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Keywords

philippines, long-range, radiative, forcing, effect, transport, aerosols, asia, its, northern, southeast, direct, east

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Abstract

Two elevated fine particulate mass concentration events were observed in a span of a week in northern Philippines on March of 2017. Results from chemical characterization, lidar observation, and model simulations of particulate matter show the high aerosol concentration events to be caused by long range transport (LRT) of anthropogenic pollutants from northern East Asia at the surface and biomass burning emission from Indochina aloft. In this study, we investigated the transport path of these LRT aerosols and estimated their direct radiative forcing. A strong Siberian high and a confluent flow induced by a continental high over the main Asian continent and a cyclonic circulation over the south of Japan produced a strong northerly wind

that carried pollutants from northern East Asia to northern Philippines. At the same time, strong westerlies 2-4 km aloft carried biomass burning emissions from Indochina, which also had an impact on ground concentration. Mass extinction efficiency of LRT aerosols was estimated to be in the range of 2.06 to 6.44 m^2g^{-1} with a mean value of $4.32 \pm 1.32 \text{ m}^2\text{g}^{-1}$. In addition, we calculated the direct radiative forcing effect under clear sky condition and found that the transported aerosols had a mean net negative forcing (cooling) effect of $-50.80 \pm 12.38 \text{ Wm}^{-2}$ and $-11.98 \pm 3.96 \text{ Wm}^{-2}$ on surface and at the top of the atmosphere, respectively. Such pollutant transport during wintertime cold surges had been shown to reduce local surface pollutants in northern China. Consequently, its impacts shift to downwind regions as far as the Philippines. As winter cold surge frequency increases due to the warming arctic, more LRT events may be expected as a result of climate change.

Keywords: aerosol, long range transport, radiative forcing

1. Introduction

Exposure to fine particulates is a major global health concern that affects millions annually. In the recent past decades, pollutant emissions in Asia have surpassed that of western developed countries (Akimoto, 2003; Smith et al., 2011). As a result, air pollution is linked to premature mortality across countries in Asia (Lelieveld et al., 2015; Organization et al., 2015; Zhang et al., 2017). Other than the serious health effects it poses, particulate matter also undergo complex interaction in the atmosphere that can have considerable impacts on Earth's radiative forcing (Twomey, 1977; Ackerman et al., 2000).

10 Combined with the effects of climate change, its potential impacts are still
 11 not well fully understood (Stocker et al., 2013). Aerosols have average res-
 12 idence times in the order of hours to weeks (Carter et al., 2005; Wu et al.,
 13 2007; Wang et al., 2011). Ambient concentrations of aerosols depend on
 14 distance from source location, as well as transport and deposition processes
 15 (Thomaidis et al., 2003). As a result, aerosol concentrations have large spa-
 16 tial variability. Consequently, this uneven distribution leads to high spatial
 17 and temporal variability in its radiative forcing. This may result to differ-
 18 ent climatic effects occurring in separate regions (Akimoto, 2003). For this
 19 reason, aerosol-cloud-climate interaction has been the focus of several past
 20 and future field campaigns in different specific regions of East and Southeast
 21 Asia, including the tropical western North Pacific (WNP).

22 Certain weather patterns that serve as long range transport (LRT) path-
 23 ways of aerosols extend their impacts from local to regional and global scales.
 24 Therefore, understanding the role of meteorology in air pollution studies is
 25 of utmost importance. Several air pollution outflow transport mechanisms
 26 across Asian regions have been reported in past literatures (Stohl et al., 2002;
 27 Akimoto, 2003; Yienger et al., 2000; Chakraborty et al., 2015). Elevated con-
 28 centration of pollutant gases and particulates are reported in countries such
 29 as Taiwan (Lin et al., 2009; Lee et al., 2011), Korea, and Japan transported
 30 from the main Asian continent (Aikawa et al., 2010; Oh et al., 2015; Kashima
 31 et al., 2016; Kim et al., 2017) due to these weather patterns (Eckhardt et al.,
 32 2004; Itahashi et al., 2010).

33 Southeast Asia is one of the most vulnerable regions to impacts of cli-
 34 mate change (Yusuf and Francisco, 2009). Biomass burning for the purpose

35 of land clearing is ubiquitous in this region (Betha et al., 2014). In 2002,
 36 the Association of Southeast Asian Nation (ASEAN) established a coordi-
 37 nating center for transboundary haze pollution to manage the impact of land
 38 and forest fires in the region ([http://haze.asean.org/asean-agreement-on-](http://haze.asean.org/asean-agreement-on-transboundary-haze-pollution/)
 39 [transboundary-haze-pollution/](http://haze.asean.org/asean-agreement-on-transboundary-haze-pollution/)). Nevertheless, haze from forest fires in SEA
 40 countries such as Myanmar, Lao, Cambodia, Thailand, Vietnam, Malaysia
 41 or Indonesia continue to have significant impacts. In springtime when open
 42 burning is at its peak, an eastward flow aloft from Indochina transports par-
 43 ticulates associated with burning, as well as CO and O₃ to East Asia (Lin
 44 et al., 2013). Conversely, strong northerly wind along the East China Sea
 45 (ECS) moves dust and anthropogenic pollutants towards the Asian subtrop-
 46 ics. As a result, the northern region of South China Sea (SCS) has been the
 47 site of several aerosol characterization experiments in the recent years. Its lo-
 48 cation in the middle of the main Asian continent, Indochinese Peninsula and
 49 the Maritime Continent make its location useful in investigating emissions
 50 from different source regions.

51 There is limited literature on the extent of LRT of pollutants to the
 52 tropical western North Pacific like the Philippines. The most recent is the
 53 report in a previous study (Bagtasa et al., 2018) where high fine particulate
 54 mass concentration was observed in northern Philippines during the north-
 55 east monsoon season, particularly during the boreal spring season. Wind
 56 backtrajectory and chemical source apportionment analysis indicated that
 57 one-third of measured aerosols in the sampling site are due to LRT from
 58 northern East Asia. However, the study did not elaborate on the transport
 59 mechanism of LRT pollutants. The aim of the present study is twofold; (1)

we report a LRT event in northern Philippines in March of 2017 and investigate the sources and mechanism of aerosol LRT to the tropical WNP. (2) We also estimated the clear sky direct radiative forcing effect of the LRT aerosols over northern Philippines using the Fu-Liou radiative transfer model. This paper is organized as follows: The next section will describe the observational data, modeling and methodology used in the analysis. The results are presented in the third section followed by a brief discussion, and is then summarized in the final section.

2. Data and method

2.1. *sampling site*

Burgos (18.52°N, 120.65°E) is a town located in the northwestern-most part of Luzon Island in the Philippines. The small rural town has a population of about 10,000. Burgos is classified as a Type 1 climate under the modified Coronas climate type classification (Bagtasa, 2017; PAGASA, 2018) where the region experiences wet season from June to September, and a distinct dry season from October to April. The site is surrounded by the SCS to the west, Luzon Strait to the north, and the northern hills of Cordillera Mountain range to the east. In the present study, we performed simultaneous measurements using a filter-based chemical speciation of fine particulates, a Raman lidar, and the chemical transport model WRF-CHEM for a one week period in March of 2017. During the sampling period, the sampling site is characterized by ambient temperature ranging from 20° to 32°C and strong winds averaging 41.0 km hr⁻¹ and gusts of 52.2 km hr⁻¹ induced by the convection of the northeast monsoon wind flowing around the northwest edge

84 of the Cordillera mountains, or the corner and obstacle effect (Brody and
85 Nestor, 1985).

86 2.2. *PM_{2.5} chemical analysis*

87 Fine particulate (PM_{2.5}) samples were collected every 12 hours from 9
88 March to 16 March 2017. An air-sampler (BGI PQ200, USA) with 47 mm
89 quartz fiber filter at a flow rate of 16.7 L min⁻¹ was used. Each particulate
90 sampling was conducted from 0800H to 2000H Philippine Standard Time
91 (PST; UTC +8), and from 2000H to 0800H PST of the following day. The
92 0800H - 2000H PST and 2000H - 0800H PST sampling times will be referred
93 to as daytime (D) and nighttime (N) sampling, respectively. The quartz
94 fiber filters were dried and weighed before and after sampling. The filters
95 were then cut into four identical parts: One for the analysis of carbonaceous
96 components (Carlo Erba, Model 1108), another for water-soluble ionic species
97 (Dionex DX-120), for metallic elements (ICP-AES, Perkin Elmer, Optima
98 2000DV) and for the analysis of anhydrosugar (HPIC Dionex ICS 5000+).
99 A detailed description of chemical analysis is described in Bagtasa et al.
100 (2018).

101 2.3. *Lidar system*

102 The lidar system utilized in this study is part of the Total Carbon Col-
103 umn Observing Network (TCCON) station that was established in north-
104 western Philippines in 2017 (Velazco et al., 2017b). The lidar system is a
105 two-wavelength (532 and 1064 nm) polarization (532 nm) Mie and Raman
106 (607 and 660 nm) lidar system primarily designed to complement column
107 CO₂ retrievals by Fourier Transform Spectrometry (FTS) of the TCCON

108 site (Uchino et al., 2012). The Mie lidar data have a range resolution of 7.5
 109 m and 1 min integration time. System specification is summarized in ta-
 110 ble 1. Backscattering and extinction profiles were calculated using Fernald’s
 111 inversion method (Fernald, 1984) with the assumption of pure molecular
 112 backscattering at the far-end boundary condition at 22.5 km above ground
 113 level (AGL). The PM_{2.5} sampling site is located approximately 5 km west
 114 of the TCCON station. CALIPSO total attenuated backscatter and feature
 115 mask lidar data (Omar et al., 2009) were also utilized and downloaded from
 116 [https : //www – calipso.larc.nasa.gov](https://www-calipso.larc.nasa.gov) (accessed Jan 2019).

117 2.4. WRF-Chem and Hysplit

118 Weather Research and Forecasting - Chemistry (WRF-Chem) model (Ska-
 119 marock et al., 2005; Grell et al., 2005) was used to simulate PM_{2.5} transport
 120 and dispersion in the East and Southeast Asian regions. The WRF-Chem
 121 model with spectral nudging was used to downscale the ECMWF ERA5
 122 reanalysis from 0.3° horizontal resolution to 12 km. Gridded emission in-
 123 ventory of PM_{2.5} and PM₁₀ used in the simulation were from the Emis-
 124 sion Database for Global Atmospheric Research (EDGAR) version 4.3.1 data
 125 (<http://edgar.jrc.ec.europa.eu/>, downloaded January 2019). Table 2 shows
 126 the summary of the WRF-Chem model settings.

127 Analysis of wind back trajectories was done using the HYSPLIT model
 128 (Draxler and Hess, 1997). Meteorological conditions were driven by the same
 129 output from the WRF-Chem model run with 5-day spin up time. 96-H wind
 130 back trajectories were then plotted.

131 2.5. Radiative transfer model

132 The Fu-Liou (Fu and Liou, 1992, 1993) radiative transfer model (RTM)
 133 was used to estimate the radiative forcing impact of observed aerosols. It is
 134 a delta-four stream solver with fifteen shortwave (SW) and twelve longwave
 135 (LW) spectral bands. Atmospheric optical properties are calculated by the
 136 correlated k distribution method for each spectral band (Fu and Liou, 1992).
 137 Surface albedo spectral dependencies were taken into consideration using the
 138 IGBP land classification. In this study, lidar-derived aerosol optical depth
 139 (AOD) of up to 10 km was used to calculate clear sky (cloud free) aerosol
 140 net direct radiative forcing effect. Three surface types, cropland, evergreen
 141 broadleaf forest, and ocean, were used to depict land cover in the region of
 142 the sampling site.

143 3. Results

144 The WNP monsoon winds modulate the seasonal upwind and downwind
 145 regions of northern Philippines. It plays a major role in fine particulate
 146 transport that affects atmospheric aerosol loading variability of the region
 147 (Bagtasa et al., 2018). Prior to the March 2017 observation period, fine par-
 148 ticulate sampling in one to two-week durations for each season were done
 149 from 2015 to 2017. Seasonal fine particulate mean mass concentration and
 150 the corresponding standard deviation observed from 2015 to 2017 are shown
 151 in Fig. 1. Henceforth, June-July-August (JJA), SON, DJF and MAM sam-
 152 pling periods will be referred to as summer, fall, winter and spring, respec-
 153 tively. The summer and fall seasons coincide with the southwest monsoon
 154 and monsoon transition period, respectively. These two seasons show lowest

155 seasonal mean particulate mass concentrations of $8.40 \pm 3.86 \mu\text{g m}^{-3}$ in sum-
 156 mer, and $7.86 \pm 2.18 \mu\text{g m}^{-3}$ in fall. In the summer season, sea-salt is the
 157 dominant component of $\text{PM}_{2.5}$. Summer is characterized by the prevailing
 158 southwest winds and peak average rainfall of 450 mm per month in August
 159 (climatological normal rainfall of SYNOP 98223 located 40 km south of sam-
 160 pling site (PAGASA, 2019)). Frequent afternoon and evening thunderstorms
 161 in this season leads to low open burning activities around the sampling site
 162 that contribute to the overall low $\text{PM}_{2.5}$ concentrations. The transition pe-
 163 riod between southwest and northeast monsoon occurs in fall where easterly
 164 winds prevail. Wind backtrajectory analysis (Fig. 2 of Bagtasa et al. (2018))
 165 suggests air parcels arrive from the Philippine Sea or Pacific Ocean, where no
 166 known large pollutant emitters are present, hence, the observed low partic-
 167 ulate mass concentration. On the other hand, higher particulate loading in
 168 the region occurs during the northeast monsoon season in winter and spring.
 169 Mean particulate concentration is $10.87 \pm 7.15 \mu\text{g m}^{-3}$ in winter and 18.07
 170 $\pm 7.30 \mu\text{g m}^{-3}$ in spring. In these two seasons, measured concentrations of
 171 components associated with anthropogenic, industrial and biomass burning
 172 emissions were also elevated.

173 3.1. March 2017 $\text{PM}_{2.5}$ observation

174 In the present study, we focus on the springtime measurement in 2017
 175 to investigate the transport mechanism of the observed high fine particulate
 176 mass concentration. Figure 2a and 2b show the seven-day 12-hourly $\text{PM}_{2.5}$
 177 mass concentration and the corresponding water soluble (WS) ionic compo-
 178 nents, respectively. During the northeast monsoon season, observations show
 179 daytime concentrations to be higher than nighttime concentrations. This sug-

gests that the sporadic backyard burning of leaves and detritus prevalent in the region during daytime is likely a major factor in the diurnal PM_{2.5} concentration cycle. The emissions are then advected away by strong winds before nighttime. In addition, observed daytime wind speed is generally higher than that of nighttime with mean values of 55 km hr⁻¹ for daytime and 32.4 km hr⁻¹ for nighttime. More LRT pollutants carried by the stronger daytime monsoon winds may also have contributed to the higher daytime concentration. Both aforementioned sources, local burning and LRT pollutants, comprise two-thirds of the annual ambient concentration in the region (Bagtasa et al., 2018). During the seven-day sampling period from 9 to 15th of March 2017, two instances of elevated PM_{2.5} concentrations were observed. The averaged PM_{2.5} mass concentration observed on March 09D, 09N, 15D and 15N is 23.95 $\mu\text{g m}^{-3}$, which is more than 222% higher than the averaged concentration of all the other sampling days in between. These high particulate concentration days will be referred to as HC and conversely, LC for all other sampling days. Various chemical components were also found to be significantly higher during the HC days. Components associated with secondary inorganic aerosols SO₄²⁻, NO₃⁻ and NH₄⁺ were 1.4, 1.5 and 1.5 times higher, respectively. Several metallic components were also found to be significantly elevated. For instance, Zn and Pb were both at 2.4 times, Ni at 1.6 times, Al at 2.9 times, and Ca and V are both 2.1 times higher than LC days. Carbonaceous components EC and OC were 2.1 times higher during the HC days. Most of these components are associated with anthropogenic emissions from urban, industrial processing, and transportation (i.e. shipping) sectors (Haddad et al., 2011; Pérez et al., 2016; Xiao et al., 2018). OC/EC

equivalent to 2 or higher is generally an indicator of aerosols from distant sources (Chow et al., 1996). The OC/EC ratio was 2.32 and 1.97 for the HC and LC samples, respectively. This suggests that a large portion of all the observed aerosols for the whole spring 2017 sampling were of distant origin, consistent with our previous findings (Bagtasa et al., 2018). In addition, K^+ and levoglucosan concentration also showed significant increases of 1.5 and 1.9 times higher on HC days, respectively. Both these components are used as tracers for biomass burning (Urban, 2012).

3.2. Particulate transport modeling

To determine the possible sources of fine particulates during the HC days, 96-hour wind back trajectories were simulated using the WRF-HYSPLIT model. Figure 3a shows the wind backtrajectories associated with the HC aerosol loading days and fig. 3b for LC sampling days. A difference in upwind source locations for the HC and LC sampling days can be inferred from the calculated trajectories. Wind trajectories during HC days came from northerly regions of northeast China and the Korean peninsula, which then traverses the ECS before the air parcels reach northern Philippines. In the LC days, air parcels mainly originate from ECS or the region north to northeast of the Philippines. The air parcels are then transported southwestward along the tropical trade winds at around 20°N before reaching the northern part of the Philippines. The contours of fig. 3 are the Mean Sea Level Pressure (MSLP) isobars averaged from 96 to 48 hours prior to the $\text{PM}_{2.5}$ observations. In the days leading to HC samples, the 1015-1020 hPa MSLP isobars are meridionally oriented along the east coast of China that results in a northerly wind as depicted by the windbarbs. For the LC days, the 1015 and 1017 hPa

isobars show a ridge over southwest Japan extending to 145°E and 137°E, respectively. The ridge leads to a northeasterly upwind source. In addition, simulated northerly winds on HC days were stronger than the northwesterly LC days in the northern Philippine region, consistent with observed mean wind speeds at the sampling site of 59.4 km hr⁻¹ on HC and 29.2 km hr⁻¹ on LC days.

Figure 4 shows the simulated regional PM_{2.5} mass concentration from March 5-8 leading to the first HC sampling day. The upper (Fig. 4a-d) and lower (Fig. 4e-h) rows show the simulated PM_{2.5} concentration and windbarbs for surface and PM₁₀ concentration at 750-700 mb height aloft, respectively. In Fig. 4a), high PM_{2.5} concentration in the northeast region of China and the Korean peninsula is simulated in March 5 at 0 UTC, 4 days prior to HC observation. On March 6 (Fig. 4b), the air mass with high aerosol load moves and spreads south and southeastward from the Yellow Sea to ECS, and parts of western Japan. Strong northerly wind is seen across the Yellow Sea. On March 7 (Fig. 4c), the air mass is transported by strong northerly wind further south in the subtropics along 22°N with reduced concentration. The following day on March 8 (Fig. 4d), the air mass reaches northern Philippines wherein the LRT aerosols persist for two days. We also looked at the transport of emissions from a westerly airstream at 750-700 mb altitude during March described by Lin et al. (2009). In their study, they showed that a synoptic scale vertical transport of tropospheric pollutants from northwestern China extending southward to Indochina was due to the existence of a deep trough on the leeward side of the Tibetan plateau and Indochina mountains. The WRF-Chem simulation shows large quantities of

upper-air pollutant transport eastward from the main Asian continent. We note that the EDGAR emission inventory does not include large scale burning. Here, the simulated PM_{10} is to illustrate the upper-air transport in the region rather than estimate biomass burning emissions. Model result also shows emissions from the Indochina region lifted aloft which contributed to the southern periphery of the transported polluted air mass. As the northerly wind strengthens from March 5 to 8, the stream of polluted air shifts southward until the southern periphery go over northern Philippines.

The WRF-Chem simulation for the second HC day from March 12 to 15 is shown in fig. 5. Figure 5 shows a similar pattern as the first HC day in fig. 4. Figure 5a starts with an increase in simulated concentration in the northeast part of the main Asian continent that is transported southwards by a strong northerly wind along the Yellow and ECS (Figs. 5b-d). At 750-700 mb height, a stream of polluted air moves eastward from the main Asian continent and Southeast Asia. The strong northerly wind pushes the stream southward where the southern periphery reaches the northern region of the Philippines.

3.3. Synoptic conditions

The MSLP in fig. 4a shows an eastward moving anticyclone located to the south of Japan ($30^{\circ}\text{N}, 140^{\circ}\text{E}$) created an extended ridge of the 1015-1017 hPa isobar along the subtropics at 20°N . This ridge temporarily shifts the northeast monsoon to an east-northeasterly prevailing over northern Philippines. At this time, a cyclonic circulation over the northeast China plain is displaced by a strengthening Siberian high, which induces a stable atmosphere in that region and results to high particulate concentration in northeast China. On

280 March 6 (Fig. 4b), the displaced cyclonic circulation had move eastward over
 281 northeast Japan. The strengthening Siberian high induces a strong north-
 282 westerly wind along its eastern flank and extends the high pressure ridge
 283 across most of the main Asian continent. This continental high pushes an-
 284 other low pressure system emerging from the main Asian continent eastward
 285 to southern Japan, which in turn displaces the anticyclone to the east. This
 286 continental high and the cyclonic circulation over southern Japan produced
 287 a confluent northerly wind flow along the Yellow and ECS regions. This
 288 northerly wind induced by the confluent flow sustains the southward trans-
 289 port of the polluted air masses in the succeeding two days. This wind flow
 290 is characterized by meridionally-oriented 1015-1020 hPa isobaric lines along
 291 the eastern China coastline. As the low pressure moves towards the Pacific
 292 Ocean on March 7 (Fig. 4c), the 1015-1020 hPa isobars from the continen-
 293 tal high further extend southward to Luzon Strait at 18° - 22° N. The strong
 294 northerly wind in ECS continues to prevail and affects northern Philippines.
 295 On March 8 (Fig. 4d), an eastward moving anticyclonic circulation emerges
 296 from the continental high towards the Pacific. This anticyclone blocks the
 297 northerly flow and extends the high pressure ridge to the Pacific subtropics,
 298 this condition resembles the wind-backtrajectories on LC days shown in fig.
 299 3b.

300 Similar changes in synoptic conditions that led to the second HC event
 301 are shown in fig. 5a-d. March 13 shows the 1015-1017 hPa ridge extended to
 302 the north central Pacific. At this time, a low pressure system in the northeast
 303 China plain is displaced eastward by the Siberian high and leads to a stable
 304 boundary layer condition in that region. On the following day, March 14,

the low pressure system moves eastward to the northeast of Japan. At the same time, another low pressure system moves to southern Japan. This synoptic pattern induces the same confluent strong northerly flow along the Yellow and ECS as characterized by meridionally-oriented isobars along the eastern China coast. This strong northerly wind prevails for two days. After which, an eastward moving high pressure system emerges from the main Asian continent and returns the 1015-1017 hPa isobar back to its initial position.

To illustrate both the surface and upper-air particulate transport, fig. 6 shows the vertical cross-section of the simulated mean $\text{PM}_{2.5}$ and cloud and ice mixing ratios (gray mask) along 126°E longitude averaged from 5 to 9 of March 2017. On the other hand, fig. 7 shows the CALIPSO lidar total attenuated backscatter and feature mask data taken on March 6 along the same longitude. The WRF-Chem simulation was able to resolve the general features of particulate distribution cross-section, in particular, the characteristic of upper-air transport from 18°N to 35°N . The upper-air transport shows varying altitude with respect to latitude. Simulated $\text{PM}_{2.5}$ along the upper airstream is at lower heights of about 2 km and less in lower latitude regions of about 18°N . The height of this polluted airstream gradually increases to approximately 8 km AGL at 35°N . This feature is seen on both the model simulation and CALIPSO lidar data. In addition, elevated surface $\text{PM}_{2.5}$ concentration that extends to 2-3 km AGL is also seen from 33°N to 45°N on both simulation and CALIPSO data. The gray mask of fig. 6 is the combined simulated cloud and ice mixing ratio to depict cloud distribution in the model domain.

330 3.4. Lidar observation

331 Figure 8a and 8b show the 532 nm observed range-corrected signal and the
 332 extinction coefficient profile, respectively, of the TCCON lidar from March
 333 5 to 16. Lidar observation at the sampling site shows two distinct layers
 334 of high backscatter signal. One on the surface up to 2 km and one aloft
 335 from 2 to 4 km AGL. These layers are more apparent during the first HC
 336 sampling day from March 8 to 10. Extinction coefficient on the first HC day
 337 reached approximately 0.4 km^{-1} from 2.5-4 km AGL. The calculated AOD for
 338 March 9 had an average value of 0.78, and 0.68 for the second HC sampling
 339 day (March 16) at the end of the sampling period. It is apparent that the
 340 aerosol transport aloft only changed in terms of quantity but was present all
 341 throughout the sampling period. The average AOD obtained for LC days
 342 was 0.56. It is also worth noting that the high altitude cirrus clouds detected
 343 by the lidar at 14-16 km is consistent with the CALIPSO observation for
 344 18.5°N latitude, and to a certain extent, well simulated by the WRF-Chem
 345 model.

346 4. Discussion

347 According to a previous study, Velazco et al. (2017a) reported an increase
 348 in observed xCO concentration from a mean baseline value of 90-100 ppb
 349 to 140 ppb that peaked in 8-9 March 2017 at the TCCON sampling site.
 350 Furthermore, another increase of approximately 25 ppb xCO that peaked in
 351 March 16-17 was observed. Analysis in the present study shows that other
 352 than the increase in column CO, increase in ground fine particulate are due to
 353 surface pollutant transport from East Asia and transport of biomass burning

354 emission aloft from Indochina.

355 The southward transport of surface pollutant induced by a strong Siberian
 356 high had been reported to influence pollutant concentration in certain regions
 357 of East Asia (Jeong and Park, 2017; Wang et al., 2017; Zhao et al., 2018) .
 358 Consequently, the southward movement of pollutants led to an increase in
 359 observed concentration in central and southern China (Wang et al., 2017).
 360 Here, this weather pattern was shown to transport pollutants that reach the
 361 tropical WNP region of northern Philippines. The reduction of pollutant
 362 concentration in northern China can be compared to a 50% reduction in
 363 emission (Zhao et al., 2018), this is beneficial for the large population in
 364 that region who are continuously exposed to high pollutant concentration.
 365 However, the impact of these aerosols is shown to shift to downwind regions.

366 The other LRT aerosol pathway found is the upper-air westerly wind flow
 367 originating from the northern Indochina region to the WNP. This westerly
 368 wind is known to transport biomass burning emission to the subtropics of
 369 the WNP in spring (Lin et al., 2009). However, unlike the result of Lin
 370 et al. (2009) where biomass burning aerosols aloft were not measured near
 371 the ground, the large increase in biomass burning markers levoglucosan and
 372 K^+ during the HC days suggests biomass burning pollutants aloft were able
 373 descent and affect ground concentrations in northern Philippines. The range-
 374 corrected lidar signal shows a downdraft motion from 2-2.5 km to the lower
 375 detection height limit (< 1 km). Aerosol intrusion from the free troposphere
 376 to the boundary layer started from early morning to evening of March 8 that
 377 led to a high backscatter signal and extinction coefficient values from the
 378 evening of March 8 to morning of March 9. The same case was also observed

379 on March 15 to March 16. This is consistent with the flow model of Lin et al.
 380 (2013) where the downward branch of a transient local East-west circulation
 381 due to a cold-surge anticyclone was shown to bring down high altitude pol-
 382 lutants in southern Taiwan (Yen et al., 2013). This downwash mechanism
 383 apparently also affects ground concentration in northern Philippines. The
 384 observed lidar depolarization ratio and lidar ratio show values ranging from
 385 4-8% and 45-70 sr during the HC sampling days, respectively. Aerosols de-
 386 tected aloft at 2 - 4 km recorded depolarization ratios of about 6-8% and lidar
 387 ratios ranging from 57-73 sr. These values were higher compared to near-
 388 ground depolarization and lidar ratio of 4-5% and 40-50 sr, respectively. This
 389 implies that surface pollutants were still dominated by anthropogenic rather
 390 than biomass burning pollutants, consistent with observed fine particulate
 391 chemical analysis and model.

392 A conceptual model of the transport mechanism is illustrated in fig. 9. On
 393 the surface, confluent wind flow due to a high over the main Asian continent
 394 and low pressure system in the vicinity of southern Japan creates a strong
 395 northerly flow along the Yellow sea and ECS. This strong northerly flow
 396 transports aerosols from northeast China and the Korean peninsula south-
 397 wards to northern Philippines. At the same time, an eastward stream of
 398 strong upper-air westerly flow which carries biomass burning emissions from
 399 the Indochina shifts southward due to the same strong northerly flow from
 400 northern East Asia. The transported biomass burning emissions traverses as
 401 far south as northern Philippines.

402 4.1. Aerosol radiative effect

403 The transported pollutants from both surface and upper-air vertically ex-
 404 tend from surface up to more than 4 km AGL. This leads to a mean AOD
 405 of approximately 0.73 derived from the integration of the lidar extinction
 406 profile during the HC sampling days. To estimate the radiative effect of the
 407 LRT aerosols in the northern region of the Philippines, we start by examin-
 408 ing the relationship between the optical and mass properties of these LRT
 409 fine particulates. We used the parameter called mass extinction efficiency or
 410 MEE. MEE is commonly used in describing the total light extinction (sum of
 411 scattering and absorption) per unit mass of aerosol (Hand and Malm, 2007).
 412 The MEE value is useful in estimating an equivalent optical depth to de-
 413 rive aerosol radiative effects from observed mass concentration essential in
 414 determining the impact of aerosols on Earth's climate (Myhre et al., 2004;
 415 Hansell Jr et al., 2011). Here, we calculated the MEE from the 12-hourly
 416 averaged extinction coefficient derived from the lidar and PM_{2.5} mass con-
 417 centration measurements. The lidar-derived extinction coefficient used was
 418 the column-averaged extinction coefficient from 700 m to 4 km height. The
 419 column integrated extinction was significantly correlated with ground PM_{2.5}
 420 mass concentration at $r = 0.85$ ($p < 0.05$) as shown in fig. 10.

421 The calculated MEE ranges from $2.78 \text{ m}^2\text{g}^{-1}$ to $8.69 \text{ m}^2\text{g}^{-1}$ and an average
 422 of $5.38 \pm 1.91 \text{ m}^2\text{g}^{-1}$. Hygroscopicity of aerosols can lead to an overestima-
 423 tion of MEE (Bagtasa et al., 2007). During the sampling period, relative
 424 humidity observed in SYNOP 98223 station ranged from 68% to 84%. The
 425 aerosol mass concentration derived from the quartz fiber filters were measured
 426 after being stored in a dry environment, thus, mass concentration presented

427 here is mass of dry aerosols. To correct the hygroscopic effect, we determine
 428 the ambient aerosol mass concentration by calculating the density change of
 429 several aerosol components (i.e. Na^+Cl^- , $\text{NH}_4^+\text{NO}_3^-$, $\text{NH}_4^+\text{SO}_4^{2-}$) due to am-
 430 bient humidity, details of this method is described in Bagtasa et al. (2007).
 431 After the correction, mean MEE yielded a value of $4.32 \pm 1.32 \text{ m}^2\text{g}^{-1}$. The
 432 result here agrees with the observed MEE of 3.0 to $4.7 \text{ m}^2\text{g}^{-1}$, with a mean
 433 value of $4.3 \pm 0.6 \text{ m}^2\text{g}^{-1}$, from the Asian Pacific Regional Aerosol Charac-
 434 terization Experiment (ACE-ASIA) campaign in spring of 2001 for polluted
 435 Korea/Japan region (Quinn and Bates, 2005; Hand and Malm, 2007).

436 The radiative effect of aerosols is one of the major uncertainties in our
 437 understanding of our climate (Stocker et al., 2013). Due to the high spatial
 438 and temporal variability of aerosol concentration, as well as variability in
 439 chemical makeup, it is difficult to assess their impacts on the climate by
 440 using globally-averaged measurements. There is a strong need for regional or
 441 area specific aerosol direct radiative forcing estimation. The direct radiative
 442 forcing effect of LRT aerosols in the northern Philippines was estimated using
 443 the Fu-Liou radiative transfer model. Compared to pristine summer/fall
 444 extinction profiles, the estimated net direct radiative effect of LRT aerosols
 445 at TOA and surface had mean values of $-11.98 \pm 3.96 \text{ Wm}^{-2}$ and $-50.80 \pm$
 446 12.38 Wm^{-2} , respectively. In the HC days, there were approximately 40%
 447 more cooling effect compared to the LC spring sampling days. An estimated
 448 mean uncertainty of 17% for surface and 8% TOA radiative forcing were
 449 mainly due to the underlying land-cover type used in the calculation.

5. Conclusion

Two-year seasonal fine particulate sampling was conducted in the town of Burgos, Ilocos Norte, located in the northwestern edge of the Philippines. The seasonal variability of measured $\text{PM}_{2.5}$ is influenced by the prevailing monsoonal wind flow. Lowest fine particulate concentration is found in fall and highest in spring. In addition, observed springtime particulates are from local and LRT anthropogenic sources (Bagtasa et al., 2018). In this research study, we focus on the spring measurement of March 2017 sampling where two high aerosol loading days were observed in a span of a week.

We present a conceptual model to describe the mechanism of pollutant transport to the region of northern Philippines. We found two source regions where emitted pollutants were advected on the surface and aloft. On the surface, confluent wind flow from a high pressure system over the main Asian continent and low pressure south of Japan creates a strong northerly flow in East China Sea. This strong northerly transports mainly anthropogenic aerosols from northeast China and the Korean peninsula towards northern Philippines. On the other hand, a strong upper-air westerly flow carries biomass burning emissions from the Indochina region eastward to northern Philippines. We estimated the radiative effect of these transported fine particulate pollutants. The combined radiative effect of the LRT aerosols have a net cooling effect of $-11.98 \pm 3.96 \text{ Wm}^{-2}$ and $-50.80 \pm 12.38 \text{ Wm}^{-2}$ at TOA and surface, respectively.

The strong Siberian high that initiated the advection transport is reportedly affected by climate change. Zhao et al. (2018) found that warmer arctic air temperature, which is inversely correlated to loss of sea ice in the arc-

475 tic region, is correlated to the strengthening of the Siberian high. While
 476 a stronger high leads to stronger than usual winds in parts of China that
 477 lessens wintertime air pollution loading in that region of Asia, it also leads
 478 to higher LRT aerosols in downwind regions like the Philippines. This may
 479 have implications on the health impacts, as well as shifts in the climatic
 480 effects of LRT of particulates in the future.

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Table 1: Lidar system specification

Transmitter				
Laser	Nd:YAG			
Wavelength	532	1064		
Pulse energy	210 mJ	180 mJ		
Pulse frequency	10 Hz			
Pulse width	5 ns	6 ns		
Beam divergence	0.1 mrad	0.1 mrad		
Receiver				
Telescope	Advanced Coma-Free			
Diameter	356 mm (f = 2845 mm)			
FOV	1 mrad			
Wavelength (nm)	532	1064	607	660
Polarization	P and S			
Channel	3	1	1	1
Filter bandwidth	0.32	0.38	0.25	0.3
Detectors	PMT	APD	PMT	PMT
Quantum eff.	0.08	0.68	0.06	0.04
Signal	16 bit A/D + PC			
Time resolution	1 min			
Range resolution	7.5 m			
Maximum range	120 km			

Table 2: WRF-Chem model configuration

WRF-Chem configuration	
Microphysics	WSM6
LW radiation	RRTM
SW radiation	Goddard
Land surface	NOAHL LSM
PBL	Yonsei University Scheme
CU physics	Kain-Fritsch
Chemistry	MADE/SORGAM
Emission data	EDGAR ver. 4.3.1
Spectral nudging	Yes
Domain resolution	12 km

Figure 1: Observed seasonal PM_{2.5} mass concentration from 2015 - 2017.

Figure 2: 12-hourly a) PM_{2.5} mass concentration and its b) water soluble ionic components from 9 March to 16 March 2017.

Figure 3: 96-H wind backtrajectory using WRF-Hysplit and composite mean sea level pressure 96 to 48 hours before the a) HC and b) LC sampling days in March 2017.

Figure 4: WRF-CHEM simulated surface $PM_{2.5}$ (upper row) and 750-700 mb height (lower row) PM_{10} each at 0 UTC from 5 to 8 March 2017

Figure 5: WRF-CHEM simulated surface $PM_{2.5}$ (upper row) and 750-700 mb height (lower row) PM_{10} each at 0 UTC from 12 to 15 March 2017

Figure 6: WRF vertical cross section of $PM_{2.5}$ (color) and cloud/ice mixing ratio (gray mask) at $126^{\circ}E$ averaged from 4 to 9 March 2017.

Figure 7: CALIPSO (UTC56190) a) attenuated backscattering and b) vertical feature mask data for 6 March 2007.

Figure 8: a) Range corrected and b) extinction coefficient profile of 532 nm Mie lidar signal from 5 to 17 March 2017. Colorbar unit: a) arbitrary and b) km^{-1}

Figure 9: Conceptual synoptic flow for springtime LRT events in northern Philippines

Figure 10: PM_{2.5} mass concentration vs. extinction coefficient from 9-16 March 2017.

Long-range transport of aerosols from East and Southeast Asia to northern Philippines and its direct radiative forcing effect

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Highlights:

1. Spring-time aerosol transport pattern from East Asia to the Philippines.
2. Biomass burning emissions from Indochina observed in northern Philippines.
3. Conceptual flow of long-range transport of aerosols during spring.
4. Transport of aerosols has cooling effect in the region.