



Research article

Quantifying PM_{2.5} from long-range transport and local pollution in Taiwan during winter monsoon: An efficient estimation method

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ABSTRACT

From autumn to the following spring, annually, the northeast monsoon transports PM_{2.5} (particles less than 2.5 μm in aerodynamic diameter) from the Asian continent to downstream areas. Naturally, this triggered a question: What are the contributions of PM_{2.5} from long-range transport (LRT) and local pollution (LP) at any downstream location? To answer that question, the present study developed an economical and efficient method that can easily estimate the contribution of PM_{2.5} from LRT (LRT-PM_{2.5}) and PM_{2.5} from LP (LP-PM_{2.5}). The method used PM_{2.5} and meteorological observation data in Taiwan from 2006 to 2015 and a short-term simulation from January to May in 2010. The analysis classified the data into three types of PM_{2.5} source patterns: LRT-Event (high concentration plume at the front edge of southward moving anticyclones/strong northeast wind), LRT-Ordinary (less concentration in common strong northeast wind), and LRT/LP Mix or Pure LP (PM_{2.5} was from both LRT and LP or from only LP under weak northeast wind). During the ten-year period, the average LRT-PM_{2.5} values at the northern tip of Taiwan were 31–39 μg m⁻³, 12–16 μg m⁻³, and 4–13 μg m⁻³ for the LRT-Event, LRT-Ordinary, and LRT/LP Mix or Pure LP patterns, respectively. The 10-year average LRT-PM_{2.5} and LP-PM_{2.5} contributions were approximately 70:30 in northern Taiwan, 50:50 in central Taiwan, and 30:70 in southern Taiwan for the LRT-Event pattern; 60:40 in northern and 40:60 in central and southern Taiwan for the LRT-Ordinary pattern; and 30:70 in northern and 25:75 in central and southern Taiwan for the LRT/LP Mix or Pure LP pattern. Interestingly, LRT-PM_{2.5} peaked in 2013 but has decreased annually since then, whereas LP-PM_{2.5} has roughly decreased in the past ten years.

1. Introduction

PM_{2.5} not only influences the tropospheric oxidants via heterogeneous reactions (Tie et al., 2005) but can also absorb and scatter solar radiation, which impairs visibility (Na et al., 2004), affects the atmospheric radiation budget, and balances and changes the global climate (Hu et al., 2017). Moreover, PM_{2.5} inhaled into the human respiratory organs damages human health and quality of life (Zhu et al., 2011). In recent years, Asian haze episodes often occurred inland or in coastal areas of the Asian continent. The frequency was highest in the spring and winter (Fu et al., 2014; Yang et al., 2016). As the cold high-pressure systems originating from Siberia move southward, the peripheral circulation usually transports the Asian haze to downstream areas, such as Korea, Japan, and Taiwan (Zhang et al., 2015).

To protect citizens' health, on May 14, 2012, the Taiwan Environmental Protection Agency (TEPA) announced PM_{2.5} daily and

annual standards of 35 μg m⁻³ and 15 μg m⁻³, respectively. However, the announcement was just a blind regulation based on those of other countries. There is no plan for implementing a practical approach. During the summertime, the PM_{2.5} level is low and the pressure from legislators is minimal. However, the PM_{2.5} level is elevated during the wintertime due to the impact of the Asian haze (Chuang et al., 2008a,b; Yang et al., 2018). The public then asks the TEPA and the local Environmental Protection Bureaus (EPB) to find a solution immediately. However, the TEPA or local EPBs cannot control emissions in the distant Asian continent. It is helpful if they can understand the individual contributions of PM_{2.5} from long-range transport (LRT) and local pollution (LP) so that they can set a goal to achieve or evaluate the control measures in their approach.

In recent years, more and more studies tried to quantify the contributions of LP and LRT pollutants. Two methods were most involved: TS (Trajectories Statistics) and CTM (Chemical Transport Modeling).

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The TS method is to count the frequency of backward-trajectories in passing through grids. The more frequent the trajectory passed a grid, the more contributed from emissions in that grid. Trajectory information usually results from the meteorology models like MM5 (Mesoscale Model version 5, [Dudhia, 1993](#)) or WRF (Weather Research and Forecasting, [Skamarock and Klemp, 2008](#)), which provides wind fields to a trajectory model like Hysplit model ([Stein et al., 2015](#)). For example, [Pawar et al. \(2015\)](#) utilized a back-trajectory climatology analysis of air masses to assess the contribution of LRT Particulate matter at the Indian Institute for Science Education and Research (IISER) Mohali in north-west Indo-Gangetic Plain. They classified the air masses into six clusters, which represented LP and different LRT sources. Very similar methods have been applied for quantifying the contribution of LRT PM₁₀ for South Hessen in Germany ([Garg and Sinha, 2017](#)), LRT PM_{2.5} during a short severe haze in Beijing, China ([Yang et al., 2018](#)), LRT PM_{2.5} in the Ordos region, Inner Mongolia, China ([Khuzestani et al., 2017](#)), LRT PM₁₀ in Tibetan Plateau Uplift Area: Xining, China ([Xin et al., 2016](#)), LP and LRT PM_{2.5} in the Po Valley, Italy ([Squizzato et al., 2012](#)), etc. Another method applied the CTM, which generally contains the BFM (Brute Force Method) and the AM (Apportionment Method) methods. The BFM method adapted a simple concept of denoting the difference between a normal simulation and another simulation that zeros out emissions of a specific region as the contribution from that region. This method is direct and widely applied ([Marmur et al., 2005](#); [Burr and Zhang, 2011](#); [Chen et al., 2014](#)). In addition, the BFM method is often extended to assess the effects of some specific regulations or control measurements ([Li et al., 2017a](#)). AM method is more complicated and incorporates the source apportionment modules into CTM. For example, [Skylakou et al. \(2014\)](#) applied a regional chemical transport model called PMCAMx ([Fountoukis et al., 2011](#)) that used the particulate matter source apportionment technique (PSAT, [Wagstrom et al., 2008](#)) to estimate PM_{2.5} from the local, mid-range (50–500 km), and long range transport (> 500 km) emissions. [Kwok et al. \(2013\)](#) also developed an integrated source apportionment method (ISAM) and implemented it into CMAQ model ([Byun and Schere, 2006](#)). However, the modeling components such as the emissions, meteorological modeling, chemical mechanisms, and numerical deviations all contain uncertainty. Moreover, long periods of simulation requires very expensive computing resources. Therefore, above studies could not be applied for long-term period study and it is necessary to build an economical and efficient method for estimating the contributions of PM_{2.5} from LRT and LP.

Here, a review of the literature on the quantitative analysis of transboundary transport from the Asian continent to Taiwan is provided. [Chang et al. \(2000\)](#) applied CTM to simulate the contribution of LRT from East Asia to Taiwan for six episodes in 1993. The sulfate deposition from LRT ranged from 9% to 45% and the nitrate deposition ranged from 6% to 33%. For all types of LRT source patterns, the impact was the highest when the northeast wind came from the Asian continent. [Lin et al. \(2004\)](#) examined the meteorological and air quality data from November 1999 to May 2000 and November 2000 to May 2001. They classified the wintertime LRT into dust storm transport, front transport, and background air mass transport with mean PM₁₀ concentrations at the northern tip of Taiwan of 127.6, 85.0, and 32.8 $\mu\text{g m}^{-3}$, respectively. These three types of LRT episodes accounted for 25.2% of the 14-month research period. Meanwhile, LP accounted for 71.7% and a mean PM₁₀ concentration of 47.4 $\mu\text{g m}^{-3}$ (Data Missing accounts for 3.1%). On average, LRT contributed 50%–75% of the PM₁₀ abundance for the LP episodes in northern Taiwan during winter and spring. [Chuang et al. \(2008a\)](#) connected LRT events with the push from high-pressure systems. [Chuang et al., 2008b](#) further used the CMAQ ([Byun and Schere, 2006](#)) model to simulate an anticyclone and the Asian haze plume simultaneously arrived in northern Taiwan and showed the combination nearly dominated the PM_{2.5} level in Taipei city. The local pollutants in Taipei were rapidly diluted and pushed southward by the LRT air masses. In addition, [Chen et al. \(2014\)](#) also

utilized the CMAQ to simulate the contributions of various sources of PM_{2.5} in Taiwan in 2007. They found that the contributions from direct LRT (LRT precursors directly forming PM_{2.5}) and indirect LRT (LRT precursors interacting with local precursors forming PM_{2.5}) were 27% and 10%, respectively. In other words, the total LRT contributed 37% for a whole year. Among the seasons, LRT contributed more during autumn and winter, with contributions of 39% and 41%, respectively. [Wang et al. \(2016\)](#) adapted the AGAGE method ([O'Doherty et al., 2001](#)) to assess the Asian haze events. First, they used the AGAGE method to process PM_{2.5} observations at the northern tip of Taiwan. Next, they applied backward trajectories with the HYSPLIT model ([Draxler and Rolph, 2013](#)) to determine whether the air currents of the LRT events were from the Asian continent. Furthermore, they used the spatial distribution of AOD to assess the occurrence of the events since AOD strongly correlates with PM_{2.5} concentrations. Usually, the spatial peak of AOD began to move from the Asian continent to Taiwan three days in advance. From the above reports, although many discussed the impact of LRT on Taiwan, they lack an economical and efficient method for quantifying the contribution of LRT or LP for any place in Taiwan over a long period, such as ten years. Then the policy makers can understand the trend of LRT-PM_{2.5} and LP-PM_{2.5} and be certain about the effects of control measures exerted in the past.

To estimate the contributions of PM_{2.5} from LRT and LP in Taiwan, this study developed an economical and efficient method. First, this study classified three types of source patterns: episodes that are high PM_{2.5} concentration events from LRT high concentration plume at the front edge of southward-moving anticyclones/strong northeast wind (LRT-Event), episodes that are not high PM_{2.5} concentration events under common LRT strong northeast wind (LRT-Ordinary), and PM_{2.5} episodes that are mix of LRT and LP or are pure LP (LRT/LP Mix or Pure LP) under a weaker prevailing northeast wind. Accordingly, LRT-PM_{2.5} at the northern tip of Taiwan was estimated for the above three source patterns. Next, this study used air quality modeling to determine the proportion of change (PoC) of PM_{2.5} from the northern tip of Taiwan to any downstream area. Then, the contribution of LRT was estimated using PoC and LRT-PM_{2.5} from 2006 to 2015. The difference between the total observed PM_{2.5} and LRT-PM_{2.5} is the LP-PM_{2.5} (PM_{2.5} from LP, i.e. the emissions from the island of Taiwan). The details of the estimation method are described in section 2.3. In addition to quantifying LRT-PM_{2.5} and LP-PM_{2.5}, this study also discusses the trend in LRT-PM_{2.5} and LP-PM_{2.5} over ten years. The results from this study are very valuable, especially to the TEPA and local EPBs. The long-term trend/projections is essential for the policy makers to evaluate the trend of emissions, effects of control measures in the past and upper limit of their control strategy, etc.

2. Methods

2.1. Geographical location of meteorological and air quality observation sites

[Fig. 1](#) shows that Taiwan is located in the West Pacific. When the Siberia cold high-pressure systems move to East Asia, it brings a prevailing northeast wind to East Asia and the West Pacific. From late autumn to the following spring, the northeast monsoon transports pollutants from the Asian continent to downstream areas. The WL station (#1 in [Fig. 1](#)) is the northernmost station that is first exposed to LRT air masses. In addition to the WL station, there are four other air quality stations nearby, including KL (#2 in [Fig. 1](#)), TS (#3 in [Fig. 1](#)), IL (#4 in [Fig. 1](#)), and DS (#5 in [Fig. 1](#)). We will discuss the correlation relationship between WL and those four stations and prove that the LRT haze plume is a nearly uniform air mass with a horizontal scale that can cover the east-west width of Taiwan. These five stations are called the upstream stations in the following narrative. This study aims to understand the impact of LRT on any location in Taiwan. Eight stations: SJ (#6 in [Fig. 1](#)), TY (#7 in [Fig. 1](#)), HC (#8 in [Fig. 1](#)), CM (#9 in [Fig. 1](#)), JI

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