Dear reviewers and editor:

We are really grateful to reviewers who spent much time reviewing the original manuscript. Through the review processes, we totally understand that this manuscript could not be accepted without the reviewers' valuable comments. Please notice that the revision according to reviewers' comments are written in **red words.** In this response, we have attached three files: the manuscript of the main context, the supplement, and the one-to-one response. Also, we sincerely thank for the editor and ACP staff's effort again.

Best regards.

Ming-Tung Chuang

Response to Reviewers

Manuscript *acp-2019-762*

We greatly appreciate the insightful comments and suggestions of the reviewers. Below please find a list of the Reviewers' remarks in contrast to our responses to them:

Review #1

Major Concerns		
(1) comments from Anonymous Referee #1	(2) author's response	(3) author's changes in manuscript.
The manuscript has been revised thoroughly well according to the reviewer's comments and become more scientifically focused than before. However, there still are several sentences not easy to understand their meaning properly which will require further English editing. Furthermore, there are several sentences which describe a figure or table, but it is not clear which part of figure or table they are describing. I strongly recommend the authors to check all	On behave of all authors the first author sincerely thank the reviewer spent a lot of time reading the manuscript, finding numerous errors, and giving many valuable comments. The first author has to be responsible for unclear expression of narratives in the manuscript. Before the last submission, the first author has asked two co-authors and a professional editing company to revise the manuscript. However, the revised manuscript still contains some improper writing. Due to limited time, the authors have asked a senior colleague to help revise it instead for this revised manuscript. Furthermore, the first author has tried to read carefully and repetitively to ensure the clarification of writings.	**In addition to the response regarding to comments in this review, the authors have revised other narratives in this revised manuscript.** On line 19-20 When the Asian anticyclone moved from the Asian continent to the West Pacific, e.g., on Jan 9, 2017, the contributions from BRIR and YRDIR to northern Taiwan could reach daily averages of 8 and 11 μg m^{-3}.

such descriptions and revise them if necessary. One example of such description is in the line 267 starting from “The increase of PM2.5…”. I could not understand which part of the Figure 5 you want to describe. There still are such unclear descriptions which greatly deteriorate the readability of the manuscript. Another big concern is that the introduction section is still long and redundant. It’s good to have a thorough review of the background, but it should cite only indispensable papers. Followings are specific points for further revision.	**In this revision, the authors have tried to reduce some cited literatures in the Introduction section already, listed below:** The EAH has started to spread out from Asia Continent to East Asia in spring and winter due to the movement of anticyclones. (Fu et al., 2014~~; Yang et al., 2016~~). The trajectories could be calculated from, for example, the archived meteorological data of NOAA ARL (www.ready.noaa.gov/archives.php) or ~~the model outputs of MM5 (Mesoscale Model version 5, Dudhia, 1993), or~~ WRF (Weather Research and Forecasting, Skamarock and Klemp, 2008) simulation results. ~~Yang et al. (2018) also used this method to evaluate the influence of $PM_{2.5}$ from the Bohai Sea, Yangtze River Delta, and Pearl River Delta regions on Beijing.~~ The BFM method has been widely used for estimating the contribution of a specific source or the effect of a control strategy (~~Marmur et al., 2005;~~ Burr and Zhang, 2011; Chen et al., 2014; Li et al., 2017) because this method is easy and straightforward. Skyllakou et al. (2014) applied the particulate matter source apportionment technique (PSAT, Wagstrom et al., 2008) in the PMCAMx model (Fountoukis et al., 2011) to assess the impact of local pollution (LP), short distance transport ~~(50–500 km)~~, and LRT ~~(>500 km)~~ on Paris, France. ~~Chuang et al. (2008a) classified the pollution weather patterns for the Taipei $PM_{2.5}$ events. They named the weather system during LRT events as “high-pressure pushing”, in which the high-pressure systems transported the pollutants from the Asian continent to Taiwan.~~	On line 67-68 Nevertheless, this method is not perfect because it potentially ignores chemical reactions between the specific sources within~~in~~ the remaining sources. On line 86-87 Chuang et al. (2008b) utilized CMAQ to simulate the chemical evolution of $PM_{2.5}$ compositions in the moving plume from Shanghai to Taipei. On line 91-92 Chuang et al. (2017) simulated three types of $PM_{2.5}$ episodes in the LRT, the LP and the LRT/LP mix. On line 92-94 Both the simulation and observation showed the proportion of NO_3^- in $PM_{2.5}$ was very small in the EAH and a strong north-to-northeast wind increased the proportion of sea salt at the northern tip of Taiwan. On line 94-95 Chuang et al. (2018) developed an efficient method which make use of five-month simulation results to estimate the LRT-$PM_{2.5}$ and LP-$PM_{2.5}$ at any place in Taiwan. On line 145-146

	~~Subsequently,~~ ~~Lin et al. (2004) examined the meteorological and air quality data from November 1999 to May 2000, and from November 2000 to May 2001 in Taiwan. They classified the LRT in winter into dust transport, frontal transport with pollutants, and LRT of background air masses, which contributed an average PM_{10} level of 127.6 μg m^{-3}, 85.0 μg m^{-3}, and 32.8 μg m^{-3}, respectively. Furthermore, the frequencies of LRT events and LP events were 25.2% and 71.7% (missing data accounts for 3.1%).~~ ~~Wang et al. (2016) combined backward trajectories and AOD distributions to estimate the impact of EAH on Taiwan. Their results suggested the $PM_{2.5}$ level was 57.1±13.6 μg m^{-3} for haze events, which is four-fold higher than the background events (13.7±7.4 μg m^{-3}) from 2005 to 2013.~~ ~~On average, the ratio of LRT-$PM_{2.5}$ and LP-$PM_{2.5}$ for an LRT-Event was 70:30 for northern Taiwan, 50:50 for central Taiwan, and 30:70 for southern Taiwan; for an LRT-Ordinary the ratio was 60:40 for northern Taiwan and 40:60 for central and southern Taiwan; for LRT/LP&Pure LP it was 30:70 for northern Taiwan and 25:75 for central and southern Taiwan. Their results also showed the annual LRT-$PM_{2.5}$ decreased since 2013, which implied the emissions from the Asian continent decreased since then.~~ ~~The above studies all showed the East Asian continent was the dominant source of LRT $PM_{2.5}$ for Taiwan in the winter period. Therefore, i~~If we can identify the sources contributing the most to the LRT $PM_{2.5}$ and the transport pathway, then we can enhance the ability to predict the LRT $PM_{2.5}$, i.e., the EAH.	They used the MetOne SASS $PM_{2.5}$ samplers (Met One Instruments, Inc.) for collection of the 24-hour (00:00 to 00:00) $PM_{2.5}$ composition samples ~~at six stations~~ every six days. On line 170-172 This study used statistical indexes such as MB (Mean Bias), MAGE (Mean Average Gross Error), and IOA (Index of Agreement) to evaluate temperature, wind speed, and relative humidity, and used WNMB (Wind Normalized Mean Bias) and WNME (Wind Normalized Mean Error) for wind direction in the fourth domain. On line 176-177 The MB performance for *Base* case shows that the temperature was slightly overestimated for PJY (Table 1), which is located in the outer sea of northern Taiwan. The MAGE of simulated temperatures at all stations are reasonable for both months. On line 205-207 Although the proportion of contribution from LRT was higher in July than January; however, the $PM_{2.5}$ levels in January were much higher than those in July due to the impact of EAH. On line 223-224 In particular, the contributions from BRIR and

	In addition, this study applied the Integrated Process Rate (IPR) technique (Byun and Schere, 2006; Liu and Zhang, 2013~~; Zhu et al., 2015~~) in CMAQ to discuss the process analysis during transport from the industrial regions to Taiwan.	YRDIR to northern Taiwan could reach daily averages of 8 and 11 μg m^{-3} on Jan 9, 2017. On line 227-228 For the stations on flat western Taiwan, there was slight influence on the 8th to 12th January 2017 (Fig. 3(c-1)-3(c-3)). On line 228-230 It was found that there was a stationary front from the sea north of Taiwan that extended southwest to Fujian and Guangdong provinces (letter *F* and *G* indicated in Fig. 1) on January 7th (Fig. S4.6(a)). On line 249-250 Through the value of each term in the process analysis, we can understand which term can produce or remove $PM_{2.5}$ at these positions and therefore realize the physical and chemical processes during LRT. On line 316-320 This suggests the ascent and subsidence of air parcels might enhance the formation and removal of aerosols below and above 760 m, respectively. It is possible that the ascent motion of the air parcel near the warm surface moved to a cold environment at a higher altitude, up to 760 m. This may cause condensation and trigger

		heterogeneous reactions of aerosols. In contrast, the descent motion of the air parcel above 760 m may cause the evaporation of aerosols due to a warmer environment at lower altitude than aloft. On line 330-331 ~~Although #2 and BQ were most affected by YRDIR, t~~The major contribution processes at BQ below 200 m (layer 7) was HADV, followed by AERO and above 200 m ~~it~~ were either VDIF, ZADV, or CLDS, or mixture of them. On line 375-377 This suggested the $PM_{2.5}$ was mainly from local pollution and background atmosphere on July 18th. On the other hand, on July 30th the 72-hour backward trajectory ensemble starting from the end at BQ/ZM/CY went through a cyclone near Taiwan and then to the South China Sea and Philippines (Fig. S4.7(d-1)-(d-3)). On line 387-388 As illustrated in Fig. 11, on both Jan 12th and Jan 13th, the major simulated compositions were sulfate and OC for #1 - #4. On line 390-391 The simulated proportions of Na^+ and Cl^- in $PM_{2.5}$ at # 19 and #4 were higher than those at #1 and #2.

		On line 393-394 In addition, the simulated proportions of nitrate in $PM_{2.5}$ at BQ, ZM, and CY were higher than those over #1 - #4. On line 400-402 As mentioned earlier, #1 was influenced by upstream YRDIR, the simulated proportion of nitrate in $PM_{2.5}$ at #1 was higher than further upstream #2, #3, and #4. The simulated proportion of nitrate in $PM_{2.5}$ at #3 and #4 was higher than #2, which implies #3 and #4 were influenced more by PRDIR than #2. On line 405-407 In addition to the local emission inventory, the underestimation of sulfate could possibly be related to underestimation of emissions from uncertain sources, e.g. ships around Taiwan or local point sources, since the local line and area sources of SO_2 are both low. On line 416-417 On that day the contribution from BRIR and YRDIR on northern Taiwan could reach daily averages of 8 and 11 μg m^{-3}, respectively. On line 417-419 ~~In contrast, t~~The influence of PRDIR on Taiwan was much less than BRIR and YRDIR.

		However, the $PM_{2.5}$ from PRDIR can influence Taiwan ~~with a monthly average impact of approximately 0.5 μg m^{-3}~~ via transboundary transport and boundary layer mixing (VDIF), and this is enhanced when a cold surge passes Taiwan.
-L121: What does "integrated emissions" mean?	The authors have recover the "integrated emissions" back to "all emissions".	On line 105-108 It applied the CTM with the BFM method to simulate four scenarios: *Base* (control case with all emissions), *BRIR* (all emissions except BRIR), *YRDIR* (all emissions except YRDIR), and *PRDIR* (all emissions except PRDIR) scenarios and thus resulted in the determining the contributions of each industrial region.
-L139: Why "therefore" here?	Thanks the reviewer finding this improper writing. It was an error made in the last submission. The authors have modified the narratives and make the sentence smooth.	On line 122-126 In previous studies (Zheng et al., 2018; Chuang et al., 2018), the anthropogenic emissions in China have obviously decreased since 2013; therefore, a year after 2013 was chosen. Moreover, in order to show the difference of transport between winter and summer, this study chose January and July 2017 to represent the LRT in the winter and summer period and their contrast, with more discussion on the winter transport due to greater impact of EAH.
-L172: MIX inventory is not only for China	Yes, the authors have removed the words "Multiresolution Emission	On line 157-158

	Inventory for China" which can be short to MEIC instead of MIX and in order to avoid misleading.	The anthropogenic emissions for East Asia and Taiwan island were obtained from MIX (~~Multiresolution Emission Inventory for China,~~ Li et al., 2017) and TEDS 10.0 (Taiwan Emission Data System, TEPA, 2017), which are based on the years 2010 and 2016, respectively.
-L174-176: Did you do this adjustment for MIX emission in entire model domain?	The authors only adjusted the MIX emissions for Chinese mainland and have modified that sentence to avoid misleading.	On line 158-161 The MIX emissions of SO_2, NO_X, NMHC, NH_3, CO, PM_{10}, and $PM_{2.5}$ covering Chinese mainland were adjusted with changes of -62%, -17%, 11%, 1%, -27%, -38%, and -35%, respectively, according to the change of annual emissions between 2010 and 2017 (Zheng et al., 2018).
-Table1: Which scenario did you use for this table? This kind of basic information should be described in the table caption or manuscript. Temporal resolution of the observation should be described somewhere in the manuscript. The same comments go to Table2.	The model performance was for the base case. The authors have added the basic information in the manuscript and bottom of Table 1 and Table 2.	On line 176-177 The MB performance for *Base* case shows that the temperature was slightly overestimated for PJY (Table 1), which is located in the outer sea of northern Taiwan. On line 190-191 For the *Base* case, the simulated $PM_{2.5}$ was overestimated at all stations except CY and HC in January 2017 (Table 2). The performance of the trend (correlation coefficient, R) is acceptable or good for all stations except HC.

		At the bottom of Table 1 and Table 2 Note: 1. The standard of statistical evaluation is based on Emery (2001) and TEPA (2016); 2. The above evaluation was for base scenario; 3. The observation and simulation data for above evaluation was in hourly resolution.
-L192-193: Does this sentence describe only for July case?	Thank the reviewer for pointing out this mistake. The authors have modified that sentence to indicate that is only for July case	On line 177-178 The MAGE of simulated temperatures at all stations are reasonable for both months. However, the IOA indicates the simulated temperature at PJY and KH in July was less correct.
-L219: How did you estimate these values of the contributions of local pollution?	We have improved the narratives for clearer description. We made a very simple assumption that the $PM_{2.5}$ at HC is the $PM_{2.5}$ from background atmosphere for all sites. The difference between measured $PM_{2.5}$ at each site and the background $PM_{2.5}$ is attributed to local pollutions for each sites, respectively.	On line 201-205 Even if we ignore the LP and simply assume the measured $PM_{2.5}$ at HC represents the background air quality for all sites, from Table 2, it is estimated that the contributions of local pollution was the difference between measured $PM_{2.5}$ at each sites and the background $PM_{2.5}$, for northern (BQ and PZ), central (ML and ZM), and southern Taiwan (CY, TN, and ZY) were 41–42%, 54–63%, and 75–78% of measured $PM_{2.5}$ in January, and 22–32%, 33–48%, and 36–39% in July, respectively.
-Figure 2: Figure caption should state that these figures show the difference between Base	The authors totally agreed with the reviewer's suggestion and have added information both in the manuscript and the caption of Figure 2.	On line 211-213. For the impact of the three industrial regions on

case and the other sensitivity simulation case for the sake of clarity. The term “impact” is not so clearly describing what you show here.		$PM_{2.5}$ in Taiwan in January 2017, the monthly mean impact from BRIR (difference between *Base* and *BRIR* scenario) was approximately 0.7–1.1 µg m^{-3} as illustrated in Fig. 2(a). The caption of Figure2 **Figure 2: The monthly average wind field and impact of $PM_{2.5}$ from BRIR (difference between Base and zero-out scenarios): (a) concentration and (b)percentage ; YRDIR: (c) concentration and (d)percentage ; PRDIR: (e) concentration and (f) percentage on Taiwan in January 2017**
-Figure 3: The same indications for Figure2 above. Furthermore, the Y-axis of Fig 3 should be delta(concentration).	The authors have added similar information in caption of Fig. 3. In addition, the authors have modified the label of Y-axis to “Δ concentration” for Fig. 3 and Figure S4.8 in the manuscript of this submission.	The caption of Figure3 **Figure 3: The daily average impact of PM2.5 from BRIR, YRDIR, PRDIR on air quality stations in Taiwan in January 2017. a,b, and c denote the impact on BQ, ZM, and CY from 1 (BRIR), 2 (YRDIR), and 3 (PRDIR). The impact was calculated with BFM method, i.e., the difference between the base and zero-out scenarios.**
-L226: The “relative” impact was..	Yes, the authors have added “relative” into that sentence which is more clear than the original.	On line 213-214 The relative impact was higher in northern Taiwan, approximately 5% of total $PM_{2.5}$.
-L239: 2(f)) --> 2(f)	Thanks the reviewer for finding this error which have been revised	On line 226-227

	already.	The spatial distribution of influence from PRDIR was totally different from BRIR and YRDIR, as shown in Fig. 2(e) and Fig. 2(f).
-L266: appeal --> appear?	Thanks the reviewer for finding this error which have been revised already.	On line 254-255 The physical or chemical terms in Fig 5 (a-1) and Fig. (a-2) did not always appear synchronously, and their proportions in total were not equal.
-L275-276: Fig5(c-2) and Fig5(c-3) should be switched.	Thanks the reviewer for finding this error which have been revised already.	On line 264-266 For #3, $PM_{2.5}$ was influenced mainly by YRDIR (Fig. 5(c-3)) and occasionally by BRIR (Fig. 5(c-2)), but it was also influenced by PRDIR from the 8th to 12th (Fig. 5 (c-4)) with positive contribution of CLDS, which could be attributed to high relative humidity environment over Taiwan Strait.
-L276-277: Is this consistent with the fact that CLD is the main production process in Fig5(d-4)?	Thanks the reviewer finding this inconsistent narrative and have modified that in the revised manuscript. Meanwhile, the authors have removed "Fig. 5(d-4)" on line 280 of the original manuscript.	On line 264-266 For #3, $PM_{2.5}$ was influenced mainly by YRDIR (Fig. 5(c-3)) and occasionally by BRIR (Fig. 5(c-2)), but it was also influenced by PRDIR from the 8th to 12th (Fig. 5 (c-4)) with positive contribution of CLDS, which could be attributed to high relative humidity environment over Taiwan Strait. On line 269-270 Although #4 is very near PRDIR, it was

		influenced more by YRDIR (Fig. 5(d-3)-~~5(d-4)~~) and other sources in eastern and northern China rather than three industrial regions since the prevailing wind was mainly northeast wind in January.
-L281: What is the "north" here?	The authors would like to express sources in east and northern China other than BRIR and YRDIR. They have modified that sentence in the revised manuscript.	On line 269-270 Although #4 is very near PRDIR, it was influenced more by YRDIR (Fig. 5(d-3)) and other sources in eastern and northern China rather than three industrial regions since the prevailing wind was mainly northeast wind in January.
-L299: What does "the lower 20 averaged layers" mean here? Does Figure 5 show the daily process contributions averaged in the lower 20 layers? If so, you must clearly state it in the figure caption and/or in somewhere in the manuscript.	The authors have added the information in the manuscript and caption of Fig. 5 and Figure S4.9.	On line 287-291 On Jan 8th to 10th, the negative ZADV indicated the concentration was decreasing in the lower 20 averaged layers, where the daily processes occur, but the concentration gradient was positive ($\frac{\partial PM_{2.5}}{\partial z} > 0$, the concentration of $PM_{2.5}$ from PRDIR was higher at a high altitude than that at a low altitude over Taiwan), which implies the vertical velocity had to be negative, i.e., a downward motion. The caption of Figure 5 **Figure 5: The daily contributions of individual processes averaged over the lower 20 layers to the concentrations of $PM_{2.5}$ in January 2017,**

		a,b,c,d,e,f, and g represent #1, #2, #3, #4, BQ, ZM, and CY, respectively；1, 2, 3, and 4 represent influence of total emissions (base case), BRIR, YRDIR, and PRDIR, respectively
-L310: This is not always true. Could you specify when and where this statement is true?	Thanks the reviewer for pointing out this error. The authors have modified that sentence in the revised manuscript.	On line 300 On most days in winter period, northeast wind prevailed over East Asia.
-L331: What does “nonuniform” mean here? What do you want to mean?	The authors would like to express some process is not consistent in continuous layers. The have used “inconsistent” to replace “nonuniform” in that sentence.	On line 261-264 From Fig. 5(b-1)-(b-4), among the three industrial regions it is apparent that #2 was influenced by both the BRIR and YRDIR, mainly produced through inconsistent HADV, VDIF, ZADV, and CLDS; and removed through AERO and occasional HADV and DDEP processes, and almost unaffected by PRDIR. On line 320-322 Although #1 was slightly influenced by YRDIR, the contribution of different processes from YRDIR on #1 was less and inconsistent (Fig. 8(a-3)). The contribution of different processes from PRDIR to #1 was also inconsistent and even less (Fig. 8(a-4)). On line 370-371 The positive and negative contribution processes were inconsistent below 80 m (layer 4).

-L382: Fig S4.12 --> Fig S4.11?	Thanks the reviewer for finding this error which have been revised already.	On line 372-373 Fig. S4.11 shows that the influence of the three industrial regions on #2, #3, #4, BQ, ZM, or CY were almost ignorable.
-Figure 5, 8. 9: BR --> BQ	Thanks the reviewer for finding this error which have been revised for Figure 5, 8. 9, S4.9, S4.11, and S4.13.	
-Figure 11: #17, 18, 19, 20 should be modified.	Thanks the reviewer for finding this error. The authors have modified the Fig. 11 and Fig. S4.14 already.	
-L445: overestimated <--> underestimated	Thanks the reviewer for finding this error which have been revised already.	On line 434-435 The simulated proportion of nitrate and ammonium in $PM_{2.5}$ during the winter was slightly underestimated, but the simulated K^+, Ca^{2+}, Mg^{2+}, Na^+ was overestimated at BQ, ZM, and CY.

Review #2

General Description		
(1) comments from Reviewers	(2) author's response	(3) author's changes in manuscript.
For final publication, the manuscript should be accepted as is suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)	We sincerely thank the reviewer who provided many valuable comments in previous reviewing processes. We have to say this manuscript could not be (if) accepted without the improvements regarding to those comments. Honestly, we did know this manuscript is not an excellent work but we will continue to study hard on unresolved issues of atmospheric chemistry.	