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A review of the technology and applications of methods for evaluating the transport of air pollutants

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ABSTRACT

A variety of methods based on air quality models, including tracer methods, the brute-force method (BFM), decoupled direct method (DDM), high-order decoupled direct method (HDDM), response surface models (RSMs) and so on forth, have been widely used to study the transport of air pollutants. These methods have good applicability for the transport of air pollutants with simple formation mechanisms. However, differences in research conclusions on secondary pollutants with obvious nonlinear characteristics have been reported. For example, the tracer method is suitable for the study of simplified scenarios, while HDDM and RSMs are more suitable for the study for nonlinear pollutants. Multiple observation techniques, including conventional air pollutant observation, lidar observation, air sounding balloons, vehicle-mounted and ship-borne technology, aerial surveys, and remote sensing observations, have been utilized to investigate air pollutant transport characteristics with time resolution as high as 1 sec. In addition, based on a multi-regional input-output model combined with emission inventories, the transfer of air pollutant emissions can be evaluated and applied to study the air pollutant transport characteristics. Observational technologies have advantages in temporal resolution and accuracy, while modeling technologies are more flexible in spatial resolution and research plan setting. In order to accurately quantify the transport characteristics of pollutants, it is necessary to develop a research method for interactive verification of observation and simulation. Quantitative evaluation of the transport of air pollutants from different angles can provide a scientific basis for regional joint prevention and control.

Introduction

Over the past four decades, sustained and rapid economic development and urbanization, including a sharp increase in the vehicle population, have caused severe environmental prob-

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lems (Song et al., 2017; Bai et al., 2021; Wang et al., 2017), among which air pollution issues are particularly prominent. To effectively improve the atmospheric environment, governments at all levels have released policies and measures since 2013, including the "The Action Plan for Preventing and Controlling Air Pollution", "the Clean Air Action Plan", "the Three-year Action Plan for Winning the Battle for Blue Skies" etc. As such, implementing such measures has significantly improved air quality (Wang et al., 2018; Ding et al., 2019). The United Nations Environment Programme released two reports in 2016 and 2019, highlighting improvements in Beijing's air quality in response to effective control strategies related to clean fuel, construction dust, traffic dust, and coal-fired boilers (Wang et al., 2021d; Lang et al., 2016; Zhao et al., 2020). This achievement has also provided a suitable example for other developing countries. However, persistent heavy pollution processes still occur in many areas, especially during the autumn and winter seasons (Zhong et al., 2018; Yang et al., 2015, 2021a; Tian et al., 2021). Many studies have been conducted on the causes and mechanisms of air pollution. With advances in research, it has been found that dealing with the regional characteristics of air pollution has become a key issue in improving urban air quality (Xiao et al., 2021; Guo et al., 2014; Wang et al., 2021b). Thus, establishing a technical method that can accurately quantify the transport of air pollutants is significant for regional joint prevention and control mechanisms.

A variety of observational data sets have been used to study the qualitative or quantitative transport of pollutants. Wind field information at different heights can be used to analyze air pollutant transport during specific periods, and multiple ground and vertical observation sites can be applied to investigate the transport of air pollutants at urban and regional scales (Wang et al., 2021a). In addition, mathematical models based on observational data have been established (Duan et al., 2021; Ge et al., 2018). In general, observational data can objectively characterize the temporal and spatial distributions of air pollutants. However, due to the low density of urban observation sites, it is difficult to conduct large-scale observation experiments, along with a general lack of long-term air pollutant transport research across regions.

Air quality models can compensate for this defect. They have been widely used in the study of atmospheric pollution. The Hybrid Single-Particle Lagrangian Integrated Trajectory model can be used to qualitatively analyze the source of polluted air masses in different periods (Wang et al., 2015; Li et al., 2017; Jongbae et al., 2015; Sun et al., 2018). It can deal with the input of various meteorological elements and physical processes, and thus simulate relatively complete transport, diffusion and deposition processes. The Potential Source Contribution Function (PSCF) and Concentration Weighted Trajectory (CWT) method can also identify the transport trajectories and high source regions. These have been used to study the transport pathways of typical cities (Zhang et al., 2018b, 2019a). Small-scale models have been widely applied in air pollutant diffusion and source identification at the local level (Mao et al., 2020a, 2020b). These models usually contain no chemical mechanism modules (e.g., Gaussian plume model) or only simple chemical reaction mechanisms (e.g., Calpuff model). The physical dispersion process of atmospheric pollutants is generally described by the dispersion coefficients. For instance, the Calpuff model (Joseph et al., 2000) can simulate the chemical reaction for the conversion SO_2 to SO_4^{2-} and NO_x to NO_3^- based on the MESOPUFF II and RIVAD/ARM3 schemes, respectively. The three-dimensional air quality models, including the Community Multiscale Air Quality model (CMAQ), Comprehensive Air Quality model (CAMx), Weather Research & Forecasting, and Chemical Transport model (WRF-Chem), which comprehensively consider the physical diffusion and chemical transformation of pollutants, can reproduce the spatial and temporal distribution of pollutants in historical periods. Some studies have clarified the impact of a single type of emission source diffusion based on multi-scenario simulation program settings (Dai et al., 2021; Eunhye et al., 2021; Lang et al., 2021; Li et al., 2021). Combining different modules, the transport of air pollutants at urban and regional scales has also been discussed (Jingkyul et al., 2019; Han et al., 2021). The contribution of local and non-local emission sources can be identified based on receptor and source tags, yielding an intercity air pollutant matrix. In addition, the pollutant transport flux at different scales can be calculated by combining interface wind field and pollutant concentration information. Based on this, some scholars have shown that the calculation of transport flux combined with variations in the boundary layer height can more effectively characterize and quantify the transport of pollutants. Moreover, the material exchange between the boundary layer and the free atmosphere has also been shown to have an impact on pollution formation (Zhang et al., 2018a, 2021b; Qu et al., 2021a; Chang et al., 2018). Therefore, research on the temporal and spatial distribution of the boundary layer height and boundary-free atmospheric matter exchange is necessary to further understand air pollution transport.

In addition to pollutant transport research based on environmental impact, the implicit transfer of pollution emissions based on regional trade is also an effective technique for identifying the transport of air pollutants (Wang et al., 2020a; Li et al., 2019). Due to differences in the energy production and consumption levels between regions, energy production overflow or lack of energy exists in some regions. Inevitably, pollutant emissions are implicitly transferred based on the trade between regions. The multi-regional input-output (MRIO) model is widely used to recognize the transport of air pollutants (including $PM_{2.5}$, SO_2, NO_x, volatile organic compounds (VOCs), and CO_2) among various regions (Zhao et al., 2015; Zhu et al., 2018). This can clarify the trans-boundary migration mechanism of air pollutants from an economics perspective and is essential for the coordinated development of the environment and economy between regions.

This study explores the advantages and differences of different technical methods in air pollutant transport evaluation technologies from multiple angles based on a review of previous studies, summarizing the research results of the past ten years, combined with the research results of the present research group. This article provides data and scientific support for the next stage of regional joint prevention and control in China.

1. Transport matrix based on modeling approach

Three-dimensional air quality models have been widely applied in the study of air pollutant transport for inter-city and inter-regional scales because of their high temporal and spatial resolution and the flexibility of their research scheme design (Skyllakou et al., 2014; Eliana et al., 2013; Yang et al., 2021b; Zheng et al., 2015). These models include various advection and diffusion mechanisms, and thus can better characterize the transport and diffusion of pollutants in different physical processes. In terms of chemical mechanisms, complex aerosol mechanisms and gas-phase mechanisms can simulate each atmospheric process that affects the transport, transformation and removal of ozone, particulate matter and other pollutants. Although there are shortcomings in the mechanism of generation of secondary inorganic aerosol and secondary organic aerosol, the continuous improvement of parameterization schemes can reproduce the actual air quality situation well.

The contributions of various sources to air pollutants such as $PM_{2.5}$ and O_3 in target regions can be obtained directly utilizing tracer methods including CMAQ-ISAM and CAMx-PSAT/OSAT. A series of achievements have been made regarding the transport of pollutants at regional and urban scales (Zhang et al., 2019b; Dong et al., 2020). The annual average $PM_{2.5}$ in most of the previous studies was dominated by local emissions, with contributions larger than 50%. The primary components mainly came from local emissions, while the secondary components were closely related to mid- or even long-range transport (Ying et al., 2014; Hu et al., 2015; Ming et al., 2017; Qu et al., 2021b). O_3 is affected more by regional transport than $PM_{2.5}$ based on OSAT analysis. The tracer methods are mainly designed to understand the seasonal and inter-annual variations under specific meteorological and emission scenarios (Chen et al., 2021; Dong et al., 2020; Ge et al., 2021). These methods are suitable for transport research under various pollutant simplification scenarios, rather than study of the nonlinear characteristics of pollutant transport.

The brute-force method (BFM) is meant to simulate and compare baseline and reduction scenarios. The difference between two simulation projects has been used to represent source impacts (Fang et al., 2021; Lang et al., 2013). Some studies have focused on the sources of $PM_{2.5}$ in Beijing during heavily polluted periods. Results over the past years showed that local emissions dominated clean periods, while nonlocal emissions contributed more during polluted periods, with contribution more than 70% (Gao et al., 2016; Zhang et al., 2021a; Hu et al., 2021). In general, similar conclusions focusing on $PM_{2.5}$ transport could be obtained applying both BFM and tracer methods. However, marked differences have been reported in studies aimed at secondary pollutants. Qu et al. (2021b) developed a simulation based on BFM to identify the mechanism of indirect NO_3^- transport in the PRD and demonstrated the notable effects and complex process of cross-regional NO_3^- transport. It has also been reported that BFM is reliable for estimating single-source impacts on secondary sulfate in $PM_{2.5}$ (James et al., 2015). As for ozone, due to the sensitivity of O_3 to NO_x and VOCs, sharp reduction in NO_x emission might lead to the aggravation of O_3 pollution, which is different from the result of OSAT.

The decoupled direct method (DDM), high-order decoupled direct method (HDDM) and response surface models (RSMs) based on air quality models have also been applied in estimating the contribution of sources and regions to $PM_{2.5}$ and O_3. DDM and HDDM could calculate the sensitivity of precursors to changes in emissions. A previous study reported that DDM coupled with receptor models could give acceptable results for spatially resolved source apportionment (Ivey et al., 2015). Case studies also have been conducted to recognize the impact of sources on $PM_{2.5}$ and its components, as well as O_3 (Liao et al., 2007; Bates et al., 2018; James et al., 2015). RSMs obtained the response relationship between emissions and pollutant concentrations through multiple scenario simulations and mathematical statistical fitting. This approach has been widely utilized in linear and nonlinear analysis for $PM_{2.5}$ and O_3 (Xing et al., 2011, 2017, 2018).

Since there are obvious differences among the various technical methods, suitable technologies should be screened according to the research objectives and plans. Fang et al. (2021) evaluated the response relationship between sources and O_3 concentrations in PRD by using different techniques. The results show that BFM, RSM and HDDM can suitably evaluate the response between a single source and O_3 concentrations, but differences were obtained under different reduction scenarios. OSAT is more suitable for multi-source response relationship analysis. Comparative evaluation of technical methods was also conducted focusing on $PM_{2.5}$ and its major components. The SO_4^{2-} and NO_3^- should compete to combine with NH_4^+. The formation of the three components showed nonlinear responses to their precursors SO_2, NO_x and NH_3. Disagreement among methods is evident in calculations on NO_3^- (James et al., 2015). In general, the response relationship between target pollutants and their precursor emissions should be considered when screening applicable technologies. Various technologies have good applicability for the transport of pollutants with simple chemical mechanisms (such as primary particles), while for complex secondary pollutants, such as O_3, nitrate, etc., technical methods should be carefully selected.

2. Transport flux and pathway analysis

The transport flux method was applied to quantitatively evaluate the transport characteristics of different air pollutants. It is calculated by utilizing the mass concentration of $PM_{2.5}$ and wind field information and represents the mass of $PM_{2.5}$ passing through a specific vertical section. The formula for $PM_{2.5}$ flux (F) calculation is derived from a previous study of our research group as follows (Zhang et al., 2018a):

$$F = \sum_{k=1}^{h} \sum_{l} L \cdot H_k \cdot c \cdot v \cdot n \tag{1}$$

where, h (m) refers to the top height, l (m) refers to the boundary line between two adjacent cities, L (m) refers to the grid width of the modeling set, H_k (m) refers to the vertical height difference between layers k and k+1, c (μg/m^3) refers to the

$PM_{2.5}$ concentration in the vertical grid cell, v refers to the wind vector, and n is the normal vector corresponding to the vertical grid cell.

Various observation datasets have been used in previous studies to calculate the transport flux of atmospheric pollutants. The observational results of gaseous pollutants (including SO_2, NO, NO_2, O_3, and CO) and meteorological factors at Yufa Station from August 2006 to October 2008 were used to evaluate the transport between Beijing and the North China Plain (NCP). The results showed seasonal differences in transport flux observed among the above air pollutants passing through the NCP-Yufa-Beijing channel. In addition, the decreased surface flux intensity during the Olympics demonstrated the positive effect of joint prevention and control on air quality improvement (Li et al., 2016). Lv et al. (2017) applied a vehicle-based mobile lidar technique to estimate the transport flux based on the $PM_{2.5}$ and vertical wind-data profiles, finding that the southwest was an important regional transport pathway in Beijing. Dickerson et al. (2007) observed the regional distribution in the downwind transport process of the SO_2 pollution belt driven by a cold front in spring in northern China using ozone monitoring instrument (OMI) data, and then reconstructed the spatial distribution of pollutants based on the OMI aerosol index, Moderate-resolution Imaging Spectroradiometer aerosol optical depth, and other satellite observation data, as well as trajectory calculations and other means. The spatial distribution characteristics of the pollution belt during regional transport were confirmed.

Air quality models are also the leading technology used in transport flux studies. A study focusing on the transport characteristics of the four seasons in 2016 showed noticeable seasonal differences in the net flow flux among cities, closely related to altitude and location. In addition, the following three main transport pathways during the four seasons were obtained based on the transport flux results: northwest-southeast (January and April), southeast-northwest (July and October), and southwest-northeast (for the four seasons) (Zhang et al., 2019a). The differences between the BTH and Yangtze River Delta (YRD) regions and inland and coastal cities were also discussed. It was found that the seasonal inflow for the BTH region ranked in the order of April > October >July > January and January > October > April > July in the YRD region, indicating that it was more prominent in inland (Beijing and Shijiazhuang) cities than in a coastal city (Tianjin) in the BTH region (Guan et al., 2021). The vertical distribution of $PM_{2.5}$ transport flux has also been investigated. It was found that the vertical distribution of flux was similar during the pollution processes in autumn and winter in Beijing, with transport occurring at distinctly higher altitude in heavy pollution periods (Zhang et al., 2019a, 2021a).

The results of other research groups were further analyzed. Chang et al. (2018) selected Beijing, Tianjin, and Shijiazhuang as target cities, and the transport flux in the base year of 2010 was utilized using the WRF-CMAQ model. Beijing was mainly affected by inflow from Baoding and Zhangjiakou in winter and Langfang and Baoding in summer. Three major pathways were identified. In addition, transport during polluted periods was significantly stronger than that during clean days (Chang et al., 2018). The seasonal and annual transport fluxes of $PM_{2.5}$ in “2+26” cities in the base year of 2015 were also analyzed using the WRF-Chem model, and the transport pathways were investigated. Results showed that the seasonal horizontal $PM_{2.5}$ fluxes ranked in the order of October > April >January >July, differing from the results of Zhang et al., possibly due to the choice of study period. Several pathways at the boundary of the “2+26” cities were identified, similar to previous studies (Qi et al., 2021).

As mentioned above, the observational data can accurately quantify the horizontal and vertical transport characteristics of air pollutants at the interface. However, complex observation schemes are difficult to implement over large areas. In contrast, air quality models have the advantage of flexibility, although previous studies have demonstrated a certain deviation between the simulated and observed values. This is closely related to the incompleteness of the model’s physical diffusion and chemical transformation mechanisms. Overall, it is reasonable to carry out transport flux research based on air quality models and verify the results with observations.

3. Transport analysis based on observation

Vehicle-mounted and ship-borne instruments and aerial surveys have been applied to investigate the horizontal and vertical transport of air pollutants along the measurement route. Portable weather monitors can be utilized to obtained meteorological datasets such as temperature and relative humidity with resolution ranging from 1 to 10 sec. $PM_{2.5}$ and O_3 concentrations can be measured by aerosol monitors and O_3 photochemistry monitors with time resolution of 10 sec. The major components of aerosol, including BC, sulfate, nitrate, ammonium, chloride, organics and so on forth, could be measured by on-line observation equipment with time resolution of 1 min (in’t Veld et al., 2021; Tian et al., 2019; Wang et al., 2021a). Vertical observation is also considered one of the primary technical methods for exploring the diffusion of air pollutants (Kumar et al., 2017; Qu et al., 2021a; Wang et al., 2021a). Ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) and scanning lidar systems were found to contribute to a better understanding of air pollutant distribution (Itahashi and Irie, 2022; Xu et al., 2021; Huang et al., 2021; Xian et al., 2020). In addition, the column concentration of air pollutants could be obtained by applying satellite inversion technology, which thus provides a supplement for understanding the vertical characteristics of air pollution (Georgoulias et al., 2020; Wu et al., 2021).

A multidimensional observational project, comprising surface measurements, a microlight aircraft, a helium balloon, and remote sensing data, was conducted in southern Spain to investigate the transport of O_3; the local/remote source contributions were clarified during a pollution episode (in’t Veld et al., 2021). Tian et al. (2019) conducted an aircraft measurement to investigate the transport of air pollution in December 2016 in Beijing. The chemical composition and size distribution of the aerosols at various heights were observed. The differences in the vertical aerosol profiles before, during, and after the pollution periods were further analyzed. This demonstrates the influence of air pollutants transported from high levels on the air quality in Beijing. Multiple lidars were used to investigate the vertical profiles and regional transport of aerosols and ozone in the YRD region during the September 4–5, 2016 G20 summit. It was found that vertical and horizontal transport mainly occurred at a height of 1–2 km during the ozone pollution periods, affected by the northeast wind (Wang et al., 2021e). The interaction between aerosols and the planetary boundary layer (PBL) has played an important role in accumulating atmospheric pollution in urban areas (Huang et al., 2020; Lv et al., 2020a; Wang et al., 2020b). This has a significant effect on the long-range transportation of air pollutants. The aircraft meteorological data relay includes the flight phase (ascent and descent), time, latitude, longitude, absolute altitude, temperature, pressure altitude, wind speed, and direction, collected at a time resolution of 35 sec during the ascending phase and 60 sec during the descending phase (Lv et al., 2020b). Moreover, this data can be applied to the study of vertical structure and atmospheric boundary layer-free troposphere exchange, potentially providing data support for the interaction between the PBL and aerosol pollution (Jin et al., 2021). According to our previous research results, we analyzed the material exchange between the atmospheric boundary layer and free troposphere of typical cities based on aircraft meteorological data relay and air quality models. It was found that differences in pollution transport characteristics between Beijing and Shijiazhuang were mainly reflected in the exchange effect. The free troposphere mainly flowed into the atmospheric boundary layer in Beijing, while outflow from the atmospheric boundary layer to the free troposphere in Shijiazhuang was more intense. This is closely related to the vertical wind profiles.

Regional transport can also be studied and judged based on the concentration of pollutants at ground observation sites, combined with the meteorological conditions and the evolution of the meteorological field (Han et al., 2015; Zhao et al., 2020). Mathematical models based on observational data have also been applied to pollutant transport research. Distributed lag nonlinear models can characterize the nonlinear exposure-lag-response relationship between air pollutant concentrations and influencing factors. Duan et al. (2021) used the wind direction as a dummy variable to recognize the effect of transport and wind speed factor on $PM_{2.5}$ and O_3 in Handan, identifying the major wind direction associated with high concentrations. A mathematical method was established based on air pollutant concentration information from two urban observation stations and a background observation station, and the minimal local contribution and maximum transport effect of urban Beijing were obtained with hourly resolution (Ge et al., 2018). A complex network method was built to characterize the transport of $PM_{2.5}$ in the YRD region. An improved ranking algorithm was applied by considering two metrics to identify the important nodes in the network, yielding a north-south transport pattern in YRD regions (Wang et al., 2021c). These approaches provide new methods for calculating transport based on observations.

4. Pollutant transfer based on MRIO models

From physical diffusion and chemical mechanisms perspectives, air pollutants can be transported to areas hundreds of kilometers away, affecting the air quality in urban centers. In addition, from a trade perspective, the impact of supply and demand, productivity level, and other factors will also lead to emissions transfer. In particular, previous studies have shown that cross-border air pollution is a significant concern (Holloway et al., 2002; Mo et al., 2021). Pollutant transfer amount calculations between different regions through trade characteristics can provide a new point of view for the study of pollutant transport across regions.

Single region input-output (SRIO) and MRIO models have been applied in trade-based air pollutant transport analyses. The SRIO model represents the relationship between the material flow and transformation between different industry sectors in a single region. Previous studies have applied this model to investigate the carbon transfer process in a typical region or country (Weber and Matthews, 2007; Sun et al., 2017; Machado et al., 2001). The transfer of carbon emissions from bilateral trade is also discussed, revealing that, due to the differences in emission control technologies in various countries, the impact of bilateral trade on global carbon emissions significantly differs (Shui et al., 2006; Jayanthakumaran et al., 2016). The MRIO model supplements the intermediate input-output matrix and expands the product flow in different sectors in a single country to the product flow in different sectors in multiple countries; that is, compensating for the defects of different pollution emission coefficients in different countries. This model improves the accuracy of the implicit transfer of air pollutants in multiple regions. Some studies have focused on carbon emissions transfer on a global scale. The relationship between developed and developing countries has been studied, indicating that the responsibility of carbon emissions reduction should be shared (Davis and Caldeira, 2010; Chen et al., 2011; Zhu et al., 2018). In addition to carbon emission transfer analysis, conventional pollutants including SO_2, NO_x, PM_{10}, and non-methane VOCs (NMVOCs) have also been discussed. It was pointed out that China has become an exporter of implied SO_2, NO_x, and PM_{10} emissions through export products and an importer of implied NMVOC emissions through imported products. The European Union, East Asia, and the United States import China's energy-intensive products (such as heavy industry, mining and processing, electricity, gas, and water supply), accounting for approximately 70% of China's consumer air pollutant emissions (Wang et al., 2020a; Li et al., 2019). These results explain China's high emission load from a trade perspective. As such, these results can provide an in-depth understanding of the amount of pollutant transfer caused by trade between regions or countries.

5. Conclusions and perspectives

Atmospheric pollution is often a regional process. Clarifying the characteristics of atmospheric pollutant transport across borders is a vital prerequisite for regional joint prevention and control. In this paper, studies focused on the regional transport of air pollutants during past years were reviewed, and the advantages and disadvantages of the evaluation of the transport of air pollutants using different technical methods were discussed by combining them with the results of our research group.

In general, both observational technology and simulation technology can successfully characterize the transport of atmospheric $PM_{2.5}$ across boundaries. Based on ground observations, vertical observations, and the application of different mathematical algorithms, the characteristics of air pollutant transport can be objectively elucidated. However, the design of observational experiments has limitations in terms of the temporal and spatial sets. Air quality models can better compensate for this defect, but due to the imperfect physical diffusion and chemical mechanisms in the meteorological and air quality models, especially the representation of the secondary pollutant concentrations and wind field simulation performance, deviations between observations and simulations will occur. This is more noticeable during high-concentration periods. Therefore, it is necessary to select suitable technologies for the evaluation of air pollutants to carry out research on transport characteristics, especially to distinguish simplified scenarios and complex reduction scenarios, as well as air pollutants with simple and complex generation mechanisms. In addition, we should comprehensively consider the advantages of observation and simulation technologies, forming an air pollutant transport evaluation method based on the interactive verification of stereo observation and air quality simulation. The transportation technology system can quantitatively study the transport of air pollutants at different scales. In addition, the technical methods used in pollutant transfer volume research based on implicit trade can also better characterize pollutant transfer from an economic perspective on regional or international scale, providing novel ideas and technical support for transnational pollutant transportation research.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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