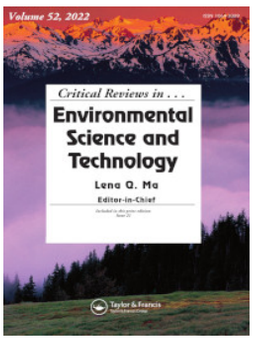

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Source-receptor relationship of transboundary particulate matter pollution between China, South Korea and Japan: Approaches, current understanding and limitations

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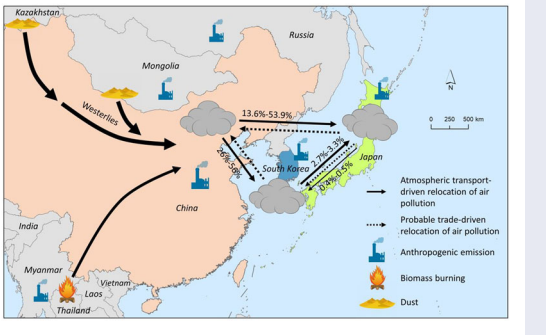
ABSTRACT



Transboundary particulate matter (PM) pollution in Northeast Asia has raised tremendous concerns in China, South Korea, and Japan, leading to a proliferation of publications in recent years. This article summarizes the existing knowledge on the source-receptor relationship (SRR) of transboundary PM pollution between China, South Korea, and Japan with a focus on approaches, current understanding, and limitations. We found that eastern-, northern- and northeastern China are the most contributing source areas within China to PM pollution in both South Korea and Japan, but it remains debatable whether China contributes more PM pollution to South Korea or Japan than those countries themselves. Considerable differences have been reported in the estimations of China's relative contributions to receptor countries, and higher estimations were usually obtained from studies that focused on short time periods, used outdated emission inventories, and had few or no international collaboration. China's contributions range from 26% to 56% for South Korea and from 13.6% to 53.9% for Japan if the analysis periods are limited to one or several years and the receptors are limited to an entire country. We attributed these differences to the discrepancies in the analysis periods, analytical approaches, modeling settings, definitions of source and receptors, and international collaboration. We also demonstrated that current SRR studies face the challenges from data quality issues in PM measurement data and emission inventories, limited temporal and spatial scales in modeling, and limited analytical perspectives concerning the allocation of environmental responsibilities. Suggestions for future research are provided to address these challenges.

Abbreviations: NEA: Northeast Asia; PM: particulate matter; $PM_{2.5}$: fine particulate matter; MICS-Asia: Model Inter-Comparison Study for Asia; SRR: source-receptor relationship; HYSPLIT: Hybrid Single-Particle Lagrangian Integrated Trajectory Model; NOAA: the United States National Oceanic and Atmospheric Administration; PSCF: Potential Source Contribution Function; CWT: Concentration Weighted Trajectory; RTWC: Residence Time Weighted Concentration; CTM: Chemical Transport Model; WRF-Chem: Weather Research and Forecasting model coupled with Chemistry; CAMx: Comprehensive Air Quality Model with Extensions; CMAQ: Community Multiscale Air Quality Modeling System; NAQPMS: Nested Air Quality Prediction Modeling System; RAQM2: Regional Air Quality Model 2; TSSA: tagged species source apportionment algorithm; PSAT: particle source apportionment technology; NIER: Korean National Institute of Environmental Research; NASA: the United States National Aeronautics and Space Administration; MEIC: Multi-resolution Emission Inventory for China; INTEX-B: Intercontinental Chemical Transport Experiment-Phase B; REAS:

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Supplemental data for this article can be accessed on the publisher's website.

Regional Emission inventory in Asia; CAPSS: Clean Air Policy Support System; LTP: Joint Research Project for Long-range Transboundary Air Pollutants in Northeast Asia

KEYWORDS Transboundary particulate matter pollution; source area; source-receptor relationship; Northeast Asia; data quality

HANDLING EDITORS Daniel Tsang and Robert Letcher

1. Introduction

Countries in Northeast Asia (NEA), including China, South Korea and Japan, have suffered from severe particulate matter (PM) pollution for decades. The extremely high PM concentration in NEA and its adverse health impacts have raised tremendous public concern in NEA (Li et al., 2016; Taniguchi et al., 2017). Driven by the increasing concern, many efforts have been devoted to studying the transboundary PM pollution in NEA with a focus on the source areas and source contributions of transboundary PM pollution, leading to a proliferation of publications in recent years (C. Bae et al., 2020; S. Lee et al., 2019; Li et al., 2018). Despite an increasing number of studies being conducted, existing knowledge on the source areas of transboundary PM pollution and their contributions in recent years has not been systematically and comprehensively reviewed. A review conducted in 2014 summarized studies on the source-receptor relationship (SRR) of sulfate and nitrate in East Asia (Qu et al., 2016); however, no study has provided a systematic and comprehensive review on the SRR of lumped PM pollution. Consequently, some important questions of great interest to the general public and policy makers in NEA remain unanswered: Which parts of East Asia are the sources of the transboundary PM pollution in NEA and how much PM pollution does each source area contribute to the receptors? Has a consensus been reached on the source areas of transboundary PM pollution and their contributions to PM pollution in NEA? How different are the extant findings and why?

Unfortunately, the lack of answers to the above questions and an incomplete and unclear understanding of the SRR of transboundary PM pollution among NEA countries, combined with the growing public concern, have led to the recurrence of finger-pointing, diplomatic disputes, and potential conflicts among NEA countries. For example, the South Korean Ministry of Environment claimed that China was responsible for 70%–80% of $PM_{2.5}$ pollution in Seoul (Ock, 2019). In response, Chinese environmental officials refuted the accusation and asserted that China would not take the blame for South Korea's own problem (Zhang, 2018). Clearly, there is an urgent need to summarize the existing knowledge in the literature to gain a comprehensive and systematic understanding of the source areas and source contribution of transboundary PM pollution in NEA. A better understanding and a more extensive knowledge base would help build consensus among countries and create a closer coalition for reducing transboundary PM pollution in the future.

Therefore, this paper aims to provide an up-to-date review of existing knowledge on the SRR in current transboundary PM pollution research in NEA, clearing up doubts and building clarity from the literature by summarizing findings on source areas of transboundary PM pollution and their contributions of the transboundary PM pollution to the receptors. We first provide summaries of the identification of source areas (Sec. 2) and quantification of source contribution (Sec. 3) of the transboundary PM pollution in NEA. In each summary, we provide an overview of the methods employed in current transboundary PM pollution studies to identify the source areas and quantify the source contribution. We highlight the uncertainties in these methods that plague the evidence base, emphasizing the need for critical thinking in reviewing the existing evidence. We compare extant findings on the relative contributions by source countries to the receptor countries and offer explanations regarding the inconsistencies between these findings. We then

discuss the challenges facing current SRR studies in NEA (Sec. 4). Finally, we offer a set of suggestions (Sec. 5) to help future SRR research address the challenges.

2. Source areas of transboundary PM pollution in Northeast Asia

One of the main challenges in improving the air quality in the region is to identify the spatial origins of the pollution to ensure that air quality plans target the appropriate source areas for effective results. Therefore, before implementing any mitigation strategies and policy measures, the source areas that contribute most to pollutant concentration levels should be identified. This is particularly true for transboundary PM pollution that is transported from remote areas. However, accurate identification of source areas for transboundary PM pollution is not an easy task. There are several approaches for identifying the source areas of transboundary PM pollution, and each method has its advantages and disadvantages. Naturally, the credibility of the conclusions on the source areas depends on the strength of these approaches.

2.1. Approaches for identifying source areas of transboundary PM pollution

The most frequently used methods for identifying the source areas of transboundary PM pollution in NEA can be classified into the following five categories.

1. **Back trajectory model**. The early development and application of back trajectory models can be traced back to the 1940s (Stein et al., 2015). This model can be used to understand the spatial origins of air masses over the span of a few days, as it is able to trace the paths of air parcels back in time and space and indicate where the air parcels have traveled before reaching the receptor sites. This is done by reconstructing transportation and dispersion routes of air parcels from a particular location using regional meteorological data (wind fields) and Lagrangian functions (Tang et al., 2007). The most widely used tool for back trajectory modeling in NEA is the Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT, http://ready.arl.noaa.gov/HYSPLIT.php) developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (NOAA) (Draxler & Hess, 1998). For example, in a study of high-PM_{10} episodes in Seoul of South Korea, a 72-h back trajectory analysis was conducted using the HYSPLIT model that led to the conclusion that high-PM_{10} pollution in Seoul originated from severely polluted regions in northeastern China (Oh et al., 2015).

A typical back trajectory model, such as the HYSPLIT model, is very helpful in providing supportive and indirect evidence for identifying the source areas of transboundary PM pollution; however, it is worth noting that there are limitations and uncertainties in using this model. First, the model cannot differentiate transboundary pollution from locally emitted air pollution. The strength of this model is that it shows the potential transport pathway of air parcels. However, the results of back trajectory modeling can never demonstrate with certainty that the air pollution originates from the source area. The cause of the high levels of pollution at the receptor sites might be transboundary PM pollution from remote areas but can also be local emission sources. Moreover, the potential pollution source area is likely also a receptor (Chen & Taylor, 2018). Second, the back trajectory models suffer from accuracy problem. A sensitivity study on back trajectory modeling shows that the average horizontal distance deviations are approximately 100 km for a 24 hours' model run and between 200 and 700 km for a 96 hours' model run (Rolph & Draxler, 1990). Thus, the back trajectory model cannot link the transboundary PM pollution definitely to the source areas; it can only provide weak evidence as to the potential source areas.

2. **Potential Source Contribution Function (PSCF)**. PSCF is a widely used statistical analysis method based on back trajectory modeling that can deduce potential source areas (Ashbaugh et al., 1985; Heo et al., 2009; B. M. Kim et al., 2016). PSCF assumes that if the trajectory of an air parcel passes a geographical grid cell (i, j), the air parcel will collect the pollutants emitted from cell (i, j) and be transported along the trajectory to the receptor site. The PSCF measures the probability that a source is located at cell (i, j). If n_{ij} represents the number of times that the trajectories pass through cell (i, j) and m_{ij} is the number of times that the trajectories pass through cell (i, j) when the pollutant concentrations at the receptor site are higher than a pre-specified threshold, then the PSCF value at cell (i, j) can be calculated using Eq. (1).

$$PSCF_{i,j} = \frac{m_{ij}}{n_{ij}} \tag{1}$$

Cells with high PSCF values are likely to be source areas, as high PSCF values indicate that the air parcels have crossed those cells, presumably collected and transported the emitted pollutants to the receptor site, and, thus, increased the pollutant concentration levels at the receptor site (Han et al., 2008). For example, a study that aimed to identify the source area of long-range transport of $PM_{2.5}$ species in Niigata of Japan used PSCF and concluded that the major source region of pollution due to biomass combustion was northeastern China in autumn but local area in Japan in other seasons (Li et al., 2018). Other similar methods exist that use the trajectory statistics; these methods include the Concentration Weighted Trajectory (CWT) (Seibert et al., 1994) and the Residence Time Weighted Concentration (RTWC) (Stohl, 1996). However, these methods have been rarely used in transboundary PM pollution studies in NEA.

As the PSCF method is based on back trajectory models, it has the limitations and uncertainties of the back trajectory model. However, compared with the simple back trajectory model, which only provides indicative results of the source areas, PSCF provides a relatively convincing statistical justification for the potential source areas by combining the pollutant concentrations at the receptor site and back trajectories.

3. **Chemical composition analysis**. Chemical composition analysis is based on the assumption that the chemical composition of air pollutants is unique in different geographical areas, and this unique chemical composition of air pollutants can be used as a fingerprint to identify the source areas of transboundary PM pollution (Chen & Taylor, 2018; Inomata et al., 2016). The closer the fingerprint in the receptor region to the fingerprint in source areas, the more likely the transboundary PM pollution is from the source areas (Chen & Taylor, 2018). The commonly used fingerprints in transboundary PM pollution studies in NEA include sulfur isotopic ratios (Kim et al., 2018; Toshiaki et al., 2012), isotopic compositions of sulfur and boron (Sakata et al., 2013), and the ratio of nitrate to non-sea-salt sulfate (Itahashi et al., 2018). For example, an analysis of the ratio of nitrate to non-sea-salt sulfate from 2001 to 2015 in East Asia showed that SO_2 emissions in China had a significant impact on the wet deposition of sulfate in South Korea and Japan (Itahashi et al., 2018).

Chemical composition analysis is a relatively convincing tool for identifying the source areas. However, it can only provide indirect evidence regarding the source areas of transboundary PM pollution. The credibility of chemical composition analysis depends on the uniqueness of the chemical composition of air pollutants in different geographical areas; however, the chemical composition of air pollutants changes over time (Thiemens, 2006). Moreover, the data required for chemical composition analysis are samples collected in various sampling points; thus, the accuracy and extent of the identified source areas of transboundary PM pollution are limited by the number of samples and the locations of the sampling sites.

4. **Eulerian Chemical Transport Models (CTMs)**. As opposed to Lagrangian models, such as the HYSPLIT model, which describe the motion of air parcels, Eulerian models can describe a variety of coupled physical and chemical processes of air pollutants over time, including emission, transport, chemical transformation, and deposition (Jacob, 1999). An Eulerian CTM is usually built using an Eulerian grid model based on a fixed coordinate system. Driven by meteorological data and pollutant emission inventory data, the model simulates chemical reactions using a chemical reaction database and calculates the resulting changes in pollutant concentrations for each grid in each time step (Chen & Taylor, 2018). Eulerian CTMs that have been used in transboundary PM pollution studies in NEA include the following: Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) (H. J. Lee et al., 2019b), Comprehensive Air Quality Model with Extensions (CAMx) (Oh et al., 2020; Wang et al., 2016), GEOS-Chem (Jeong & Park, 2019; Lee et al., 2017), Community Multiscale Air Quality Modeling System (CMAQ) (C. Bae et al., 2020; J.-H. Kim et al., 2016), Nested Air Quality Prediction Modeling System (NAQPMS) (Li et al., 2014), and Regional Air Quality Model 2 (RAQM2) (Kajino et al., 2013).

The ability of the Eulerian CTM to capture the physical and chemical processes of air pollutants over time and space enables the identification of source areas in a direct and accurate manner as well as the quantitative evaluation of the transport of transboundary air pollutants from source areas to receptor regions, that is, the SRR (Qu et al., 2016). Details on various methods for analyzing the SRR of transboundary PM pollution in NEA are elaborated in Sec. 3.1. Although most Eulerian CTMs suffer from incomplete emission inventories and gaps in the representation of atmospheric chemistry processes, they are able to provide the most convincing analysis concerning the identification of source areas of transboundary PM pollution, as these models directly link the source areas to the receptor regions.

5. **Statistical models**. By establishing the statistical relationship between the air pollution in source and receptor regions, statistical models can provide statistical evidence linking the receptor regions to potential source areas. For example, a panel data analysis of air pollutant measurements in five cities in China (Taiyuan, Lanzhou, Zibo, Beijing and Jinan) and major cities in South Korea from 2005 to 2008 showed that if China's Air Pollution Index exceeded 100, an increase in China's air pollution was associated with worsened air quality in South Korea (Park & Tak, 2012). Another regression analysis of PM_{10} concentration measurements in Seoul of South Korea, Shanghai and Beijing of China from 2014 to 2016 showed that a 1-unit increase in PM_{10} concentration in Beijing and Shanghai on the previous day was associated with a 0.13- and 0.132-unit increase in PM_{10} concentration in Seoul (Park et al., 2020). Further analysis using the Granger causality test concluded that the PM_{10} concentrations in Beijing and Shanghai were Granger causes of PM_{10} concentrations in Seoul (Park et al., 2020).

Statistical models provide plausible evidence for identifying potential source areas. However, a well-known limitation of all statistical regression models is that the results of regression analyses can only indicate association (sometimes called correlation), rather than a causal relationship. To establish a causal relationship that the air pollution in source areas causes increased air pollution levels in receptor regions, it is generally accepted that at least three criteria should be met: (1) association: there are statistically significant associations between air pollution from potential source areas and air pollution in receptor regions; (2) temporal precedence: the change in air pollution from the potential source areas occurs before the changes in air pollution in receptor regions; (3) no other plausible explanations: there are no confounding variables that can explain the association (Holland, 1986; Sekaran & Bougie, 2016). The study by Park and Tak (2012) only

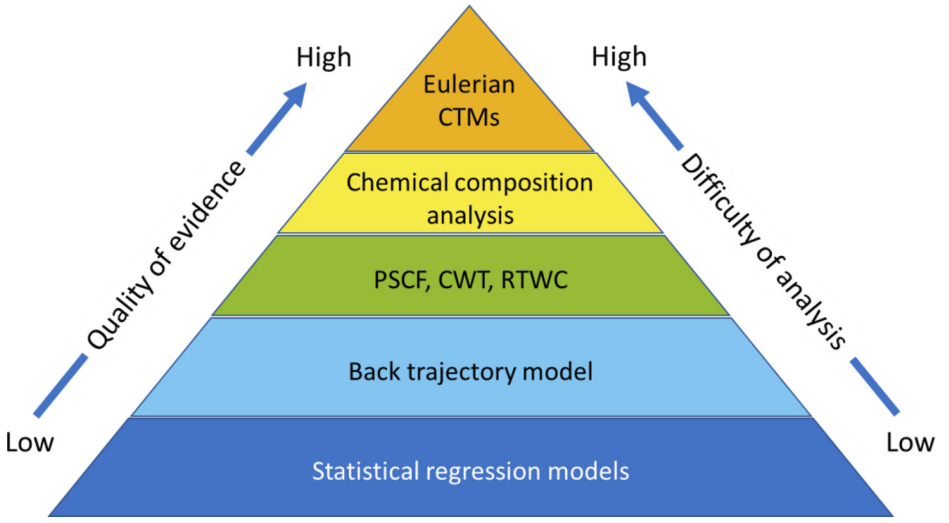

Figure 1. Pyramid of evidence-based source areas identification of transboundary PM pollution using different methods. The levels of the pyramid reflect the increasing quality of evidence produced by the method and increasing difficulty of performing analysis using the method from bottom to top.

satisfied the first criterion, the study by Park et al. (2020) presumably met the first and second criteria. However, no statistical modeling studies have fulfilled the third criterion. In fact, in addition to the transport of transboundary PM pollution, the frequent high-pressure systems residing in NEA can cause covariation of air pollution in source and receptor regions. In other words, synoptic meteorology can explain the associations between air pollution in different cities in NEA. Therefore, statistical models are limited in providing convincing evidence for identifying source areas.

As discussed above, the five categories of approaches for identifying the source areas of transboundary PM pollution, namely, the back trajectory model, PSCF, chemical composition analysis, Eulerian CTM and statistical model, have quite different strengths and capabilities in producing convincing results for the identification of source areas. Inspired by the idea of evidence-based medicine in the field of health science, which classifies evidence based on its quality and epistemological strength (Eddy, 2005; Howick et al., 2011), the five categories of methods are classified into five levels in producing high quality evidence based on the respective epistemological strength of the methods (Table S1). As shown in Table S1, Eulerian CTMs provide evidence of the highest quality, as the models are able to directly link the pollutant emissions from the source areas to the receptor regions, whereas the rest can only provide "circumstantial" evidence that indirectly link the source areas to the receptor regions. However, Eulerian CTMs require more computational time/power and more datasets than other methods. Moreover, the quality of these datasets heavily affects the accuracy of the modeling results by CTMs. The evidence pyramid in Figure 1 illustrates the different levels of evidence quality in identifying the source areas of transboundary PM pollution using these methods and the different levels of difficulty in performing analysis using these methods (difficulties in collecting high-quality data and the computational time/power required in implementing these methods).

2.2. Source areas of transboundary PM pollution in Northeast Asia

The previous discussion on the approaches for identifying source areas of transboundary PM pollution shows that Eulerian CTMs can provide the highest quality of evidence of identified source areas of transboundary PM pollution, followed by chemical composition analysis, PSCF, back trajectory models, and statistical models. Therefore, in order to obtain high quality and credible

results of identified source areas of transboundary PM pollution, we primarily rely on analyses that used the Eulerian CTMs. Results obtained from other approaches only serve as supplementary evidence.

We then used Google Scholar and Web of Science, the two most frequently used reference databases in academic research, to search relevant studies. We further conducted backward search to identify relevant work cited by the pertinent articles and forward search to identify relevant works that have cited the pertinent articles. Two inclusion criteria were applied to identify the most pertinent articles: (1) the study used the Eulerian CTMs with model evaluations showing good agreement with the observation data; (2) the study investigated the SRR of transboundary PM pollution between China, South Korea, and Japan in NEA. Finally, we found 18 records that met the two inclusion criteria. From each of the 18 studies, we extracted information on modeling time period, pollutants modeled, source attribution methods, receptor regions, source areas, emission inventories, and their relative contributions.

Table 1 shows detailed information on the studies of the SRR of transboundary PM pollution with South Korea as the receptor. Evidently, China is the primary source area of transboundary PM pollution for South Korea (Lee et al., 2017; Yim et al., 2019), followed by North Korea (Oh et al., 2020), Japan (Choi et al., 2019; NASA & NIER, 2017), and Mongolia and Russia as a whole (J.-H. Kim et al., 2016; Li et al., 2014). In particular, as China has a much larger territory area than South Korea and Japan, additional information on China's provinces and their relative contributions to PM pollution in South Korea are summarized in Table S2, providing further information on the source areas within China responsible for transboundary PM pollution transported to South Korea. As can be inferred from Table S2, at the sub-region level, eastern China and northern China, including the Beijing–Tianjin–Hebei region and the Shanxi, Shandong, Henan, Jiangsu, Anhui, and Zhejiang provinces, are the primary source areas of transboundary PM pollution transported to South Korea (Lee et al., 2017; Oh et al., 2020). North Korea and northeastern China, including the Liaoning, Jilin, and Heilongjiang provinces, are the second most contributing source areas (Choi et al., 2019; NASA & NIER, 2017). The rest of the source areas include Japan, Russia, Mongolia, and the rest of China (J.-H. Kim et al., 2016; Li et al., 2014). Figure 2(A) presents a map of the source areas in NEA that contribute transboundary PM pollution to South Korea.

Similar to South Korea, China is the primary contributing source area of transboundary PM pollution to Japan, followed by the Korean Peninsula, and Mongolia and Russian as a whole (Table 2) (Ikeda et al., 2014, 2015; Li et al., 2014). Table S3 shows that, at the sub-region level, northern-, eastern- and northeastern China are the primary source areas of transboundary PM pollution to Japan. The Korea Peninsula is the secondary source area, and the rest of the source areas include Russia, Mongolia, and the rest of China. A map of the source areas of transboundary PM pollution in NEA that contribute to pollution in Japan is shown in Figure 2(B).

Regarding the source area of transboundary PM pollution transported to China, the contributions of South Korea and Japan are insignificant (Li et al., 2014). Studies have shown that the major source areas for transboundary PM pollution to China mainly include the Indo-China peninsula in Southeast Asia (J. Li et al., 2017; Yadav et al., 2017), Mongolia (Zhang et al., 2010), and Central Asia (Jiang et al., 2013).

As can be inferred from Figure 2, the locations of the primary source areas for South Korea and Japan share a similar pattern in that their source areas are both located in their upwind proximity regions. Studies show that a typical transport pathway for transboundary PM pollution in NEA is that a low-pressure system over the Asian continent (e.g., China, Mongolia) induces ascending air currents, lifting PM and relevant precursors into the upper troposphere, then the strong persistent westerlies in this region drive the flow of pollutants from the Asian continent to the Pacific (S. Lee et al., 2011; Qu et al., 2016; Singh et al., 2009). When the outflow of air pollutants arrives at the region of South Korea and Japan, the high pressure residing over these

Table 1. Studies of the source-receptor relationship of transboundary PM pollution to South Korea as the receptor region.

Entry	Time period	Pollutants	Receptor	Method	Emission inventory	Source countries and their relative contributions
(C. Bae et al., 2020)	2012–2016	$PM_{2.5}$	Seoul, South Korea	Brute-force method using CMAQ	China (MICS-Asia 2010), South Korea (CAPSS 2010), other regions (MICS-Asia 2010)	China (42%)
(M. Bae et al., 2020)[a]	2010–2017	$PM_{2.5}$	South Korea	Brute-force method using CMAQ	China (MICS-Asia 2010), South Korea (CAPSS 2010), other regions (MICS-Asia 2010)	China (56%)
(Oh et al., 2020)[b]	Jan. 8–16 2019	$PM_{2.5}$	Seoul, South Korea	Tagged tracer method using CAMx-PSAT	China (MEIC 2010), South Korea (CAPSS 2011), other regions (REAS v1.1 2008)	China (64.4%), South Korea (24.4%), North Korea (9.8%)
(Oh et al., 2020)[b]	Jan. 8–16 2019	PM_{10}	Seoul, South Korea	Tagged tracer method using CAMx-PSAT	China (MEIC 2010), South Korea (CAPSS 2011), other regions (REAS v1.1 2008)	China (57.9%), South Korea (31.1%), North Korea (9.7%)
(LTP, 2019)	2017	$PM_{2.5}$	Seoul, Daejeon and Busan in South Korea	WRF-CAMx, WRF-CMAQ	China (emission data in 2017), South Korea and Japan (emission data in 2015)	China (32.1%), South Korea (51.2%), Japan (1.5%)
(Yim et al., 2019)	2010	$PM_{2.5}$	South Korea	Zero-out contribution method using CMAQ	HTAP_v2 2010	China (54.2%), South Korea (29.4%), Japan (0.4%)
(Choi et al., 2019)	Jun. 1–7 2016	$PM_{2.5}$	South Korea	Sensitivity analysis using GEOS-Chem adjoint model	KORUS v2.0 2015	China (26%), South Korea (57%), North Korea (9%), Japan (2%)
(Choi et al., 2019)	May 25–28 2016	$PM_{2.5}$	South Korea	Sensitivity analysis using GEOS-Chem adjoint model	KORUS v2.0 2015	China (68%), South Korea (26%), North Korea (3%), Japan (0%)
(Choi et al., 2019)	May 17–19 2016	$PM_{2.5}$	South Korea	Sensitivity analysis using GEOS-Chem adjoint model	KORUS v2.0 2015	China (59%), South Korea (34%), North Korea (4%), Japan (0%)
(Choi et al., 2019)	May 10–16 2016	$PM_{2.5}$	South Korea	Sensitivity analysis using GEOS-Chem adjoint model	KORUS v2.0 2015	China (38%), South Korea (50%), North Korea (6%), Japan (2%)
(NASA & NIER, 2017)	May 10–Jun. 10 2016	$PM_{2.5}$	Seoul, South Korea	Sensitivity analysis using GEOS-Chem adjoint model	KORUS v1	China (between 34% and 39%, the publication did not disclose an exact number), South Korea (52%), North Korea (9%), Japan (<5%)
(B. Kim et al., 2017)[c]	Feb. 23–27 2014	$PM_{2.5}$	Seoul, South Korea	Tagged tracer method using CAMx-PSAT	China (INTEX-B 2006), South Korea (CAPSS 2007), other regions (INTEX-B 2006)	China (70.4%), South Korea (20.5%), North Korea (6.7%), Japan (0.2%), Mongolia & Russia (0.05%)

(*continued*)

Table 1. Continued.

Entry	Time period	Pollutants	Receptor	Method	Emission inventory	Source countries and their relative contributions
(B. Kim et al., 2017)[c]	Feb. 23–27 2014	PM_{10}	Seoul, South Korea	Tagged tracer method using CAMx-PSAT	China (INTEX-B 2006), South Korea (CAPSS 2007), other regions (INTEX-B 2006)	China (64%), South Korea (27.7%), North Korea (6.1%), Japan (0.2%), Mongolia & Russia (0.05%)
(Lee et al., 2017)[d]	May of 2009–2013	$PM_{2.5}$	Seoul, South Korea	Sensitivity analysis using GEOS-Chem adjoint model	NIER/KU-CREATE	China (69.8%), South Korea (15.1%), North Korea (4%)
(H. C. Kim et al., 2017)	2014	PM_{10}	Seoul, South Korea	Brute-force method using CMAQ	South Korea (CAPSS 2007/10), Other regions (INTEX-B 2006, MICS-Asia 2010)	All regions outside Seoul (60.5%)
(J.-H. Kim et al., 2016)	Feb. 2014	PM_{10}	Seoul, South Korea	Decoupled Direct Method using CMAQ	China (MEIC 2010), South Korea (CAPSS 2011), other regions (REAS 2008)	China (48.2%), South Korea (25.8%), North Korea (12.4%), Mongolia (<1.7%)
(Li et al., 2014)	2010	PM_{10}	Korea peninsula	Tracer-tagged method using NAQPMS	China (2010 data from Zhao et al. (2013)), Other regions (REAS v2.1 2008)	China (26%), South Korea (41.8%), Japan (0.5%), Russia & Mongolia (<0.5%)
(Koo et al., 2008)	Jan. 2007	PM_{10}	Seoul, South Korea	Zero-out contribution using CMAQ	South Korea (CAPSS 2003), other regions (ACE-Asia 2000)	All regions outside Seoul (approx. 40%–80%)

[a]Two estimations of China's relative contributions over South Korea were calculated using two different modeling grid resolutions (M. Bae et al., 2020). The relative contribution of 56% was estimated using a higher modeling grid resolution and is believed to be more accurate (M. Bae et al., 2020).
[b]The detailed percentages of relative contributions for various source regions were calculated based on data obtained via email correspondence with the authors of Oh et al. (2020).
[c]The detailed percentages of relative contributions for various source regions were calculated based on data obtained via email correspondence with the authors of B. Kim et al. (2017).
[d]The detailed percentages of relative contributions for North Korea and Liaoning area in China were obtained via email correspondence with the authors of Lee et al. (2017).

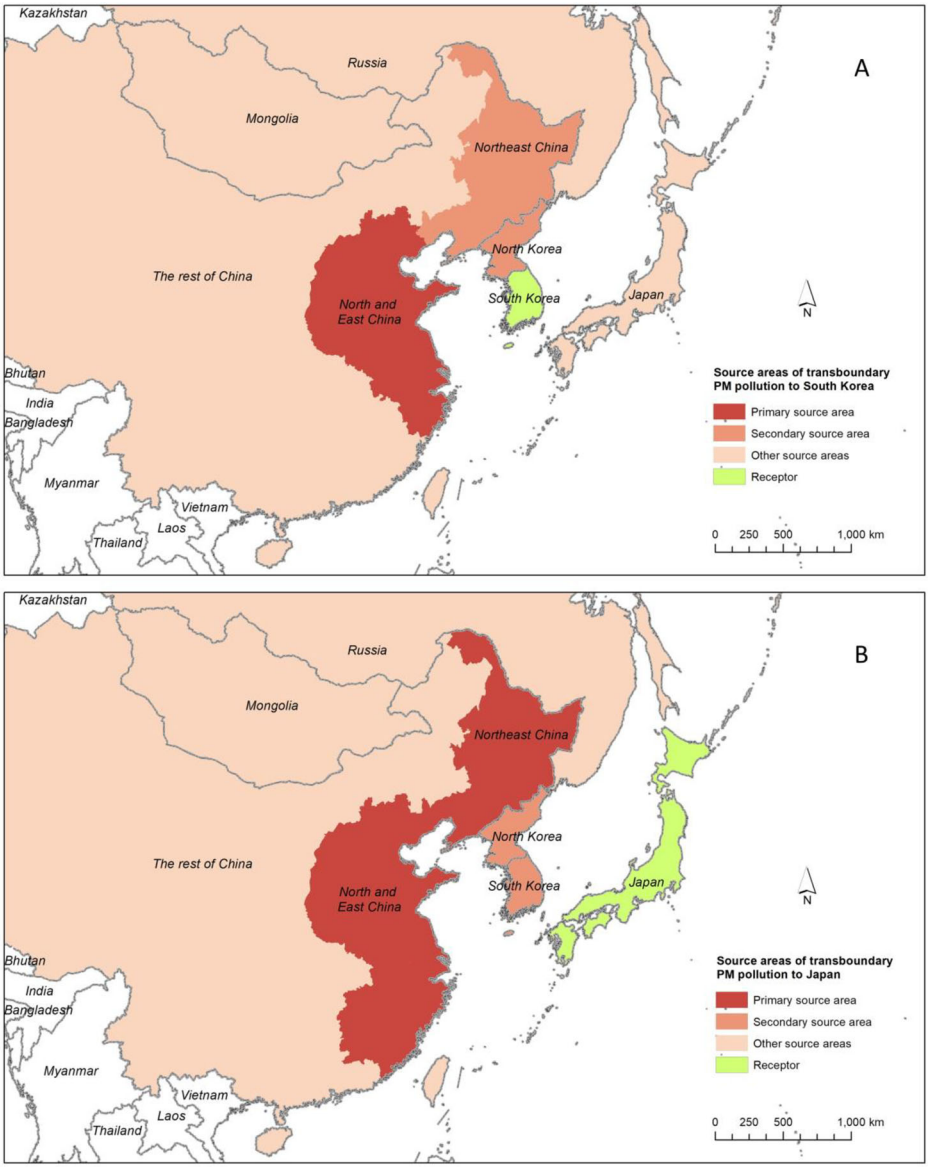

Figure 2. Source areas of transboundary PM pollution in Northeast Asia to South Korea (A) and Japan (B) as the receptor regions. The map is produced based on results of simulated source areas from extant SRR studies of transboundary PM pollution in NEA published before (including) 2020. The map does not show the source area for local PM pollution in the receptor country. Please note that the map is only for illustrative purpose and the boundaries of the simulated source areas in the map may be different from the boundaries of the actual source areas.

countries causes the air to descend and leads to the accumulation of PM pollution in these countries (S. Lee et al., 2011; Qu et al., 2016).

3. Contributions of source areas to PM pollution in Northeast Asia

3.1. Approaches for quantifying the contributions of source areas to PM pollution

The most frequently used methods to quantify the relative contributions of source areas to PM pollution in NEA can be generally classified into two groups: the sensitivity analysis techniques

Table 2. Studies of the source-receptor relationship of transboundary PM pollution to Japan as the receptor region.

Entry	Time period	Pollutants	Receptor	Method	Emission inventory	Source countries and their relative contributions
(Uno et al., 2017, 2020)[a]	Dec. 2013–Mar. 2015	$PM_{2.5}$	Fukuoka, Japan	Brute-force method using GEOS-Chem	China (MEIC 2010), other regions (REAS v2.1 2008)	China (58%), Japan (19.6%), Korea peninsula (16.3%), Mongolia and Russia (<1.9%)
(LTP, 2019)	2017	$PM_{2.5}$	Tokyo, Osaka, Fukuoka in Japan	WRF-CAMx, WRF-CMAQ	China (emission data in 2017), South Korea and Japan (emission data in 2015)	China (24.6%), South Korea (8.2%), Japan (55.4%)
(Yim et al., 2019)	2010	$PM_{2.5}$	Japan	Zero-out contribution method using CMAQ	HTAP_v2 2010	China (53.9%), Japan (29.3%), South Korea (3.3%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Kyushu, Japan	Brute-force method using CMAQ	REAS v2 2008	China (61%), Japan (21%), Korea peninsula (10%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Chugoku, Japan	Brute-force method using CMAQ	REAS v2 2008	China (59%), Japan (25%), Korea peninsula (11%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Shikoku, Japan	Brute-force method using CMAQ	REAS v2 2008	China (60%), Japan (23%), Korea peninsula (8%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Kinki, Japan	Brute-force method using CMAQ	REAS v2 2008	China (51%), Japan (36%), Korea peninsula (6%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Hokuriku, Japan	Brute-force method using CMAQ	REAS v2 2008	China (55%), Japan (33%), Korea peninsula (6%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Tokai-Koshin, Japan	Brute-force method using CMAQ	REAS v2 2008	China (45%), Japan (46%), Korea peninsula (3%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Tohoku, Japan	Brute-force method using CMAQ	REAS v2 2008	China (59%), Japan (30%), Korea peninsula (4%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Hokkaido, Japan	Brute-force method using CMAQ	REAS v2 2008	China (69%), Japan (19%), Korea peninsula (4%)
(Ikeda et al., 2015)	2010	$PM_{2.5}$	Kanto, Japan	Brute-force method using CMAQ	REAS v2 2008	China (39%), Japan (51%), Korea peninsula (0%)
(Ikeda et al., 2014)[b]	2010	$PM_{2.5}$	Fukue, Japan	Brute-force method using CMAQ	REAS v2 2008	China (78.5%), Korea peninsula (10.6%), Japan (7.8%)
(Ikeda et al., 2014)[b]	2010	$PM_{2.5}$	Oki, Japan	Brute-force method using CMAQ	REAS v2 2008	China (69%), Korea peninsula (13.5%), Japan (13.1%)
(Ikeda et al., 2014)[b]	2010	$PM_{2.5}$	Nonodake, Japan	Brute-force method using CMAQ	REAS v2 2008	China (46.1%), Japan (40.4%), Korea peninsula (5%)
(Ikeda et al., 2014)[b]	2010	$PM_{2.5}$	Rishiri, Japan	Brute-force method using CMAQ	REAS v2 2008	China (86.7%), Japan (5.3%), Korea peninsula (5%)
(Li et al., 2014)	2010	PM_{10}	Japan	Tracer-tagged method using NAQPMS	China (2010 data from Zhao et al. (2013)), Other regions (REAS v2.1 2008)	China (13.6%), Japan (39.1%), Korea peninsula (2.7%), Russia & Mongolia (<1.5%)
(Shimadera et al., 2009)	March 2005	PM	Kinki, Japan	Zero-out contribution method using CMAQ	Japan (EAGrid2000-Japan 2000), other regions (INTEX-B 2006, REAS 2005)	All regions outside Kinki (55%–82%)

[a]The percentage of relative contribution for Mongolia and Russia was estimated using WebPlotDigitizer (https://automeris.io/WebPlotDigitizer) based on Figure 5 in Uno et al. (2017).
[b]The percentages of relative contributions for China and Korea peninsula were estimated using WebPlotDigitizer (https://automeris.io/WebPlotDigitizer) based on Figure 7 in Ikeda et al. (2014).

and the tagged tracer method in Eulerian CTMs. Commonly used sensitivity analysis techniques can be further divided into the brute force method, decoupled direct method, and adjoint sensitivity analysis method based on computational efficiency and the way in which the sensitivity coefficients are calculated.

3.1.1. Brute force method

The brute force method is the simplest and most widely used method for assessing the rate of change in pollutant concentrations when small perturbations are made to the emissions (Kajino et al., 2013; Zhang et al., 2005). This method usually requires running the air quality model twice. The first model run (baseline case) is performed with standard input emission inventory data, whereas the second model run is carried out with the emission data in the source region of interest reduced by a certain percentage. Suppose the simulated pollution concentrations in the receptor region of interest in the first and second model runs are C_1 and C_2, respectively, and the percentage of adjustment in the emission data in the source region of interest is p. Then, the relative contribution of emissions in the source region of interest to air pollution in the receptor region is defined as the change ratio of the two simulated pollutant concentrations in the first and second model runs divided by the percentage of the emission adjustment (Eq. (2)).

$$\text{Relative contribution} = \frac{C_1 - C_2}{C_1 * p} * 100\% \tag{2}$$

The percentage of emission reduction varies across different studies. For transboundary PM pollution studies in NEA, some studies applied a 20% reduction rate (Ikeda et al., 2014, 2015), whereas others adopted 50% reduction (H. C. Kim et al., 2017). For an emission reduction percentage of 100%, that is, with the emission in the source region of interest set to zero, the customized brute force method is called the zero-out contribution method (Koo et al., 2008; Shimadera et al., 2009; Yim et al., 2019).

The strength of the brute force method in estimating the relative contribution of a source region to a receptor region is that it is easy to implement and routinely applied in regulatory applications (Cohan & Napelenok, 2011; Kelly et al., 2015). However, this method assumes that the model response to emission input changes is linear, which does not always hold true. The estimated relative contribution of a source region to a receptor region may vary with the changes in the emission reduction rate (Thunis et al., 2019). For example, a modeling study of the nonlinear response of $PM_{2.5}$ pollution to emissions in the Beijing–Tianjin–Hebei region of China showed that the calculated relative contribution generally increased with the emission reduction percentage (Zhao et al., 2017). Moreover, contributions from all source regions are not strictly additive, which means that the sum of all contributions to PM concentration does not always equal the baseline PM concentration (Cohan & Napelenok, 2011; Thunis et al., 2019). In addition, this method can become very expensive and inefficient when there is more than one emission reduction scenario to run.

3.1.2. Decoupled direct method

The decoupled direct method calculates sensitivity coefficients that represent the responses of pollutant concentrations to the perturbations in a few parameters (e.g., emissions in a source region) (Dunker, 1984). These sensitivity coefficients can then be used as coefficients of a Taylor polynomial to calculate the concentration change and further estimate the contribution of the source region (Cohan et al., 2005). As the decoupled direct method computes pollutant sensitivities directly within the model, this method provides a more efficient way of performing sensitivity analysis to model inputs than the brute force method (Itahashi et al., 2012; Napelenok et al., 2006).

The decoupled direct method has been implemented in a few Eulerian CTMs, including CMAQ (Napelenok et al., 2006) and CAMx (Dunker et al., 2002), and has been proved to produce consistent results in a computationally efficient way compared with the brute force method (Cohan & Napelenok, 2011). In NEA, the decoupled direct method implemented in CMAQ was used to quantify the contributions of emissions from countries in NEA on Seoul, South Korea (J.-H. Kim et al., 2016). Similar to the brute force approach, the decoupled direct method cannot consider the nonlinear effects in the emission–concentration relationship due to nonlinearities in the atmospheric chemistry processes (Clappier et al., 2017; Itahashi et al., 2012). A study showed that the decoupled direct model provided accurate estimations of sensitivities under small perturbations in emissions but inaccurate results when the perturbations were large (Zhang et al., 2005).

3.1.3. Adjoint sensitivity analysis method

The brute force approach can only calculate the model response to one perturbation in the emission at a time, whereas the decoupled direct method can compute the sensitivity coefficients of pollutant concentrations simultaneously to changes in a small number of parameters (e.g., emission or boundary condition) (Zhang et al., 2008). However, when it comes to changes in a large number of parameters, neither the brute force approach nor the decoupled direct method is feasible. The adjoint sensitivity analysis method, however, provides an efficient way for calculating the sensitivity of model outputs (e.g., tracer concentration) to numerous model parameters (Cohan & Napelenok, 2011). In an Eulerian grid model, the adjoint sensitivity analysis method calculates the sensitivity (also called gradient) of a user-defined cost function with respect to various model inputs (e.g., emissions) at each grid/location within the model domain (Wang et al., 2019). A cost function usually refers to a predefined concentration-related function that measures the model response. Then, the calculated sensitivity coefficients are used to compute the semi-normalized sensitivity coefficients, which directly quantify how emissions from sources in different locations influence the overall concentrations of pollutants of interest in a receptor region (Wang et al., 2019). Finally, the relative contributions are estimated using the semi-normalized sensitivity coefficients. Details of the adjoint model description and validation are described in Henze et al. (2007).

The adjoint sensitivity analysis method has been implemented in several modern regional and global CTMs, including CMAQ (Hakami et al., 2007) and GEOS-Chem (Henze et al., 2007). Among the studies of SRR analysis of transboundary PM pollution in NEA, we found two studies that adopted the adjoint sensitivity analysis method (Choi et al., 2019; Lee et al., 2017).

3.1.4. Tagged tracer method

The tagged tracer method, as its name suggests, keeps track of the origin of air pollutants in a model simulation by tagging and labeling the geographical locations where the pollutants are emitted or formed (Thunis et al., 2019). In the case of particulate matter, the tagged tracer method tracks primary and secondary PM components by assuming that each secondary component links directly to one specific precursor (Cohan & Napelenok, 2011). For example, NO_X emissions lead directly to the formation of particulate nitrate, SO_X emissions lead to the formation of particulate sulfate, and NH_3 emissions lead to the formation of particulate ammonium. By tracking the origin of air pollutants, the tagged tracer method provides spatially and chemically resolved source attribution information.

The tagged tracer method is currently implemented in the tagged species source apportionment algorithm (TSSA) in CMAQ (Wang et al., 2009), the particle source apportionment technology (PSAT) within the CAMx model (Wagstrom et al., 2008), and NAQPMS (Li et al., 2014). As all the source apportionment information can be obtained by executing the model once, the

tagged tracer method is computationally efficient and has been widely used in transboundary PM pollution studies in NEA (B. Kim et al., 2017; Li et al., 2014; Oh et al., 2020). The limitation of this approach is that it only considers direct chemical effects (e.g., NH_3 emissions lead to the formation of ammonium) but not the indirect effects (e.g., NH_3 emissions can affect the formation of nitrate), thereby limiting its ability to fully quantify the source contribution (Burr & Zhang, 2011; Thunis et al., 2019).

As discussed above, each of the four methods has its own strengths and weaknesses. Studies have shown that the tagged tracer method provides similar results to the brute force approach, although it cannot account for indirect chemical effects (Koo et al., 2009). Regarding the first three sensitivity analysis techniques, the ability of the three methods in obtaining accurate estimations of the relative contribution of a source region to air pollution in a receptor region is limited in the case of nonlinear relationships between pollutant concentrations and emissions (Clappier et al., 2017). However, despite this limitation, some applications do not require complete quantification of the nonlinearity. For example, Thunis et al. (2015) showed that when analysis is conducted on a yearly basis, it is reasonable to assume that the relationship between emissions and concentration is linear, suggesting that these sensitivity analysis techniques can yield reliable and accurate results.

3.2. Contributions of source areas to PM pollution in Northeast Asia

Tables 1 and 2 show the relative contributions of source countries in NEA to PM concentrations in South Korea and Japan, respectively. The relative contributions of source countries in NEA to PM concentrations in China are not shown as studies have indicated that the contributions of South Korea and Japan to PM pollution in China are insignificant (Li et al., 2014). As shown in Tables 1 and 2, varying estimates of the relative contributions of China to PM pollution in South Korea and Japan have been obtained. These studies show that the estimated relative contributions of China to PM pollution in South Korea range from 26% to 70% (Table 1) and from 13.6% to 86.7% to PM pollution in various locations in Japan (Table 2). If the analysis periods are limited to one full year or several years, and the receptor regions are limited to an entire country, the estimated relative contributions of China to PM pollution in South Korea range from 26% to 56% and from 13.6% to 53.9% to PM pollution in Japan.

Figure 3 shows the relative contributions of China to PM pollution in South Korea across different analysis time periods in various studies. As can be seen in Figure 3, higher relative contributions of China to PM pollution in South Korea (over 60%) were usually from studies that focused on shorter time periods (Choi et al., 2019; B. Kim et al., 2017; Lee et al., 2017; Oh et al., 2020). In addition, these studies were also characterized by outdated emission inventory data in 2010 or before being used as well as most authors being originally from the same country with few or no international collaboration, whereas studies that focused on longer analysis time period (C. Bae et al., 2020; M. Bae et al., 2020; Li et al., 2014; LTP, 2019; Yim et al., 2019), used recently updated emission inventory (LTP, 2019), and involved substantial international collaboration (LTP, 2019; NASA & NIER, 2017), usually produced lower relative contributions.

Results from the literature show that it is still debatable whether China contributes more air pollution to the receptor countries than the receptor countries themselves. For example, a report written jointly by a group of scientists from Korea's National Institute of Environmental Research (NIER) and the United States National Aeronautics and Space Administration (NASA) concluded that China contributed less than South Korea to $PM_{2.5}$ pollution in Seoul, South Korea (NASA & NIER, 2017); however, Oh et al. (2020) concluded that China contributed more than South Korea to $PM_{2.5}$ pollution in Seoul, South Korea.

The large disagreement in the estimates of the relative contributions of one source country to a receptor country could be explained by the following five possible reasons. First, these estimates

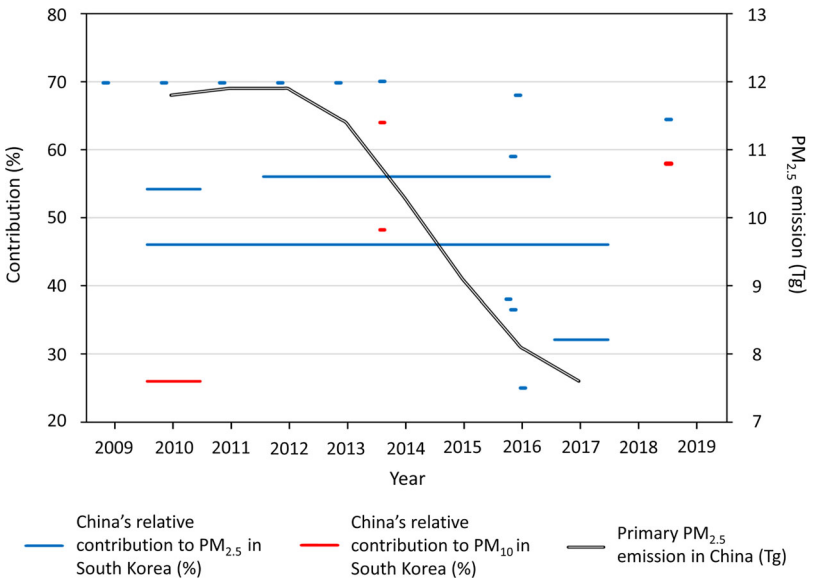

Figure 3. The relative contributions of emission in China to PM pollution in South Korea across different analysis time periods in various studies and the $PM_{2.5}$ emission trend in China from 2010 to 2017. Short dashes indicate the relative contributions for short time periods, whereas longer dashes indicate the relative contributions for a longer analysis time period of one full year or several years. Data on $PM_{2.5}$ emission trend in China are from Zheng et al. (2018).

were based on different time periods, which have different input meteorological data and emission data. For example, Choi et al. (2019) found that the percentage of Chinese contribution under the "blocking" meteorological conditions was 26%, whereas the percentage under the "extreme pollution" meteorological conditions was as high as 68%.

Second, these estimates were calculated using different analytical approaches. Some studies adopted the brute-force approach (C. Bae et al., 2020; H. C. Kim et al., 2017), whereas others used the tagged tracer method which, can only account for direct chemical effect (B. Kim et al., 2017; Li et al., 2014; Oh et al., 2020). Even for studies that used the same brute-force approach, the estimates varied due to the inherent nonlinear relationship between pollutant concentration and emission changes in the analysis of SRR.

Third, the different Eulerian CTMs used in transboundary PM pollution studies might have contributed to disagreements in the estimated relative contributions because different models have different representations of atmospheric chemistry processes and different model parameterization settings. Even the model grid resolution plays a role in producing varying estimated relative contributions. One study shows that the difference in relative contributions between a 9-km model simulation and a 27-km model simulation can reach 10% (M. Bae et al., 2020).

Fourth, different studies have different definitions of receptor and source regions. Some studies only included eastern China and part of northeastern China as the source regions of China (Lee et al., 2017), whereas other studies included all of China (Yim et al., 2019). Even for the same definition of receptor regions, different sampling methods might result in different estimates of relative contributions. For example, M. Bae et al. (2020) revealed that the relative contributions of a source region to PM pollution in the monitor location-based receptor regions and area-based receptor regions are significantly different in sparsely populated cities in South Korea.

The fifth possible reason is that the epistemic communities on transboundary PM pollution in NEA are fragmented; that is, scientists in China, South Korea, and Japan tend to work in separate groups, which makes it very difficult for each community to reach a consistent understanding of transboundary PM pollution in NEA (Kim, 2007; Lee & Paik, 2020; Yarime & Li, 2018). The epistemic community here refers to a network of experts with authority on policy-relevant

knowledge in a particular field (Haas, 1992). Each epistemic community has different opinions regarding the estimates of relative contributions in the beginning (Yarime & Li, 2018). Because of the fragmentation of epistemic communities, subsequent studies of the SRR of transboundary PM pollution in NEA were possibly influenced by the established perspectives formed in the respective epistemic communities of each country, which likely contributed to the inconsistency in the findings of the source contributions.

Although many studies have been conducted to investigate the SRR of transboundary PM pollution in NEA, when studies are segmented by whether the model simulated over a full year of PM pollution and by the definitions of source regions and receptor regions, the number of studies remains small, which is insufficient for the computation of statistics, such as the averaged relative contribution of a source country to a receptor country. Therefore, our analysis provides a basis on which future studies can build. As more studies focusing on transboundary PM pollution in NEA are published, it is important to update and refine our results.

4. Challenges of source-receptor relationship studies of transboundary PM pollution in Northeast Asia

4.1. Data quality issues

A typical SRR study usually requires emission inventories for driving the model and ground-level PM measurement datasets for model performance evaluation. The quality of these datasets is essential for accurate modeling and deriving convincing SRR analysis results.

The commonly used emission inventories in transboundary PM pollution studies in NEA include the Multi-resolution Emission Inventory for China (MEIC) (http://www.meicmodel.org) based on the Zhang et al. (2009) study on the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B), the Regional Emission Inventory in Asia (REAS) (Ohara et al., 2007), the Clean Air Policy Support System (CAPSS) for South Korea (D.-G. Lee et al., 2011), the EDGAR-HTAP dataset (Janssens-Maenhout et al., 2015), the MIX inventory for supporting the Model Inter-Comparison Study for Asia (MICS-Asia) (M. Li et al., 2017b), KU-CREATE (Woo et al., 2012), KORUS inventory (Choi et al., 2019), and others. The common problem of uncertainties in these emission inventories has been well documented in the literature (Crippa et al., 2019; Ding et al., 2017; Han et al., 2009; M. Li et al., 2017a; Zhu et al., 2019). In particular, it is worth noting that most of the SRR studies in NEA that modeled the time periods after 2010 used the MEIC 2010, MICS-Asia 2010, or INTEX-B 2006 as the anthropogenic emission data in China (see Tables 1 and 2). It is understandable that outdated emission data were chosen because the latest emission data in the source regions for the time period modeled were probably unavailable. However, the issue of data availability must not distract attention from the problem that the outdated emission data could render estimations of the SRR of transboundary PM pollution in NEA inaccurate. A study by the developer of MEIC has shown that anthropogenic emissions in China have been substantially reduced, owing to China's active and stringent clean air policies during 2010–2017 (Zheng et al., 2018). The results of the study showed that the emission reduction rates significantly increased after 2013, when China's State Council announced its Action Plan for Air Pollution Prevention and Control (see Figure 3). Meanwhile, China's anthropogenic emissions for SO_2, NO_X, CO, PM_{10}, $PM_{2.5}$, BC, and OC were reduced by 59%, 21%, 23%, 36%, 33%, 28%, and 32%, respectively, from 2013 to 2017 (Zheng et al., 2018). None of these important changes in emissions has been considered in current SRR studies in NEA, which might lead to inaccurate estimations of China's contribution to PM pollution in South Korea and Japan.

Ground-level PM measurements are usually used to compare the simulated PM concentration to assess the performance of CTMs used in SRR studies (H. C. Kim et al., 2017; Yim et al., 2019). If the ground-level PM measurements in the modeling domain have significant data quality

issues, the model evaluations are likely problematic, which lead to further concerns regarding the SRR analysis results using these models. Therefore, we conducted a quality check of the PM measurements in China, South Korea, and Japan on two potential data quality issues identified by Liu et al. (2016): $PM_{2.5}$ concentrations exceeding concurrently measured PM_{10} concentrations at the same station and repeated readings of PM concentration over consecutive hours. According to Liu et al. (2016), the two quality issues of PM measurements in China in 2014 were significant. A total of 367 and 46 stations, constituting 34.8% and 4.4% of all air quality monitoring stations in China, recorded higher readings of $PM_{2.5}$ over PM_{10} in over 1% and 5% of the records, respectively. 322 and 34 stations, constituting 30.5% and 3.2% of all stations in China, recorded over 1% and 5% of repeated $PM_{2.5}$ measurements, respectively. For South Korea and Japan, we primarily used PM measurement data from January 1, 2017 to December 31, 2018, collected from the Japan Atmospheric Environmental Regional Observation System (http://soramame.taiki.go.jp/) and the South Korea Ministry of Environment (http://www.airkorea.or.kr/). The results show that PM measurements in Japan have a much greater problem of $PM_{2.5}$ concentrations exceeding concurrently measured PM_{10} concentrations at the same station than does China. In fact, 953, 875, and 757 stations, constituting 94.2%, 86.5%, and 74.8% of all 1012 stations that can both measure $PM_{2.5}$ and PM_{10} in Japan, have $PM_{2.5}$ values greater than PM_{10} values in over 1%, 5%, and 10% of the records, respectively. Some stations even have problematic readings of over 60% (Figure S1). For the problem of repeated readings of PM concentration over consecutive hours, 218 and 37 stations, constituting 20.3% and 3.4% of all 1074 stations that measure $PM_{2.5}$ in Japan, recorded over 1% and 5% of repeated $PM_{2.5}$ measurements, respectively. Some stations even recorded over 15% of repeated $PM_{2.5}$ measurements (Figure S2). The quality problems of PM measurements in South Korea were not as significant as Japan. 114 and 13 stations, constituting 23.5% and 2.7% of all 485 stations in South Korea, exhibit higher $PM_{2.5}$ in over 1% and 5% of the records, respectively. A total of 115 and 6 stations, constituting 23.7% and 1.2% of all 485 stations, recorded over 1% and 5% of repeated $PM_{2.5}$ measurements, respectively. Therefore, it is recommended that future SRR studies in NEA that use ground-level PM measurements for model evaluation should address these data quality issues first.

4.2. Limited simulation scale in time and space

Many SRR studies of the transboundary PM pollution in NEA focused on episodic high-pollution events (Itahashi et al., 2017; Lee et al., 2017; H. J. Lee et al., 2019a; Moreno et al., 2012; Oh et al., 2020). Although these studies have contributed important and detailed insights into the transboundary PM pollution between China, South Korea, and Japan, the scope of these studies is limited to short time periods and does not provide complete information on the SRR of transboundary PM pollution in NEA over a full year. Usually, these studies focused on short time periods when transboundary PM pollution was significant. Therefore, the estimated relative contributions of a certain source area to a receptor region in these studies were found to be usually higher than the calculated relative contributions in studies that were focused on the span of a full year. For example, in a study of the transboundary $PM_{2.5}$ pollution during February 23–27, 2014, China was found to contribute 70.4% of $PM_{2.5}$ pollution in Seoul (B. Kim et al., 2017), whereas in another study focused on 2012–2016, China was found to contribute 42% of $PM_{2.5}$ pollution in Seoul (C. Bae et al., 2020). However, SRR analyses for short time periods in these studies led to unintended consequences. The general public and media typically do not carefully differentiate between the much higher percentages of relative contributions over a certain short time period and the lower percentages of relative contributions over a full year. Unfortunately, the media usually chooses the higher percentage of relative contribution to highlight the significance of transboundary PM pollution (D.-S. Kim, 2017), leading to the dissemination of misleading

information that has probably contributed to the recurring finger-pointing and discontent among stakeholders in China, South Korea, and Japan in recent years.

In addition to limitations on the temporal scale, most SRR studies are also limited in the spatial extent of the simulations (Choi et al., 2019; Lee et al., 2017; NASA & NIER, 2017). Most air pollutants have relatively long residence times in the atmosphere (Chen & Taylor, 2018), which enable these pollutants to travel thousands of kilometers away from their source areas. These air quality modeling studies that focused on a small area might have falsely attributed some of the transboundary PM pollution to the wrong source area. Studies have shown that biomass-burning emissions form the Indo-China peninsula (mainly comprising Myanmar, Thailand, Laos, Cambodia, and Vietnam) are one of the principal sources of PM pollution and other trace gases, such as O_3, over East Asia (Yadav et al., 2017). Moreover, dust from the arid region, extending from Central Asia to Mongolia and northern China, has been transporting vast quantities of air pollutants to East Asia by strong westerlies (Zhang et al., 2020). Therefore, it is recommended that SRR studies of transboundary PM pollution in NEA should include major source areas of air pollutants that were not considered previously.

In addition, as mentioned in the previous section, the different definitions of receptor regions and source regions could contribute to the inconsistent estimates of the relative contributions of one source country to a receptor country, making it very difficult to synthesize the findings of modeling studies. Therefore, it is recommended that future modeling studies on the SRR of transboundary PM pollution in NEA could follow a same modeling setting, such as applying consistent definitions of receptor and source regions as well as using the same simulation period as MICS-Asia did (Carmichael et al., 2002). In doing so, it would be easier to obtain a common understanding of model performance and uncertainties in NEA and further improve model's reproducibility, thereby helping to understand the causes of the uncertainties.

4.3. Limited analytical perspective in allocation of environmental responsibility

The SRR studies of transboundary PM pollution aim to quantify the amount of transboundary PM pollution transported from the source countries to the receptor countries. In this regard, the SRR studies of transboundary PM pollution in NEA are actually part of the important debate underway to devise appropriate methods to allocate the environmental responsibility for the damage caused by transboundary PM pollution. It was not surprising that research findings of many source-receptor analyses of transboundary PM pollution in NEA were cited by the media, government agencies, and the public when discussing the allocation of environmental responsibility (D.-S. Kim, 2017). The allocation of environmental responsibility is closely related to national interest. This may be why the transboundary PM pollution in NEA has increasingly become a politically contested and sensitive issue rather than an environmental challenge.

Current transboundary PM pollution studies approach the issue of transboundary PM pollution in NEA only from the perspective of atmospheric transport, which is limited and does not consider the process of production, consumption, and trade. For example, considering the emission of air pollutants in the industrial sector, at least three parties play important roles in the emission process: investors who initiate and fund pollution emissions, producers who produce pollution, and consumers who stimulate pollution emissions (Meng et al., 2016). All three parties benefit from the value chain and should bear certain responsibilities. The literature on greenhouse gas emissions also suggests an income-based perspective to assign environmental responsibility to earners of income from production and consumption activities (Liang et al., 2017). In other words, the relevant question is not "how much pollution has been transported from the source country to the receptor country?" but rather "how much have the earners of income in the source country and the receptor country earned from the pollution that has been transported from the source country to the receptor country?" Future SRR studies in NEA could attempt to combine

the perspective of atmospheric transport with analytical perspectives of investment, production, and consumption in attributing pollution and environmental responsibilities (Lin et al., 2014; Zhang et al., 2017).

5. Suggestions for future research

Table S4 summarizes three suggestions for future SRR studies of transboundary PM pollution in NEA to address the challenges discussed in the previous section. First, the data quality of the PM measurement data and emission inventory data in China, South Korea, and Japan should be screened and evaluated. We recommend that future studies first identify the data quality issues, estimate the extent of the data quality issues, and then remove data sources (e.g., air quality monitoring stations) that have significant proportions of problematic data points. Future studies should also use updated emission inventory data. Second, the challenge of limited simulation scale in time and space can be addressed by analyzing the SRR for a full year, including major source areas of air pollutants that were not considered before, and using the same modeling inputs, settings, and parameters for model comparison. Third, future studies should look beyond the perspective of atmospheric transport in discussing the allocation of environmental responsibilities and attempt to combine the perspective of atmospheric transport with other perspectives of industrial production and trade that take into account investment, production, and consumption.

6. Conclusion

This review provides a summary of the existing knowledge on the SRR of transboundary PM pollution in NEA. It finds that although China is the primary source area of transboundary PM pollution for both South Korea and Japan; and eastern-, northern-, and northeastern China are the most contributing source areas within China for transboundary PM pollution to both South Korea and Japan, it remains debatable whether China contributed more PM pollution to the receptor countries (South Korea and Japan) than the receptor countries themselves. Furthermore, there are substantial differences in the estimations of the relative contributions of China to PM pollution in South Korea or Japan. These differences are largely due to the discrepancies in the analysis periods, analytical approaches, modeling settings, definitions of source and receptors, and international collaboration. This review also finds that higher relative contributions are usually obtained from studies that focused on short time periods, used outdated emission inventory data, and had few or no international collaboration. If the analysis periods are limited to one full year or several years and the receptor regions are limited to an entire country, the relative contributions of China range from 26% to 56% to PM pollution in South Korea and from 13.6% to 53.9% to Japan. This review demonstrates that current SRR studies in NEA face challenges from data quality issues in PM measurement data and emission inventory data, limited simulation scale in time and space, and limited analytical perspectives in the allocation of environmental responsibilities. It is proposed that future studies screen the data quality, address modeling scales, and see the larger context through perspectives of industrial production and trade.

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