeds above the
174 planetary boundary layer.

175 **2.3 Emission inventories**

176 The Regional Emission inventory in ASia (REAS) version 2.1 (Kurokawa et al., 2013)
177 is a regional emission inventory for Asia, including monthly emissions of most major air
178 pollutants, e.g., black carbon (BC), organic carbon (OC), sulfur dioxide (SO₂), nitrogen
179 dioxide (NO₂), and greenhouse gases between 2000 and 2008. The spatial resolution of
180 REAS is 0.25 × 0.25 degrees, covering East, Southeast, South, and Central Asia and the
181 Asian part of Russia (Russian Far East, Eastern and Western Siberia, and the Ural). The
182 area coverage of REAS is from 60°E to 160°E in longitude and from 10°S to 50°N in
183 latitude, which is smaller than our domain configuration. For this reason, we use the
184 Emissions Database for Global Atmospheric Research (EDGAR) version 3.2 (the year 2000
185 emission) (Olivier et al., 2005) and version 4.2 (the year 2005 emission)
186 (<http://edgar.jrc.ec.europa.eu>) to complement the emissions over areas outside REAS
187 coverage. The emission coverage of REAS and EDGAR in our simulated domain is
188 presented in Fig. 1. We have compared the modeled results using REAS versus EDGAR
189 emission inventories in a set of one-year paired simulations: the differences between these
190 two model runs are rather limited regarding aerosol-related variables (Table S3). After
191 considering high spatiotemporal resolution of REAS emission inventory and the comparison
192 results, we decided to use REAS in this study. In addition, a detailed comparison of REAS

193 with other emission inventories in Southeast Asia was also presented by Kurokawa et al.
194 (2013).

195 The Fire INventory from U.S. National Center for Atmospheric Research (NCAR)
196 version 1.5 (FINNv1.5) (Wiedinmyer et al., 2011) is also used in the study to provide fire-
197 based emissions. FINNv1.5 classifies burnings of extratropical forest, topical forest
198 (including peatland), savanna, and grassland. The daily data are available from 2002 to
199 2014 with a 1 km spatiotemporal resolution. FINNv1.5 emission inventory also includes the
200 major chemical species (e.g., BC, OC, SO₂, CO, and NO₂) from biomass burning. A
201 modified plume rise algorithm in WRF-Chem, specifically for tropical peat fire, is described
202 in Lee et al. (2017).

203 Compared with fossil fuel emissions, biomass burning emissions vary in space and time
204 (Fig. S2). However, regarding long-term impact, both emissions are important to regional
205 air quality in Southeast Asia (Table 1). BC from biomass burning emissions, for example,
206 has significant inter-annual and inter-seasonal variabilities due to the Southeast Asia
207 monsoon and the El Niño-Southern Oscillation (ENSO) (Lee et al., 2017; Reid et al., 2012),
208 but total BC emissions are equally contributed by fossil fuel and biomass burning sources
209 (Table 1).

210 **2.4 Numerical experiment design**

211 Three numerical simulations are proposed to investigate the impacts of fire and non-fire
212 aerosols on regional air quality and visibility in Southeast Asia. Among these three runs, the
213 fossil fuel emissions only (FF) simulation and the biomass burning emissions only (BB)
214 simulation are designed to assess the impact of stand-alone non-fire and fire aerosols,
215 respectively. The simulation combining both fossil fuel and biomass burning emissions
216 (FFBB) is to demonstrate the impacts of both types of aerosols; it is also closer to real world

217 case than the two other runs. Based on available years of emission inventories, each of these
218 runs lasts 7 years (i.e., from 2002 to 2008).

219 **2.5 Deriving “Low Visibility Day” (LVD) caused by particulate pollution**

220 According to Visscher (2013), a visibility reading lower than 10 km is considered a
221 moderate to heavy air pollution event by particulate matter. As in Lee et al. (2017), we
222 define a “low visibility day (LVD)” when the daily-mean surface visibility is lower or equal
223 to 10 km, not including misty and fog days. The modeled visibility is calculated based on
224 the extinction coefficient of the externally mixed aerosols, including BC, OC, sulfate (SO_4^{2-})
225 and nitrate (NO_3^-), as a function of particle size, by assuming a log-normal size distribution
226 of Aitken and accumulation modes. Note that all these calculations are computed for the
227 wavelength of 550 nm. To make the calculated visibility based on modeled aerosols better
228 match the reality, we also consider the hygroscopic growth of OC, sulfate, and nitrate in the
229 calculation based on the modeled relative humidity (Kiehl et al., 2000; Lee et al., 2017).

230 Our focus in this study is to first identify LVDs and then to determine whether fire or
231 non-fire aerosols alone, or in combination, could cause the occurrence of these LVDs. As a
232 reference, the observed low visibility days are identified and the annual frequency in every
233 year for a given city are also derived by using the GSOD visibility data. Then, the modeled
234 low visibility days are derived following the same procedure. Using these results and based
235 on the logical chart in Fig. 2, the major particulate source (FF, BB or FFBB) that caused the
236 occurrence of observed LVDs are determined. Here, Type 1 LVD represents the cases
237 where either fire or non-fire aerosols alone can cause the observed LVD to occur. Type 2
238 means that non-fire aerosols are the major contributor to the observed LVD. Type 3 means
239 that fire aerosols are the major contributor to the observed LVD. Type 4 represents the

240 cases where the observed LVD is induced by coexisting fire and non-fire aerosols. The
241 observed LVDs that the model cannot capture are classified as Type 5.

242 **2.6 Air Quality Index (AQI)**

243 The Air Quality Index is established mainly for the purpose to provide easily
244 understandable information about air pollution to the public. The original derivation of AQI
245 in the U.S. is based on six pollutants: particulate matter (PM₁₀), fine particulate matter
246 (PM_{2.5}), sulfur dioxide (SO₂), carbon monoxide (CO), ozone (O₃), and nitrogen dioxide
247 (NO₂). Each pollutant is scored on a scale extending from 0 through 500 based on the
248 corresponding breakpoints, and then the highest AQI value is reported to the public. In this
249 study, we focus on the AQI derived from modeled 24-h PM_{2.5} and 9-h O₃. Note that the
250 original AQI is derived by using 8-h O₃. Due to the 3-h output interval of simulated O₃, we
251 use the 9-h O₃ level instead in this study. An index I_p for pollutant p is calculated by using a
252 segmented linear function that relates pollutant concentration, C_p :

$$253 \quad I_p = \frac{I_{Hi} - I_{Lo}}{B_{Hi} - B_{Lo}} (C_p - B_{Lo}) + I_{Lo}, \quad (1)$$

254 where B_{Hi} is the upper breakpoint of C_p set category and B_{Lo} is the bottom breakpoint of C_p
255 sat category in Table S4. I_{Hi} and I_{Lo} are the AQI values corresponding to B_{Hi} and B_{Lo} ,
256 respectively. For example, when the 24-h PM_{2.5} concentration is 20 $\mu\text{g m}^{-3}$, B_{Hi} , B_{Lo} , I_{Hi} , and
257 I_{Lo} are 12, 1, 35.4, 51 and 100, respectively. Then, we selected 24-h PM_{2.5} and the maximum
258 9-h O₃ AQI value in one day to represent daily AQI for PM_{2.5} (AQI_(PM2.5)) and O₃ (AQI_(O3)),
259 respectively.

260 **2.7 Health Impact Assessment (HIA)**

261 Previous observations have revealed significantly higher PM_{2.5} concentrations in the
262 cities of Southeast Asia than those in America and Europe (WHO, 2016), implying that the

263 concentration-response functions (CRFs) derived from the latter places may not be directly
 264 applicable to Southeast Asia. In this study, we adapt CRFs in Gu and Yim (2016) to
 265 estimate the annual number of premature mortalities due to ambient PM_{2.5} concentration in
 266 the corresponding region. The relative risk (RR) of four causes of death, including chronic
 267 obstructive pulmonary disease, ischemic heart disease, lung cancer, and stroke, when
 268 compared with annual incident rate, have been assessed separately. Such risks are described
 269 by a log-linear relationship with the corresponding PM_{2.5} concentration level (Burnett et al.,
 270 2014). The basic form of RR formulas is provided as follows:

$$271 \quad RR = 1 + \alpha \cdot \left\{ 1 - \exp \left[-\beta (X_j - X_0)^\delta \right] \right\}, \quad (2)$$

272 where X_j and X_0 are the particulate pollutant concentrations ($\mu\text{g m}^{-3}$) in the target cities and
 273 the threshold value below which no additional risk is assumed to exist, respectively. Here
 274 we present the uncertainty range of threshold value between $5.8 \mu\text{g m}^{-3}$ and $8.8 \mu\text{g m}^{-3}$ in a
 275 triangular distribution, as suggested by the GBD 2010 project (Lim et al., 2013).
 276 Epidemiological results are not always available in Southeast Asia. To capture both
 277 climbing and flattening out phases of CRFs curves suitable for Southeast Asia region, we fit
 278 parameters α , β , and δ in CRFs by the epidemiological samples in the East Asian cities
 279 based on Gu and Yim (2016) for China, where PM_{2.5} concentration has a comparable level
 280 to that in Southeast Asia.

281 The form of integrated CRF is calculated by the following formula:

$$282 \quad E = \sum_j (RR_j - 1) / RR_j \cdot P_j \cdot f_j, \quad (3)$$

283 where P refers to the population in the researched cities from 2002 to 2008, retrieved from
 284 statistics in their respective countries (DSM, 2010; NSCB, 2009; NSOT, 2010; CSOM,

285 2010; GSOV, 2009; DSS, 2008, 2016; NISC, 2013; BPS, 2009). f denotes the baseline
286 incident rate above 30 years of age (WHO, 2017).

287 **3 Results**

288 **3.1 Model evaluation**

289 Multiple ground-based observations are used in this study to evaluate the model's
290 performance particularly in simulating aerosol and major gaseous chemical species such as
291 ozone and carbon monoxide. $PM_{2.5}$ observations in Southeast Asia are very limited. Even
292 in Singapore, observed $PM_{2.5}$ data are only available after 2014 for the general public and
293 research community to access. Therefore, PM_{10} concentrations derived from AQI in Kuala
294 Lumpur (Malaysia) are used to present the variation of particulate matter during haze and
295 non-haze seasons. Comparing with the observations, the model accurately predicted PM_{10}
296 concentration, especially during haze seasons (July to October) (Fig. 3a); however, it
297 produced a systematic negative bias of $20 \mu\text{g m}^{-3}$ in background PM_{10} concentration during
298 non-haze periods. This discrepancy between modeled and observed background PM_{10}
299 concentration could come from either the relatively coarse resolution of the model or the
300 underestimation of primary aerosol/ aerosol precursor emissions, or both. Philip et al. (2017)
301 indicated that most global emission inventories do not include anthropogenic fugitive,
302 combustion, and industrial dust (AFCID) from urban sources, typically including fly ash
303 from coal combustion and industrial processes (e.g. iron and steel production, cement
304 production), resuspension from paved and unpaved roads, mining, quarrying, and
305 agricultural operations, and road-residential-commercial construction. In their study, they
306 estimated a $2 - 16 \mu\text{g m}^{-3}$ increase in fine particulate matter ($PM_{2.5}$) concentration across
307 East and South Asia simply by including AFCID emission. We also find that the major
308 component of $PM_{2.5}$ particles from the filtered samples of SPARAN observational network

309 is residual materials, which are mainly organic matters (Snider et al., 2016) (Fig. S1). All of
310 these analyses show the incompleteness in the current emission inventories. In addition to
311 PM₁₀ data, we have also used observed surface visibility to evaluate model performance. As
312 mentioned in Sect. 2.5, the modeled visibility values are derived from the extinction
313 coefficient of the externally mixed aerosols and simulated fine particulate concentrations.
314 As shown in Fig. 4, the model correctly predicted about 40% observed low-visibility events
315 during the fire seasons, while 60% miss-captured low-visibility events are mainly due to the
316 missing AFCID. The details of this are discussed in Sect. 4. Additional uncertainty analysis
317 of modeled LVDs by using a method for dichotomous (yes or no LVDs) cases is presented
318 in Sect. S1 of the supplementary material. On the other hand, the model has overestimated
319 the visibility range for many cases with observed visibility lower than 7 km. Such a result is
320 likely due to the 36-km model resolution used in the study, which could be too coarse to
321 resolve the typical size of air plumes containing high concentration of fine particulate
322 matters. The detailed discussion of potential uncertainty factors of modeled visibility,
323 including meteorological datasets, fire emission inventories, and the model resolution
324 can be found in Lee et al. (2017).

325 The observed CO and O₃ levels from the only WMO GAW station in the region, Bukit
326 Kototabang, Indonesia (West Sumatra) are used to evaluate the model performance in
327 simulating gas phase chemistry. Fossil fuel and biomass combustions and biogenic
328 emissions are among the major sources of CO in the region, while O₃ production is mainly
329 from photochemical reactions of precursors such as nitrogen oxides, volatile organic
330 compounds, and CO, largely from anthropogenic emissions. Due to the geographic location,
331 the primary source of CO in Bukit Kototabang is from biomass burning, hence high CO
332 levels occur during fire seasons (Fig. 3b). The model accurately captured observed CO

333 levels during the simulation. Model simulated evolution of volume mixing ratio of O₃ also
334 matches observations very well, though with a positive bias of about 20 ppbv on average
335 (34.8 versus 13.4 ppbv) (Fig. 3c). We notice that NO_x emission is higher in REAS emission
336 inventory comparing with other emission inventories and studies (Kurokawa et al., 2013).
337 The boundary condition of WRF-Chem also sets the background surface ozone quite high
338 (30 ppbv). Both could lead to the overestimated background ozone in the model.

339 **3.2 Fire- and non-fire-caused LVDs in three selected cities**

340 Based on the logical chart shown in Fig. 2, we can use the modeled results to classify
341 observed LVDs into 5 types of events with different main aerosol sources. In Bangkok,
342 there are about 165 LVDs per year during 2002-2008 based on observations. Modeled
343 results suggest that about 60% of these LVDs can be brought by either fire or non-fire
344 aerosols (the sum of Type 1, Type 2, and Type 3 in Fig. 2; see Table 2). Generally
345 speaking, fire and non-fire aerosols contribute equally towards the haze events occurring in
346 Bangkok. A more interesting finding is that 11% of LVDs need a combination of both fire
347 and non-fire aerosols to occur (Type 4). This highlights the importance of fire aerosols in
348 worsening air quality of otherwise moderate haze conditions under the existing suspended
349 non-fire aerosols. Overall, the model missed about 29% of LVDs of Bangkok during the
350 simulation period.

351 Haze occurs slightly less frequently in Kuala Lumpur than Bangkok. There are about
352 104 LVDs per year in Kuala Lumpur during 2002-2008. Thirty-six percent of these LVDs
353 are caused by either fire or non-fire aerosols; while 15% of the LVDs need a combination of
354 both aerosol sources to form haze (Table 2). Our study shows that non-fire aerosols are
355 capable of causing of 28% of LVDs occurring in Kuala Lumpur, even in the absence of fire
356 aerosols. Once we include the impact of fire aerosols, the model can capture an additional

357 23% of LVDs, of which most are Type 4 case. Overall, fire and non-fire aerosols make
358 similar contributions to observed LVDs in Kuala Lumpur.

359 In Singapore, there are about 50 LVDs per year during 2002-2008. The contribution of
360 non-fire aerosols to LVDs is about 8%. Compared with the additional 25% of LVDs owing
361 to fire aerosols, the contribution of non-fire aerosols to LVDs is small in Singapore.
362 However, the model failed to capture a high percentage of LVD cases in both Kuala Lumpur
363 (49%) and Singapore (67%) (Type 5; see Table 2). As discussed in Sect. 3.1, missing
364 AFCID in the emission inventory could explain why the model failed to capture the LVDs
365 in these two sites. Further discussion is presented in Sect. 4.

366 **3.3 Fire- and non-fire-caused LVDs over the whole Southeast Asia**

367 By comparing the annual mean $PM_{2.5}$ concentration in 50 Association of Southeast
368 Asian Nations (ASEAN) cities between three simulations, we identify that there are 13
369 ASEAN cities receiving more than 70% $PM_{2.5}$ concentration from non-fire sources, while
370 other 10 ASEAN cities where fire aerosols are the major (more than 70%) component of
371 $PM_{2.5}$ (Fig. 5). Note that although fire aerosols are the major component of annual mean
372 $PM_{2.5}$ concentration in these latter 10 ASEAN cities, the influence period of fire aerosols
373 normally is only about 3 to 5 months. The rest of the ASEAN cities are essentially
374 influenced by coexisting fire and non-fire aerosols. Note that the sum of $PM_{2.5}$
375 concentrations in FF and BB is not necessarily equal to the $PM_{2.5}$ concentration in FFBB in
376 any given city due to non-linearity in modeled aerosol processes.

377 The annual mean LVDs among 50 ASEAN cities is 192 days during 2002-2008.
378 Applying the logical chart described in Fig. 2 to analyze cases of each of these ASEAN
379 cities, we find that by considering aerosols emitted from non-fire emissions alone, about
380 59% of observed LVDs can be explained, whereas considering fire aerosols adds an

381 additional 13% of LVDs. Conversely, by considering aerosols emitted from fire alone,
382 about 47% of observed LVDs can be explained, whereas adding non-fire aerosols adds an
383 additional 25% of LVDs. About 28% of observed LVDs remains unexplained. In general,
384 non-fire aerosols appear to be the major contributor to LVDs in these cities.

385 **3.4 Impacts of ozone and PM_{2.5} on air quality and human health**

386 Similar to PM_{2.5}, O₃ also brings public health besides air quality issues (Chen et al.,
387 2007). Previously in Sect. 3.1, we have discussed that the model systematically
388 overestimated O₃ volume mixing ratio by 20 ppbv comparing with observations.
389 Overestimated 9-h O₃ could lead to a mistakenly derived high AQI_(O₃). Nevertheless, the
390 relative differences of AQI_(O₃) between various model simulations can still provide useful
391 information of the relative contributions of fire and non-fire emissions, either alone or in
392 combination, on air quality and potentially human health.

393 We find that modeled 9-h O₃ in Bangkok from non-fire emissions (FF) alone triggered
394 19% of daily AQI_(O₃) to reach moderate and unhealthy pollution level during 2002-2008,
395 while fire emissions (BB) alone can only trigger 3% of such situations (Table 3). In
396 comparison, combining fire and non-fire emissions as derived from the simulation of FFBB
397 can cause 33% of daily AQI_(O₃) to reach moderate and unhealthy pollution levels. In Kuala
398 Lumpur and Singapore, O₃ is not the major source for air quality degradation, where fire or
399 non-fire emissions alone can seldom cause O₃ levels to reach even moderate pollution
400 levels. For example, in the FF simulation, only 5% of daily AQI_(O₃) readings in Kuala
401 Lumpur and 1% in Singapore reached moderate pollution levels. Again, the majority of the
402 high AQI_(O₃) cases result from combining fire and non-fire emissions (FFBB) (Table 3).
403 Overall, non-fire emissions alone only cause 6% of daily AQI_(O₃) to reach moderate

404 pollution levels in 50 ASEAN cities, whereas about 12% of moderate and unhealthy
405 pollution cases resulted from the combined effect of fire and non-fire emissions.

406 We find that in Southeast Asia, $PM_{2.5}$ actually plays a more important role than O_3 in
407 causing high AQI cases. In Bangkok, $PM_{2.5}$ resulted in 37% and 33% high daily $AQI_{(PM_{2.5})}$
408 cases in FF and BB simulation, respectively (Table 4). Among these, three times more cases
409 with daily $AQI_{(PM_{2.5})}$ reaching unhealthy levels can be attributed to $PM_{2.5}$ from BB than
410 those from FF (Table 4). However, the unhealthy levels caused by fire aerosols alone still
411 occur relatively infrequently in Bangkok, Kuala Lumpur, and Singapore. In Bangkok, a city
412 with an 8 million population, persistent aerosol emissions from non-fire sources, aided by
413 seasonal fire aerosols, cause almost two-thirds of daily air quality readings that reached
414 moderate or unhealthy pollution levels. Kuala Lumpur and Singapore also have 48% and
415 22% bad air quality days during 2002-2008, respectively (Table 4). Examining 24-h $PM_{2.5}$
416 $AQI_{(PM_{2.5})}$ among 50 ASEAN cities shows that non-fire aerosols alone contribute to
417 moderate to unhealthy pollution levels 2.6 times more often than fire aerosols alone (23%
418 versus 9%). Compared to the modeled results in FF, $PM_{2.5}$ in FFBB increases 10% more
419 bad air quality to moderate and unhealthy pollution level (Table 4). This result is consistent
420 with the findings in Sect. 3.3.

421 We have examined the health impacts due to $PM_{2.5}$ in 50 ASEAN cities using the
422 method described in Sect. 2.7 and the results show that the top three cities for premature
423 mortality caused by particulate pollution are Jakarta (Indonesia), Bangkok (Thailand), and
424 Hanoi (Vietnam) with 910, 1080, and 620 premature mortalities per year, respectively (Fig.
425 6). The premature mortality in Jakarta is mainly due to exposure to $PM_{2.5}$ particles emitted
426 from non-fire emissions (95%), the same situation as in Hanoi (80%). However, in
427 Bangkok, the health impact due to fire and non-fire aerosols are equally critical (Figs. S3

428 and S4). In general, owing to the increasing trend of non-fire emissions during the analysis
429 period, the premature mortalities due to PM_{2.5} emitted from non-fire sources have increased
430 with time in most ASEAN cities (Fig. S3). Besides this, higher fire aerosols levels in
431 Sumatra and Borneo in 2002, 2004 and 2006 also increase the number of premature
432 mortalities in cities such as Kuching, which are exposed to particulate matters from these
433 burning events (Figs. 6 and S4).

434 Additional discussion of the impact of fire and non-fire aerosols on regional climate is
435 presented in Sect. S2 of the supplementary.

436 **4 Impact of missing components in the emission inventories on** 437 **modeled results**

438 In this study, we have noticed that the simulated PM_{2.5} concentrations in Singapore are
439 often lower than the observations of the National Environment Agency of Singapore
440 (<https://data.gov.sg/dataset/air-pollutant-particulate-matter-pm2-5>) (6.1 µg m⁻³ versus 20.3
441 µg m⁻³ in annual mean during 2002-2008). Owing to the lower simulated PM_{2.5}
442 concentration in Singapore, the model could not capture many observed LVDs (Table 2) and
443 consequently underestimated AQI levels resulting from PM_{2.5}. As mentioned before, Philip
444 et al. (2017) have pointed out that global atmospheric models can produce a 2 - 16 µg m⁻³
445 underestimation in fine particulate mass concentration across East and South Asia and most
446 current global emission inventories indeed either do not include anthropogenic fugitive and
447 industrial dusts, or substantially underestimate the quantities of these emissions (Klimont et
448 al., 2016; Janssens-Maenhout et al., 2015). The fugitive dust sources, such as road and
449 construction dust, in most major cities in Southeast Asia are apparently not well represented
450 in the emission inventory used in our study. To correct these systematic underestimates, we

451 have used crustal matter and residual matter from SPARTAN PM_{2.5} measurements as the
452 reference to fill in the modeled PM_{2.5} for the missing anthropogenic aerosol components.
453 Excluding the high concentration samples during the fire haze events, the mean
454 concentration of crustal matter and residual matter is 25.8 μg m⁻³ in Hanoi, 10.4 μg m⁻³ in
455 Singapore, 18.1 μg m⁻³ in Bandung, and 9.2 μg m⁻³ in Manila. We then added these values
456 as additional anthropogenic aerosol components in modeled aerosol abundance to
457 recalculate modeled visibility and AQI_(PM_{2.5}). Table 5 shows the calculated percentage of
458 LVDs caused by various aerosol types in Fig. 2 before and after the above correction.

459 Adding the missing anthropogenic aerosol component based on in-situ measurement in
460 the modeled results can reproduce 98% of observed LVDs in Hanoi (an increase from 79%).
461 Because the missing anthropogenic aerosols are included in non-fire aerosols, LVDs in Type
462 1 and Type 2 are heavily weighted in the new result. The results also show the LVDs in
463 Hanoi are mainly caused by non-fire aerosols and the contribution of fire aerosols is
464 relatively small. Adding the missing anthropogenic aerosol components also reduced the
465 number of missing LVDs events from 67% to 20% in Singapore. Differing from Hanoi, not
466 only Type 2 LVDs but also Type 4 LVDs increased after introducing the missing
467 anthropogenic aerosols in Singapore, implying that the fire and non-fire aerosols are equally
468 important in causing LVDs there. After applying the correction, non-fire aerosols alone can
469 explain 30% LVDs while coexisting fire and non-fire aerosols can explain 40% LVDs in
470 Singapore (Table 5). Note that the mode of the distribution of observed visibility in
471 Singapore is around 11 km. Therefore, when fire occurs in the surrounding countries, even
472 a moderate addition to the aerosol abundance from fire can worsen visibility to reach a low
473 visibility condition (visibility < 10 km). Because of the poor data quality of observed
474 visibility in Bandung (only less than 10% observations are available), introducing the

475 missing anthropogenic aerosol components did not help to characterize the major aerosol
476 contribution. In Manila, the number of missed LVDs in the model reduced 35% while Type
477 2 and Type 4 LVDs increased 26% and 9%, respectively, after introducing the missing
478 anthropogenic aerosol components. Nevertheless, even after adding the missing
479 anthropogenic aerosols to the non-fire aerosol category, the model still missed 57% of LVDs
480 in Manila. This is mainly because the model did not capture many fire events in that area,
481 likely due to underestimation of fire emissions in the emission inventory.

482 Besides LVDs, the missing anthropogenic aerosols also substantially affect the modeled
483 $AQI_{(PM_{2.5})}$. Table 6 shows the frequency of various $AQI_{(PM_{2.5})}$ levels calculated respectively
484 with and without the missing anthropogenic aerosol components in Hanoi, Singapore,
485 Bandung, and Manila. After considering the missing anthropogenic aerosol components,
486 modeled air pollution levels in Hanoi and Bandung persistently reach the moderate or
487 unhealthy pollution levels. In Singapore, modeled frequency of moderate and unhealthy
488 cases also increase from 22% to 66%, and in Manila from 8% to 36%. Furthermore, the
489 number of premature mortalities in Singapore and Manila increases significantly from 0 to
490 230 and 130, respectively (Table 7). These results indicate the importance for models to
491 include anthropogenic fugitive and industrial dusts in order to capture low visibility events
492 in the region.

493 **5 Experiment in applying machine learning algorithms to** 494 **predict the occurrence of $PM_{2.5}$ caused LVDs**

495 Traditional physical models such as WRF-Chem are developed based on equations
496 describing fluid dynamics, physical processes, and chemical reactions to link these
497 processes on different scales and to predict consequences resulting from circulation and

498 physiochemical process evolutions. However, various parameterizations, and numerical as
499 well as input data errors can all lead to the uncertainty of model prediction. Specifically, for
500 the task of forecasting the occurrence of haze events (i.e., LVDs), using these models is
501 nearly impossible due to the lack of real-time emission estimates to drive aerosol chemical
502 and physical processes. On the other hand, machine learning algorithms permit
503 interpretation of large quantity of complex historical data based on computer analyses, and
504 this capacity of machine learning seems promising for us to derive suitable conditions for
505 hazes from historical data and hence to forecast the likelihood of the occurrence of such
506 events.

507 We hence experiment using the so-called supervised learning skill that trains or
508 optimizes a machine to produce the outcomes based on input data (or features) as close as
509 possible to known results or gaining an accuracy as high as possible. In our experiment, we
510 have applied 6 different machine learning algorithms, including Nearest Neighbors
511 (Pedregosa et al., 2011), Linear Support Vector Machine (SVM) (Schölkopf and Smola,
512 2002), SVM with Radial Basis Function Kernel (non-linear SVM) (Scholkopf et al., 1997;
513 Quinlan, 1986), Decision Tree (Quinlan, 1986), Random Forest (Breiman, 2001), and
514 Neural Network (Haykin et al., 2009), to reproduce past visibility patterns or to predict haze
515 occurrence. Through the supervised learning procedure, we have also examined the
516 importance of each input variable. These machine learning machines are trained for
517 predicting LVDs at three airports in Singapore reporting to the GSOD, i.e., Changi, Seletar,
518 and Paya Labar. All the input data or features are listed in Table S5. Data are available
519 from 2000 to 2015 at Changi and Paya Labar but only between 2004 and 2015 at Seletar.

520 We have used several different classifications in the training. The first one uses two
521 classes, corresponding to haze (visibility lower or equal to 10 km) and non-haze (visibility

522 higher than 10 km) events. Another applied 2-class classification uses 7 km instead of 10
523 km in identifying the haze events. In addition, a 3-class classification has also been tested,
524 which includes two haze classes: visibility lowers than 7 km and between 10 and 7 km,
525 respectively. The training-testing ratio is set to be 60:40.

526 In our study, the highest validation accuracy and F_1 -score (Powers, 2011) in any
527 algorithm appear in the machine for Changi site, while the difference in accuracy between
528 each algorithm is small (Figs. 7 and S5). However, the accuracy for all the algorithms at
529 Seletar and Paya Labar drops dramatically by about 20-30% in 2-class classification using
530 10-km visibility and 3-class classification. The reason for the best performances in Changi
531 is likely to be the least frequency of haze events at this site (account for only 10% of the
532 total LVDs), in comparison, 37% and 44% of haze events occurred at Paya Labar and
533 Seletar during the training time period, respectively. The machines also predict non-haze
534 events with higher accuracy than haze events at Changi. Using severe haze (visibility < 7
535 km) instead of moderate haze (visibility < 10 km) to label haze event can also increase
536 accuracy (over 80%). This could be due to the fact that severe haze events are primarily
537 caused by heavy biomass burnings, whose occurrence would be well captured in the satellite
538 hotspot input data.

539 Besides accuracy and F_1 -score analysis, we have also used the *feature importance*
540 function in the scikit-learn Random Forest package to measure the importance of various
541 features (i.e. Gini importance) (Pedregosa et al., 2011). The function takes array of features
542 and computes the normalized total reduction of the criterion brought by that feature. The
543 higher the value, the more important the feature is to the forecasting machine. We find that
544 the hotspot counts from three fire regions are ranked consistently among the top three most
545 important features for most machine learning predictions in all three classifications (Fig. 8;

546 Fig. S6 and S7). The values of importance of hotspot counts are higher than 0.15. Analysis
547 also suggests that “Month” is among the top five most important features in all machines,
548 followed by wind direction and relative humidity (Fig. 8), implying that besides fire hotspot,
549 seasonal monsoon wind patterns, wind-related weather conditions (i.e., SRV in Fig. 8) are
550 also important factors in forecasting the occurrence of haze events in Singapore. In
551 addition, relative humidity is a critical variable for visibility (i.e., growth of hygroscopic
552 particles can drastically enhance the light extinction). These results are consistent with
553 previous studies of haze events in Singapore (Reid et al., 2012; Lee et al., 2017).
554 Nevertheless, previous works by Reid et al. (2012) and Lee et al. (2017) also suggested the
555 relationships between fire hotspot appearance and certain weather phenomena particularly
556 precipitation. Therefore, we are surprised that precipitation in the fire regions does not
557 appear to be a significant feature for predicting Singapore haze compared with other features
558 in our current analysis.

559 **6 Summary**

560 We have quantified the impacts of fire (emitted from biomass burning) and non-fire
561 (emitted from anthropogenic sources other than biomass burning) aerosols on air quality and
562 visibility degradation over Southeast Asia, by using WRF-Chem in three scenarios driven
563 respectively by aerosol emissions from: (a) fossil fuel burning only, (b) biomass burning
564 only, and (c) both fossil fuel and biomass burning. These model results reveal that 39% of
565 observed low visibility days in 50 ASEAN cities can be explained by either fossil fuel
566 burning or biomass burning emissions alone when they coexist, a further 20% by fossil fuel
567 burning alone, a further 8% by biomass burning alone, and a further 5% by a combination of
568 fossil fuel burning and biomass burning. The remaining 28% of observed low visibility

569 days remain unexplained, likely due to emissions sources that have not been accounted for.
570 Our results show that owing to the economic growth in Southeast Asia, non-fire aerosols
571 have become the major reason to cause LVDs in most Southeast Asian cities. However, for
572 certain cities including Singapore, LVDs are likely caused by coexisting fire and non-fire
573 aerosols. Hence, both fire and non-fire emissions play important roles in visibility
574 degradation in Southeast Asia.

575 Furthermore, we have also used air quality index or AQI derived from modeled 9-h O_3
576 and 24-h $PM_{2.5}$ to analyze the air quality of 50 ASEAN cities. The results are consistent with
577 the visibility modeling and analysis, indicating that $PM_{2.5}$ particles, primarily those from
578 non-fire emissions, are the major reason behind high $AQI_{(PM_{2.5})}$ occurrence in these
579 Southeast Asian cities. In addition to non-fire $PM_{2.5}$ stand-alone cases, coexisting fire and
580 non-fire $PM_{2.5}$ jointly caused an increase of 11% in bad air quality events with moderate
581 polluted or unhealthy pollution levels (23% versus 34%). The premature mortality among
582 the analyzed ASEAN cities has increased from ~4110 in 2002 to ~6540 in 2008. Bangkok
583 (Thailand), Jakarta (Indonesia), and Hanoi (Vietnam) are the top three cities in our analysis
584 for premature mortality due to air pollution, with 1080, 910, and 620 premature mortalities
585 per year, respectively.

586 We find the reason behind the model's miss-capturing of 28% observed LVDs averaged
587 over 50 ASEAN cities is largely due to a lack of inclusion of anthropogenic fugitive and
588 industrial as well as road dust from urban sources in the emission inventories used in this
589 study. Using $PM_{2.5}$ chemical composition data from the SPARTAN stations in Hanoi,
590 Singapore, Bandung, and Manila to fill the missing aerosol components from these excluded
591 sources can drastically increase the captured LVDs by the model in these cities, for example,
592 by 47% in Singapore. The improvement in LVD prediction is especially substantial in non-

593 fire aerosols alone cases (Type 2; from 5% to 25%) and coexisting fire and non-fire aerosols
594 cases (Type 4; from 14% to 40%). Including the missing anthropogenic aerosols in modeled
595 results also increases the occurrence of cases with moderate and unhealthy air pollution
596 levels from 22% to 66% in Singapore. Our study clearly demonstrates the importance of
597 anthropogenic aerosols along with other fugitive industrial and urban sources in air quality
598 and visibility degradation in certain Southeast Asian cities such as Singapore.

599 We have also experimented using six different machine learning algorithms to predict
600 the occurrence of LVDs caused by $PM_{2.5}$. The effort is on forecasting hazes in three surface
601 visibility observation sites in Singapore. We find that the machine learning algorithms can
602 predict severe haze events (visibility < 7 km) with an accuracy greater than 80% in any of
603 these stations. On the other hand, the accuracy is found to be sensitive to the selection of
604 features, labelling of outcome, and forecast sites.

605 The current study extends our previous effort (Lee et al., 2017) by using a model
606 including a full chemistry and aerosol package instead of a smoke aerosol module without
607 chemistry. The added model capacity provides more complete quantitative description of
608 physiochemical processes that allow us to better analyze the contribution of fire versus non-
609 fire aerosols to the regional air quality and visibility degradation. Our results show that the
610 majority of the population in Southeast Asian cities are exposed to air pollution that can be
611 mostly attributed to non-fire aerosols. On the other hand, our analysis also suggests that for
612 certain cities such as Singapore, severe air pollution are likely caused by coexisting fire and
613 non-fire aerosols. All these further complicate the options for air pollution mitigation.

614 **7 Data availability**

615 FINNv1.5 emission data are publicly available from
616 <http://bai.acom.uar.edu/Data/fire/>. REAS and EDGAR emission data can be
617 downloaded from <https://www.nies.go.jp/REAS/> and
618 <http://edgar.jrc.ec.europa.eu/overview.php?v=42>, respectively. Malaysia API records
619 can be obtained from http://apims.doe.gov.my/public_v2/home.html. The observational
620 visibility from the GSOD can be downloaded from [https://data.noaa.gov/dataset/global-](https://data.noaa.gov/dataset/global-surface-summary-of-the-day-gsod)
621 [surface-summary-of-the-day-gsod](https://data.noaa.gov/dataset/global-surface-summary-of-the-day-gsod). CO and O₃ in WHO GAW station can be obtained
622 from <http://ds.data.jma.go.jp/gmd/wdcgg/>. Fine particle data from SPARTAN are
623 publicly available in <http://spartan-network.weebly.com/>. WRF-Chem simulated data are
624 available upon request from Hsiang-He Lee (hsiang-he@smart.mit.edu).

625

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830 Table 1. Mean annual emissions of BC, OC, SO₂, CO and NO₂ from biomass burning
 831 emission (BB) and fossil fuel burning emission (FF) in the simulated domain from
 832 2002 to 2008. Parentheses show the percentage of emission from fire and non-fire
 833 sources.
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Units: Tg/yr	BC	OC	SO₂	CO	NO₂
BB	0.4 (50%)	4.1 (73%)	0.4 (7%)	71.6 (64%)	2.6 (37%)
FF	0.4 (50%)	1.4 (27%)	5.8 (93%)	39.9 (36%)	4.3 (63%)

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837 Table 2. The contribution of fire aerosols (BB), non-fire aerosols (FF), or coexisting
 838 aerosols to low visibility days (LVDs) (based on the logic chart in Fig. 2) in Bangkok, Kuala
 839 Lumpur, Singapore, and among 50 Association of Southeast Asian Nations (ASEAN) cities
 840 during 2002-2008.

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	Bangkok	Kuala Lumpur	Singapore	50 ASEAN cities
FF∩BB (Type 1)	22±10%	12±5%	3±4%	39±5%
FF (Type 2)	19±5%	16±16%	5±4%	20±3%
BB (Type 3)	19±7%	8±5%	11±13%	8±2%
FF+BB (Type 4)	11±4%	15±6%	14±8%	5±1%
Missing (Type 5)	29±5%	49±26%	67±21%	28±2%

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844 Table 3. The frequency of occurrence of air pollution level in Bangkok, Kuala Lumpur,
 845 Singapore, and 50 Association of Southeast Asian Nations (ASEAN) cities derived using 9-
 846 h Ozone (O₃) volume mixing ratio in FF, BB, and FFBB during 2002-2008.
 847

Bangkok	AQI_(O3)	FF	BB	FFBB
Good	0-50	81±3%	97±1%	69±3%
Moderate	51-100	17±2%	3±1%	21±3%
Unhealthy	101-200	2±1%	0±0%	11±1%
Very				
Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%
Kuala Lumpur	AQI_(O3)	FF	BB	FFBB
Good	0-50	95±2%	100±1%	83±6%
Moderate	51-100	5±2%	0±1%	15±5%
Unhealthy	101-200	0±0%	0±0%	2±1%
Very				
Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%
Singapore	AQI_(O3)	FF	BB	FFBB
Good	0-50	99±1%	100±0%	94±3%
Moderate	51-100	1±1%	0±0%	5±2%
Unhealthy	101-200	0±0%	0±0%	1±1%
Very				
Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%
50 ASEAN cities	AQI_(O3)	FF	BB	FFBB
Good	0-50	94±1%	99±0%	88±2%
Moderate	51-100	6±1%	1±0%	10±2%
Unhealthy	101-200	0±0%	0±0%	2±0%
Very				
Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%

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Table 4. Same as Table 3 but using 24-h PM_{2.5} concentration.

Bangkok	AQI_(PM2.5)	FF	BB	FFBB
Good	0-50	63±6%	67±5%	38±2%
Moderate	51-100	34±5%	24±3%	45±3%
Unhealthy	101-200	3±2%	9±4%	17±4%
Very Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%
Kuala Lumpur	AQI_(PM2.5)	FF	BB	FFBB
Good	0-50	73±3%	78±8%	52±7%
Moderate	51-100	27±4%	18±6%	40±4%
Unhealthy	101-200	0±0%	4±3%	8±4%
Very Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%
Singapore	AQI_(PM2.5)	FF	BB	FFBB
Good	0-50	92±5%	92±4%	78±5%
Moderate	51-100	8±4%	6±2%	19±4%
Unhealthy	101-200	0±1%	1±2%	3±2%
Very Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%
50 ASEAN cities	AQI_(PM2.5)	FF	BB	FFBB
Good	0-50	77±1%	90±3%	66±3%
Moderate	51-100	19±1%	7±2%	26±2%
Unhealthy	101-200	4±0%	2±1%	8±2%
Very Unhealthy	201-300	0±0%	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%	0±0%

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853 Table 5. The old (without missing anthropogenic aerosol components) and new (with
 854 missing anthropogenic aerosol components in FF and FFBB) calculated percentage of
 855 observed low visibility days (LVDs), categorized according the type classification explained
 856 in Fig. 2.
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	Hanoi		Singapore		Bandung		Manila	
	old	new	old	new	old	new	old	new
FF∩BB (Type 1)	38±32%	40±31%	3±4%	5±7%	41±73%	41±74%	0±0%	1±1%
FF (Type 2)	34±8%	57±13%	5±4%	25±13%	8±19%	8±20%	3±3%	29±33%
BB (Type 3)	2±2%	0±0%	11±13%	9±10%	0±0%	0±0%	3±3%	2±3%
FF+BB (Type 4)	5±3%	1±1%	14±8%	40±19%	0±0%	0±0%	2±2%	11±3%
Missing (Type 5)	21±15%	2±4%	67±21%	20±9%	51±56%	51±57%	92±41%	57±16%

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861 Table 6. The frequency of various daily air pollution levels in Hanoi, Singapore, Bandung
 862 and Manila derived using 24-h $PM_{2.5}$ concentration with (new) and without (old) the missing
 863 anthropogenic aerosol components in FFBB during 2002-2008.

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Hanoi	AQI_(PM_{2.5})	old	new
Good	0-50	43±7%	0±0%
Moderate	51-100	46±3%	32±4%
Unhealthy	101-200	10±3%	67±4%
Very Unhealthy	201-300	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%
Singapore	AQI_(PM_{2.5})	old	new
Good	0-50	78±5%	33±8%
Moderate	51-100	19±4%	59±8%
Unhealthy	101-200	3±2%	7±3%
Very Unhealthy	201-300	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%
Bandung	AQI_(PM_{2.5})	old	new
Good	0-50	36±7%	0±0%
Moderate	51-100	58±5%	52±8%
Unhealthy	101-200	6±3%	48±8%
Very Unhealthy	201-300	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%
Manila	AQI_(PM_{2.5})	old	new
Good	0-50	92±4%	64±5%
Moderate	51-100	7±3%	34±5%
Unhealthy	101-200	1±1%	2±1%
Very Unhealthy	201-300	0±0%	0±0%
Hazardous	301-400	0±0%	0±0%
Hazardous	401-500	0±0%	0±0%

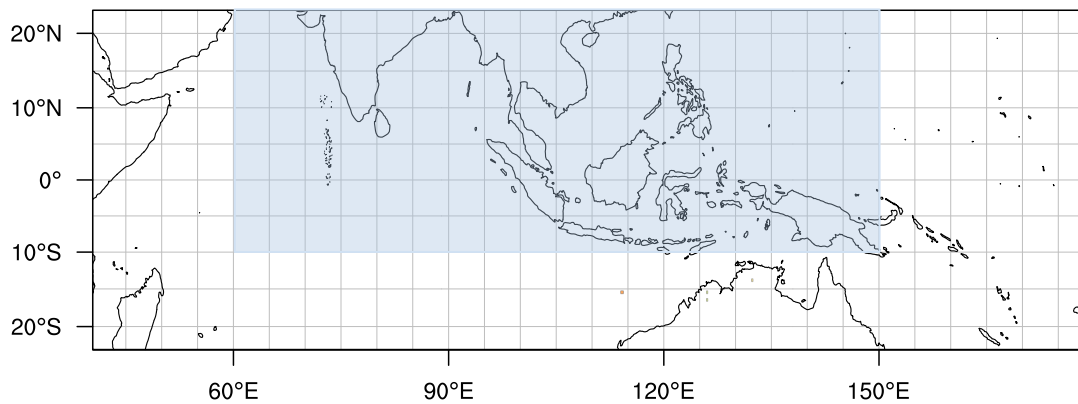
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866 Table 7. Updated PM_{2.5} concentration ($\mu\text{g m}^{-3}$) and premature mortality (95% confidence
867 intervals) in Hanoi, Singapore, Bandung and Manila with missing anthropogenic aerosol
868 components.
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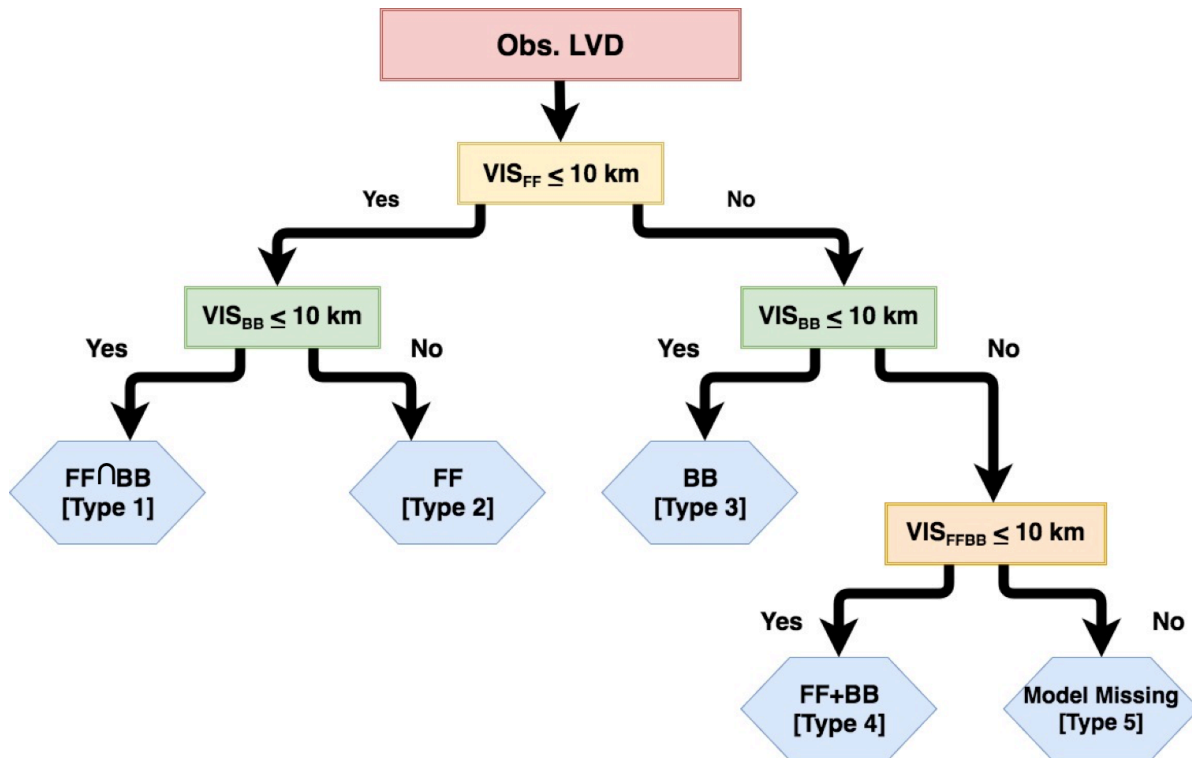
City	PM _{2.5} ($\mu\text{g m}^{-3}$)	Premature mortality
Hanoi	41.07	670 (210-1180)
Singapore	16.43	230 (20-550)
Bandung	33.18	260 (70-480)
Manila	12.38	130 (10-260)

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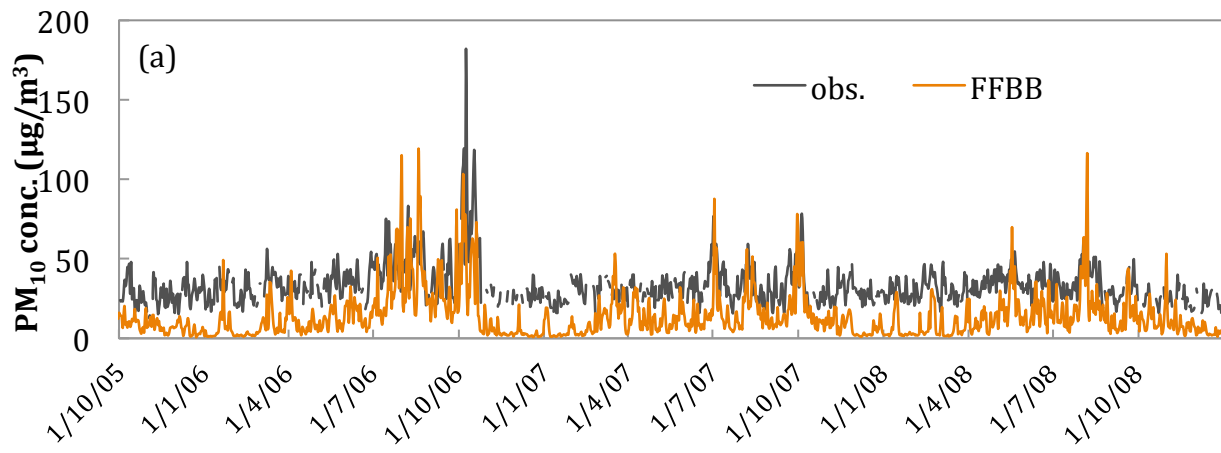


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875 Figure 1. Model domain used for simulations. The blue color region indicates the fossil fuel
876 emission coverage from the Regional Emission inventory in ASia (REAS). The rest of the
877 domain uses the fossil fuel emission from the Emissions Database for Global Atmospheric
878 Research (EDGAR).
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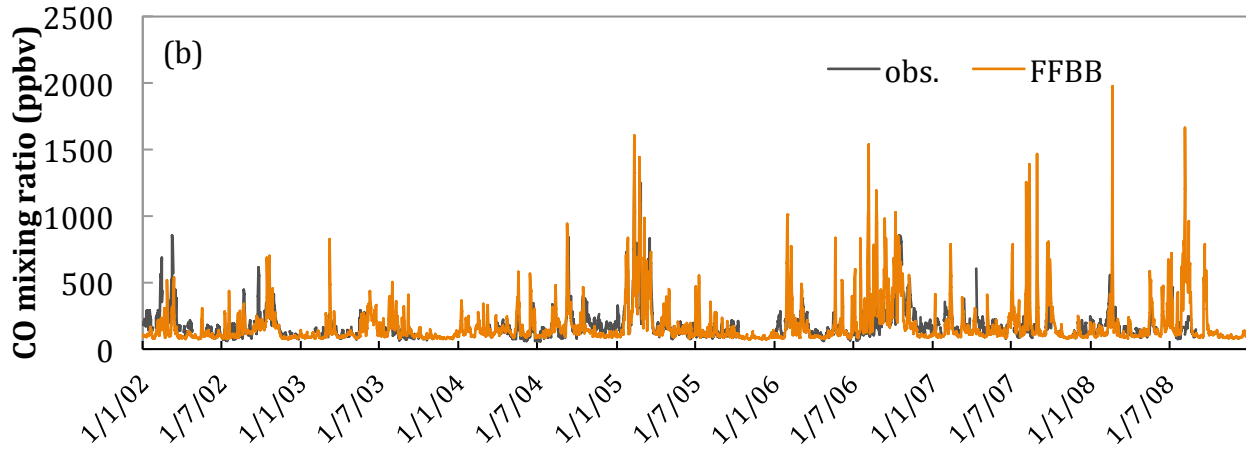


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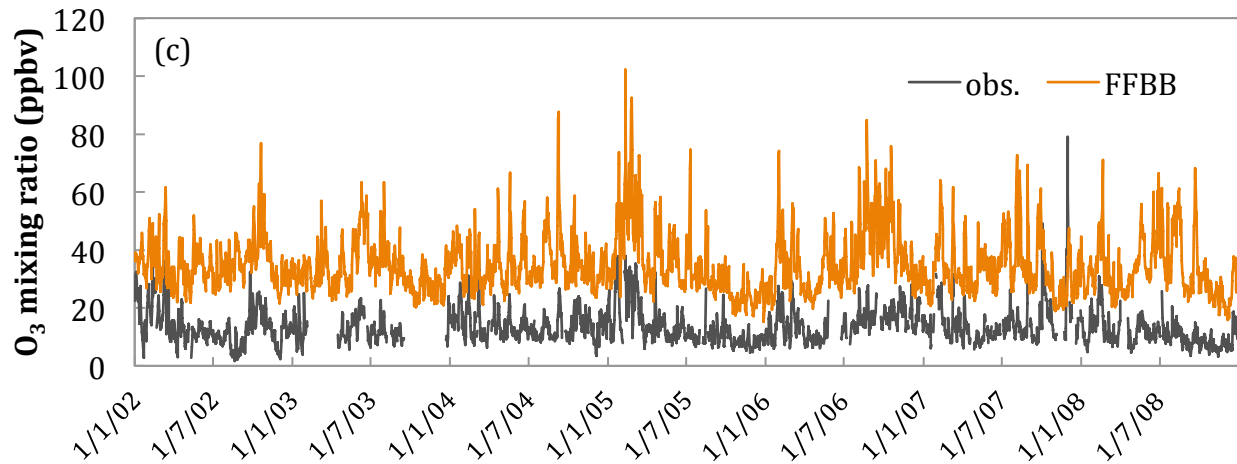
Figure 2. Logical chart for fire (BB), non-fire (FF), or coexisting fire and non-fire (FF+BB) aerosols caused Low Visibility Day (LVD). “Obs. LVD” is an identified low visibility day from observation. Then, the modeled visibility from FF (VIS_{FF}), BB (VIS_{BB}), and FFBB (VIS_{FFBB}) are used to classify observed LVD into 5 types of LVD. Type 1 LVD represents the cases where either fire or non-fire aerosols alone can cause the observed LVD to occur. Type 2 means that non-fire aerosols are the major contributor to the observed LVD. Type 3 means that fire aerosols are the major contributor to the observed LVD. Type 4 represents the cases where the observed LVD is induced by coexisting fire and non-fire aerosols. The observed LVDs that the model cannot capture are classified as Type 5.



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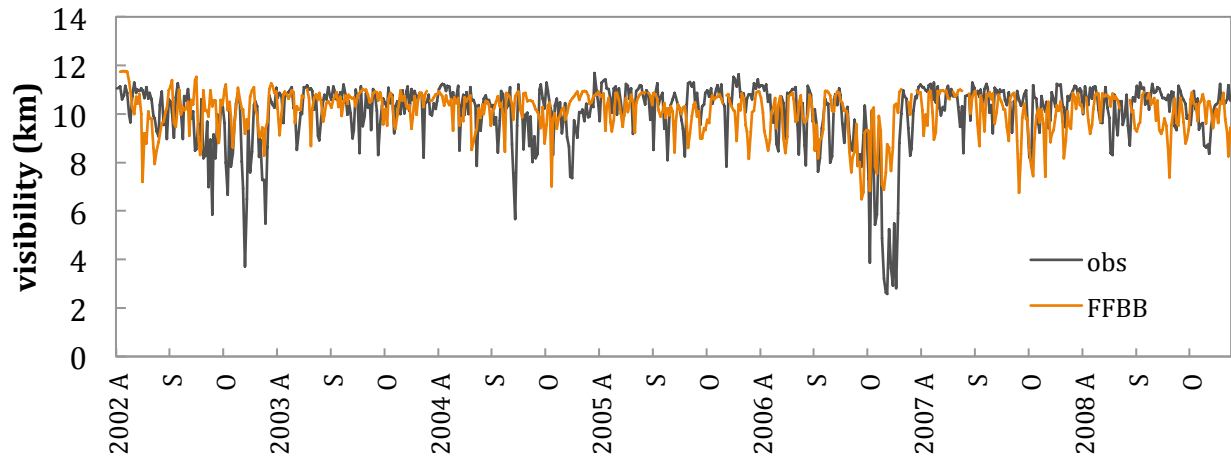
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Figure 3. (a) Time series of daily surface PM_{10} ($\mu\text{g m}^{-3}$; AQI derived) from the ground-based observations (black line) and FFBB-simulated results (orange line) in Kuala Lumpur, Malaysia during October 2005 – December 2008. (b) Time series of daily surface CO mixing ratio (ppbv) from the ground-based observations (black line) and FFBB-simulated results (orange line) in Bukit Kototabang, Indonesia during 2002 – 2008. (c) Same as (b) but surface O_3 .



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Figure 4. Comparison of daily visibility between GSOD observation (black line) and FFBB-simulated results (orange line) in Singapore during the fire seasons from 2002 to 2008. A, S, and O in the *x* axis indicates August, September, and October.

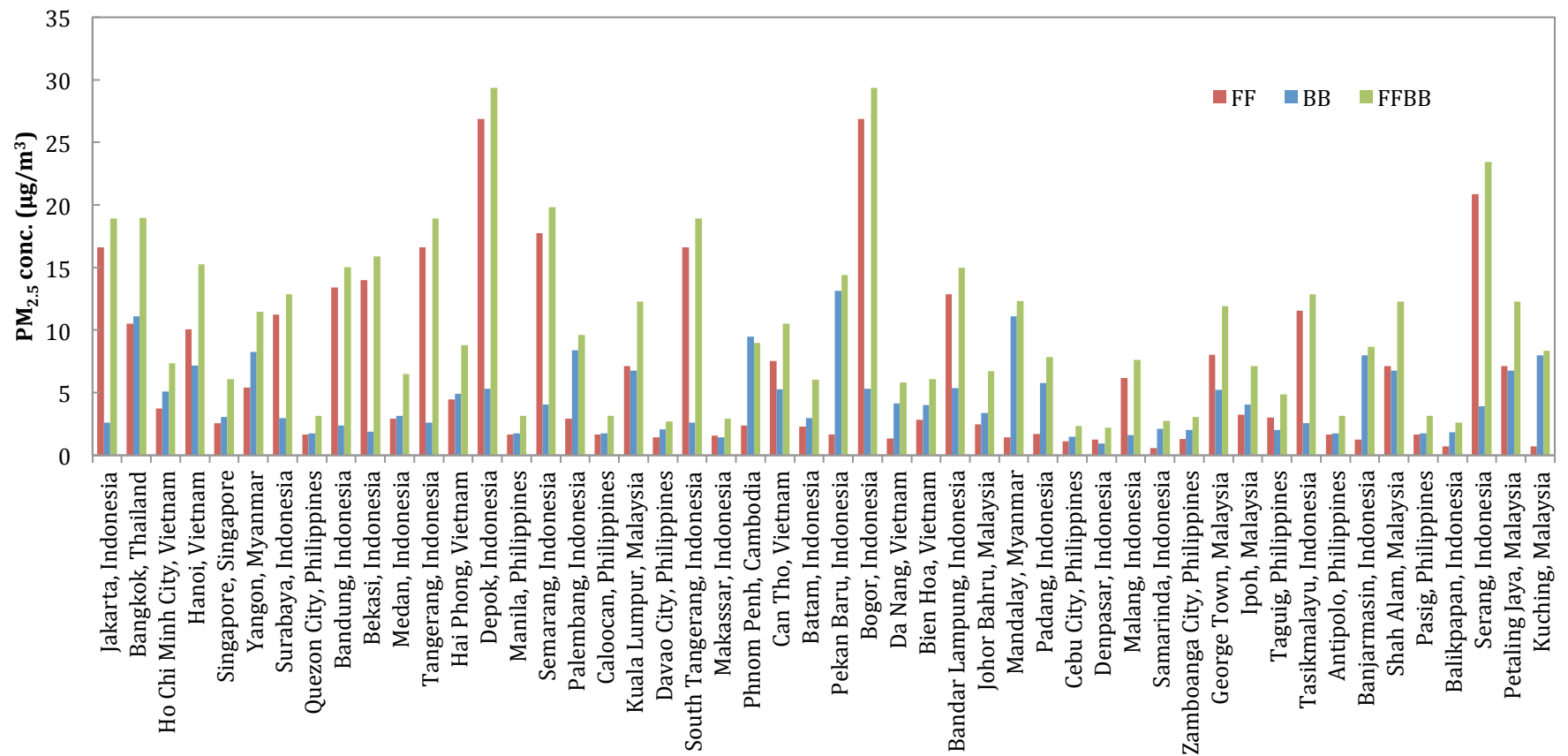


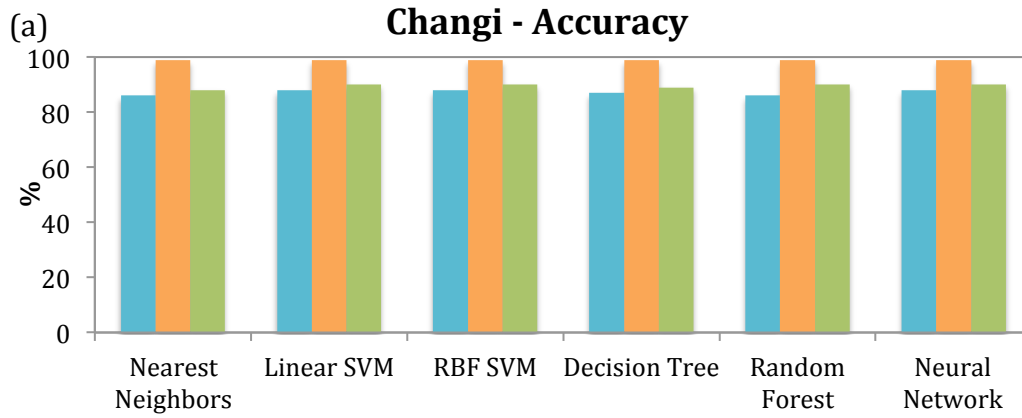
Figure 5. The annual mean simulated PM_{2.5} concentration ($\mu\text{g m}^{-3}$) in 50 Association of Southeast Asian Nations (ASEAN) cities, derived from FF (red), BB (blue), and FFBB (green) simulations and averaged over the period 2002-2008.

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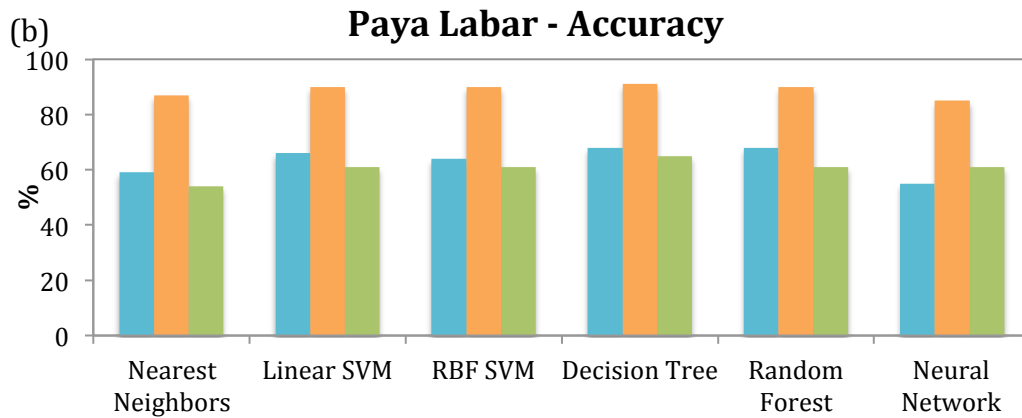
Cities	Country	2002	2003	2004	2005	2006	2007	2008
Jakarta	Indonesia	850 (150-1660)	830 (130-1650)	900 (160-1750)	950 (180-1820)	910 (150-1790)	960 (170-1870)	970 (170-1900)
Bangkok	Thailand	850 (90-1950)	1010 (130-2230)	1030 (130-2280)	1170 (180-2530)	1120 (150-2480)	1180 (160-2590)	1170 (150-2600)
Ho Chi Minh City	Vietnam	0 (0-0)	0 (0-0)	830 (80-1750)	610 (0-1590)	0 (0-1130)	230 (0-1580)	0 (0-1530)
Hanoi	Vietnam	420 (40-880)	520 (80-1020)	540 (80-1060)	560 (90-1100)	570 (80-1120)	610 (100-1190)	1150 (190-2250)
Singapore	Singapore	0 (0-0)	0 (0-0)	0 (0-260)	0 (0-190)	0 (0-290)	0 (0-290)	0 (0-0)
Yangon	Myanmar	0 (0-380)	280 (20-630)	350 (30-730)	330 (30-710)	280 (20-640)	400 (40-820)	330 (20-730)
Surabaya	Indonesia	220 (30-440)	210 (20-430)	230 (30-460)	230 (30-470)	230 (30-470)	240 (30-480)	230 (20-480)
Quezon City	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Bandung	Indonesia	200 (30-400)	200 (30-400)	210 (30-420)	230 (40-450)	200 (20-410)	220 (30-450)	220 (30-440)
Bekasi	Indonesia	150 (20-310)	160 (20-320)	180 (30-350)	190 (30-380)	190 (30-380)	210 (30-410)	210 (30-420)
Medan	Indonesia	0 (0-0)	0 (0-0)	0 (0-230)	10 (0-250)	0 (0-240)	0 (0-160)	0 (0-160)
Tangerang	Indonesia	120 (20-240)	120 (20-250)	140 (20-270)	150 (30-290)	150 (20-300)	170 (30-320)	170 (30-340)
Hai Phong	Vietnam	0 (0-0)	210 (10-480)	200 (0-480)	230 (10-510)	200 (0-500)	270 (30-580)	280 (30-590)
Depok	Indonesia	130 (30-230)	130 (30-250)	150 (30-270)	160 (40-300)	160 (40-310)	180 (40-330)	190 (40-350)
Manila	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Semarang	Indonesia	120 (20-240)	120 (20-240)	140 (30-260)	140 (30-280)	140 (30-280)	150 (30-290)	150 (30-300)
Palembang	Indonesia	100 (10-210)	0 (0-0)	100 (10-210)	0 (0-10)	150 (30-280)	0 (0-0)	0 (0-0)
Caloocan	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Kuala Lumpur	Malaysia	130 (10-290)	100 (0-260)	160 (20-340)	170 (20-360)	170 (20-360)	150 (10-340)	150 (10-340)
Davao City	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
South Tangerang	Indonesia	130 (20-250)	120 (20-240)	130 (20-250)	140 (30-260)	130 (20-250)	130 (20-260)	130 (20-260)
Makassar	Indonesia	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Phnom Penh	Cambodia	0 (0-0)	0 (0-40)	40 (10-90)	30 (0-80)	30 (0-80)	40 (0-90)	40 (0-90)
Can Tho	Vietnam	60 (0-270)	140 (10-310)	180 (20-370)	170 (20-360)	160 (10-350)	180 (20-380)	180 (20-380)
Batam	Indonesia	0 (0-0)	0 (0-0)	0 (0-50)	0 (0-60)	10 (0-80)	0 (0-90)	0 (0-0)
Pekan Baru	Indonesia	20 (0-80)	0 (0-40)	60 (10-120)	80 (20-150)	80 (10-150)	70 (10-140)	70 (10-150)
Bogor	Indonesia	100 (20-180)	100 (20-180)	100 (20-190)	110 (30-200)	100 (20-200)	110 (30-200)	110 (30-210)
Da Nang	Vietnam	0 (0-0)	0 (0-0)	90 (0-210)	0 (0-180)	0 (0-0)	0 (0-170)	0 (0-100)
Bien Hoa	Vietnam	0 (0-0)	0 (0-0)	60 (0-150)	0 (0-130)	0 (0-0)	0 (0-70)	0 (0-100)
Bandar Lampung	Indonesia	70 (10-140)	70 (10-140)	70 (10-140)	70 (10-140)	80 (10-160)	70 (10-150)	80 (10-160)
Johor Bahru	Malaysia	0 (0-0)	0 (0-0)	20 (0-170)	0 (0-160)	0 (0-200)	30 (0-190)	0 (0-70)
Mandalay	Myanmar	0 (0-0)	290 (20-610)	330 (30-670)	300 (30-640)	300 (30-650)	360 (40-740)	340 (30-710)
Padang	Indonesia	0 (0-0)	0 (0-0)	0 (0-60)	10 (0-90)	10 (10-130)	40 (0-110)	30 (0-100)
Cebu City	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Denpasar	Indonesia	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Malang	Indonesia	30 (0-100)	0 (0-50)	30 (0-100)	20 (0-100)	10 (0-100)	10 (0-100)	0 (0-100)
Samarinda	Indonesia	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Zamboanga City	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
George Town	Malaysia	110 (10-250)	100 (10-240)	140 (10-290)	140 (10-290)	120 (10-270)	120 (10-260)	120 (10-270)
Ipoh	Malaysia	0 (0-0)	0 (0-0)	50 (0-120)	50 (0-120)	0 (0-90)	0 (0-50)	0 (0-90)
Taguig	Philippines	0 (0-0)	0 (0-0)	0 (0-60)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Tasikmalayu	Indonesia	30 (0-70)	30 (0-70)	40 (0-80)	40 (0-90)	40 (0-80)	50 (10-90)	50 (10-100)
Antipolo	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Banjarmasin	Indonesia	50 (10-100)	0 (0-0)	50 (10-110)	0 (0-0)	60 (10-110)	0 (0-0)	0 (0-0)
Shah Alam	Malaysia	60 (0-130)	40 (0-110)	70 (10-150)	70 (10-150)	70 (10-150)	60 (0-140)	60 (0-130)
Pasig	Philippines	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Balikpapan	Indonesia	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)	0 (0-0)
Serang	Indonesia	50 (10-90)	50 (10-90)	50 (10-90)	50 (10-90)	50 (10-90)	50 (10-90)	50 (10-90)
Petaling Jaya	Malaysia	60 (0-120)	40 (0-110)	70 (10-140)	70 (10-140)	70 (10-140)	60 (0-130)	60 (0-130)
Kuching	Malaysia	50 (0-100)	0 (0-0)	50 (0-110)	0 (0-0)	60 (10-130)	0 (0-60)	0 (0-0)

914 Figure 6. Premature mortality in different years from 2002 to 2008 and cities in Association of
915 Southeast Asian Nations (ASEAN) due to exposures $PM_{2.5}$ in FFBB (95% confidence intervals).
916 Colors from green to red represent relative number scale.

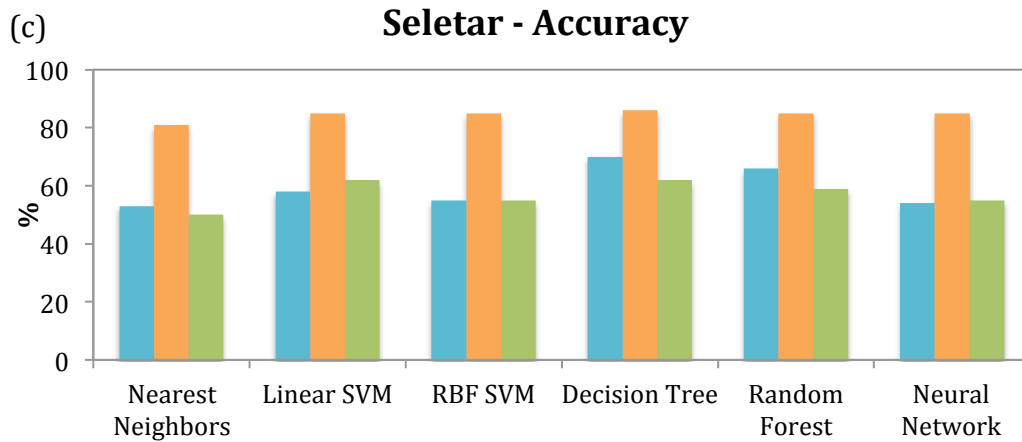
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■ 2-class (10 km) ■ 2-class (7 km) ■ 3-class

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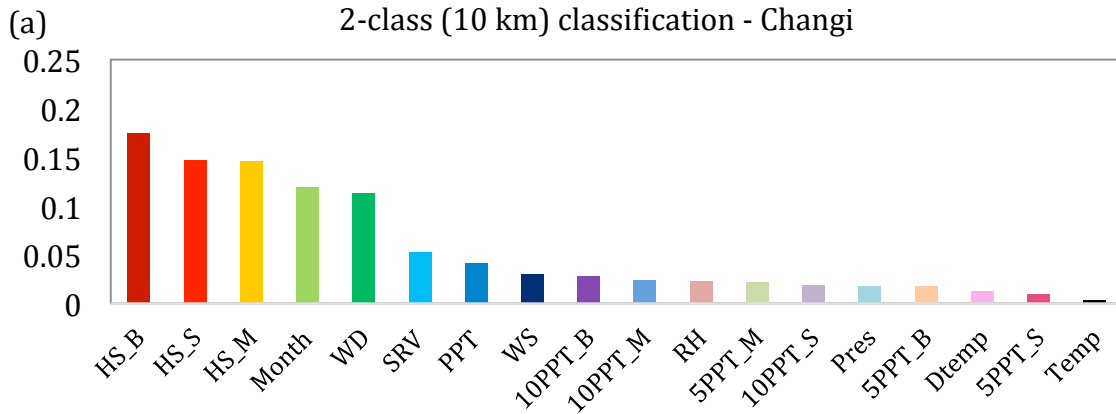
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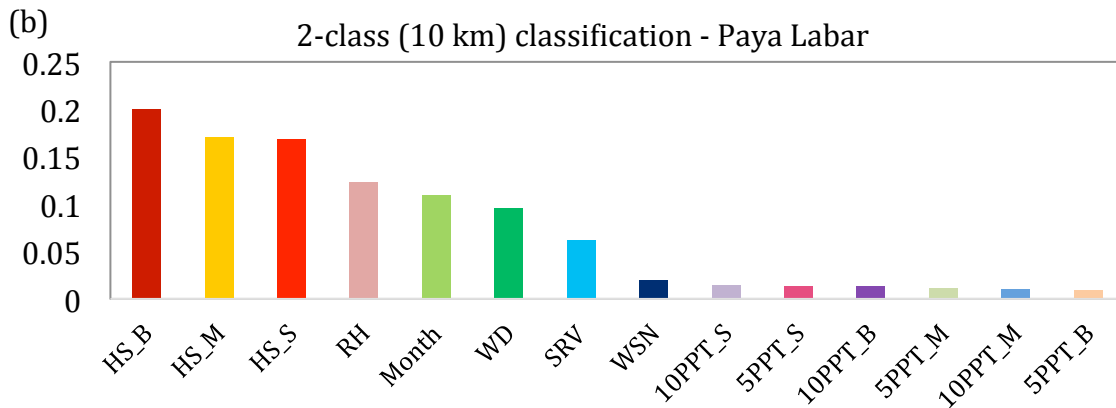
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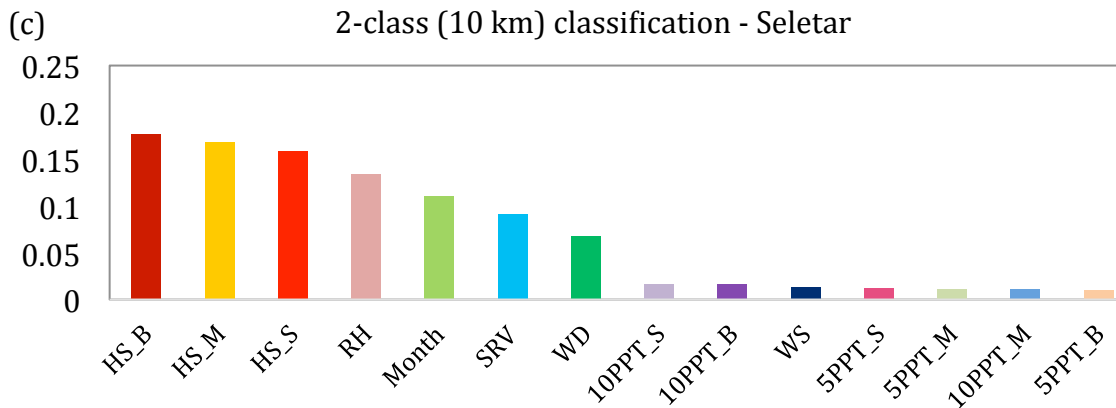
Figure 7. The testing accuracy in 6 machine learning algorithms for two 2-class (7 km or 10 km visibility as a breakpoint) and one 3-class classifications haze prediction in (a) Changi, (b) Paya Labar, and (c) Seletar.



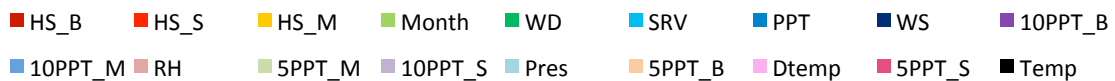
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Figure 8. Feature importance by using 2-class classification Random Forest algorithm in (a) Changi, (b) Paya Labar, and (c) Seletar. Desired outputs, haze versus non-haze events, are defined by using visibility 10 km as a breakpoint. Full name of each input feature are listed in Table S5.