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The effect of cross-regional transport on ozone and particulate matter pollution in China: A review of methodology and current knowledge

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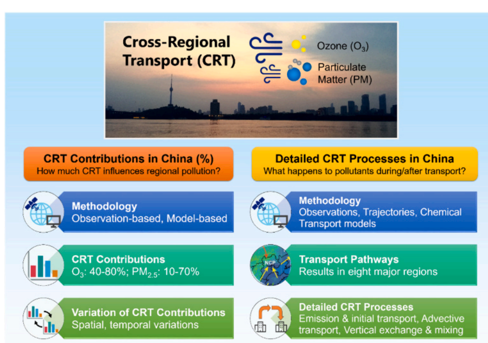
HIGHLIGHTS

- Methods and findings on cross-regional transport (CRT) influence on O_3 and particulate matter (PM) in China are summarized.
- The CRT contribution to O_3 and PM shows spatiotemporal variation.
- Transport pathways of air pollutants in the main regions have been identified.
- Studies on detailed CRT process reveal its complexity in dynamics and chemistry.
- A deeper understanding of CRT influence is needed for future air quality improvement.

GRAPHICAL ABSTRACT



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ABSTRACT

China is currently one of the countries impacted by severe atmospheric ozone (O_3) and particulate matter (PM) pollution. Due to their moderately long lifetimes, O_3 and PM can be transported over long distances, cross the boundaries of source regions and contribute to air pollution in other regions. The reported contributions of cross-regional transport (CRT) to O_3 and fine PM ($PM_{2.5}$) concentrations often exceed those of local emissions in the major regions of China, highlighting the important role of CRT in regional air pollution. Therefore, further improvement of air quality in China requires more joint efforts among regions to ensure a proper reduction in emissions while accounting for the influence of CRT. This review summarizes the methodologies employed to assess the influence of CRT on O_3 and PM pollution as well as current knowledge of CRT influence in China. Quantifying CRT contributions in proportion to O_3 and PM levels and studying detailed CRT processes of O_3, PM and precursors can be both based on targeted observations and/or model simulations. Reported publications indicate that CRT contributes by 40–80 % to O_3 and by 10–70 % to $PM_{2.5}$ in various regions of China. These contributions exhibit notable spatiotemporal variations, with differences in meteorological conditions and/or emissions often serving as main drivers of such variations. Based on trajectory-based methods, transport pathways contributing to O_3 and PM pollution in major regions of China have been revealed. Recent studies also highlighted the important role of horizontal transport in the middle/high atmospheric boundary layer or low free troposphere, of vertical exchange and mixing as well as of interactions between CRT, local meteorology and chemistry in the detailed CRT processes. Drawing on the current knowledge on the influence of CRT, this paper provides recommendations for future studies that aim at supporting ongoing air pollution mitigation strategies in China.

1. Introduction

Ambient ozone (O_3) and particulate matter (PM) are key pollutants linked to poor air quality in different regions around the globe (Fowler et al., 2020). High levels of O_3 and PM are harmful to human health, potentially causing damage to the cardiovascular and respiratory systems of humans (World Health Organization, 2021). It is estimated that exposure to outdoor air pollution leads to 8.8 million global premature deaths per year (Lelieveld et al., 2020). Additionally, both O_3 and PM influence radiative forcing, inducing changes in the regional and global climate (Fuzzi et al., 2015; Lu et al., 2019a).

Given the moderately long lifetimes of O_3 and PM (Seinfeld and Pandis, 2016), they can be transported in the atmosphere over tens, hundreds, or even thousands of kilometers away from where they are emitted or chemically produced. In this paper, long-distance transport of pollutants on the scales of tens to thousands of kilometers that crosses regional boundaries and contributes to O_3 and PM pollution in other downwind regions is defined as cross-regional transport (CRT). Such a process is also called long-range transport, cross-boundary transport, or other similar terms in previous publications. In theory, for both pollutants, the role of transport is similarly important as the role of local emission and/or chemical transformation (Vilà-Guerau de Arellano et al., 2015). While the influence of the latter two processes has been discussed in depth in earlier studies (e.g., Hallquist et al., 2009; Guo et al., 2014; Zhang et al., 2015a; Li et al., 2017a; Archibald et al., 2020), efforts are also needed to explore the complex influence of CRT on regional O_3 and PM pollution, which are normally studied from the following two perspectives:

- *The contribution of CRT*: Quantitative results regarding the proportion of O_3 or PM levels attributed to CRT within the studied region.
- *The detailed process related to the CRT of pollutants*: Qualitative and/or quantitative illustrations of the dynamic and chemical processes related to CRT of O_3, PM and their precursors.

Analyses of these topics can enhance our understanding on the causes of O_3 and PM pollution, and also support the practice of regional joint prevention and control of air pollution.

The influence of CRT on O_3 and PM pollution has been studied for several decades, and the complexity of this topic has been gradually revealed. For a given region, the contribution of CRT is determined by specific meteorological conditions, pollutant emissions and their distribution, and regional topography. Its variations in the short and long terms are often linked to the changes in meteorology and/or emissions. Explorations are often required to attribute the CRT contribution and its variations within the region to specific meteorological parameters, emission sources, and other factors. The detailed processes of CRT related to O_3 and PM pollution are often complex and cannot be fully unveiled only by Lagrangian-based trajectory analyses. Specifically, the dynamic processes involved in pollutant transport result from the combined effects of air movements on the small, meso-, synoptic, large and global scales (for details, see Section 2.1). During transport, O_3, PM and other related pollutants are produced and/or transformed through chemical reactions and/or microphysical processes, and thus their levels and composition may change continuously in the transported air parcels (for details, see Sections 2.2–2.3). In addition, pollutant transport influences the chemistry of pollutants in the receptor regions through multiple mechanisms (for details, see Section 2.4). Therefore, more in-depth studies based on state-of-the-art observations and models are needed for a systematic understanding of the influence of CRT on O_3 and PM pollution.

One of the most impacted areas by O_3 and PM pollution in the globe is China (Cheng et al., 2016; Lu et al., 2018). The combined effects of intensive anthropogenic emissions, meteorological conditions and topography favorable for the production of pollutants and/or unfavorable for their dissipation lead to frequent O_3 and PM pollution episodes in China, especially in the main city clusters including the North China Plain (NCP), the Yangtze River Delta (YRD), the Pearl River Delta (PRD), the Sichuan Basin (SCB) and the Guanzhong Basin (GZB) (Li et al., 2019a; Zhai et al., 2019). To improve air quality, emission reduction is required, and incipient efforts focusing on local emission control have led to notable decreases in the concentrations of fine PM (or $PM_{2.5}$, PM with diameters smaller than 2.5 μm) since 2013 (Zhai et al., 2019; Wang et al., 2020a). However, as reductions in local pollutant emissions become more difficult, the role of CRT contribution in PM pollution should also be a concern. This situation seems to be applicable for the populated East China or the so-called "gigacity" (Kulmala et al., 2021), where intensive anthropogenic emissions are widely distributed and pollutants can be easily transported from one region to another. At the same time, O_3 pollution has become more severe in China despite effective emission controls over O_3 precursors (Li et al., 2019a; Wang et al., 2020a). Since O_3 is mainly contributed by the transport of non-local O_3 and/or precursors in China (Liu et al., 2020a), the role of CRT in O_3 pollution and its variations might be crucial. These changes highlight the necessity to further explore the influence of CRT on regional O_3 and PM pollution in China.

Many observational and modeling studies on the influence of CRT on O_3 and PM pollution in China have been reported in recent years. This review aims to present recent advances in our knowledge through a systematic summary of these studies and shed light on existing gaps for further studies and air quality management in China. The methodology and scientific questions to be addressed in the paper can also provide new research directions concerning the influence of CRT in other countries and regions.

This paper is structured as follows. Section 2 outlines the dynamic and chemical processes related to the transport of O_3, PM and their precursors in the atmosphere, serving as the fundaments for relevant research. The next two sections aim to provide detailed information on the methodologies and current knowledge from the aforementioned two perspectives: Section 3 focuses on the contribution of CRT to O_3 and PM in China, and presents the quantification methods used, relevant results for the main regions and their spatiotemporal variations; Section 4 focuses on the detailed processes of CRT that contribute to O_3 and PM pollution in China, and summarizes the adopted observational and modeling methods, the trajectory-based transport pathways for different regions of China, along with main findings regarding the partial and complete CRT processes of pollutants. Concluding remarks and future perspectives on this topic are provided in Section 5.

2. Processes related to the atmospheric transport of O_3, PM and their precursors

2.1. Dynamic processes driving atmospheric transport

Generally, the longer the atmospheric lifetime of a pollutant is, the more likely it is transported over a long duration and distance driven by larger-scale dynamic processes. O_3, PM (especially $PM_{2.5}$) and their precursors (including nitrogen oxides ($NO_x = NO+NO_2$), carbon monoxide (CO), sulfur dioxide (SO_2), ammonia (NH_3), and volatile organic compounds (VOCs)) are mostly moderately long-lived species in the atmosphere, with lifetimes ranging from a few hours for NO_x, NH_3 and several VOCs (Seinfeld and Pandis, 2016; Dammers et al., 2019; Lange et al., 2022) to ~22 days for tropospheric O_3 (Stevenson et al., 2006; Young et al., 2013) and ~ 2 months for CO (Khalil and Rasmussen, 1990). Thus, the CRT processes of O_3, PM and precursors can be controlled by air movements on the micro-, meso-, synoptic, large and even global scales (Fig. 1), of which more descriptions are given below.

2.1.1. Atmospheric turbulence

Atmospheric turbulence drives pollutant transport on the micro- or small scales within the atmospheric boundary layer (ABL) (Lee, 2018). Strong turbulence mixes pollutants well in the convective ABL. In particular, it enhances the mixing of O_3 precursors and alters O_3 sensitivity to precursors, potentially leading to more rapid O_3 production and severer O_3 pollution in polluted urban areas (Tang et al., 2017; Tang et al., 2021). In contrast, weak turbulence and corresponding low ABL height often promote the accumulation and chemical production of $PM_{2.5}$ near the ground, resulting in the occurrence of $PM_{2.5}$ pollution event, or haze (Ren et al., 2019a; Peng et al., 2021; Ren et al., 2021; Lin et al., 2022). But besides weak turbulence, the intermittent bursts of turbulence were also reported during heavy hazes, which promote the dissipation of near-ground pollution (Wei et al., 2018; Ren et al., 2019a; Ren et al., 2019b). The diurnal cycle of the ABL is linked to turbulence changes in the ABL throughout the day (Lee, 2018), and it has a marked effect on the development of O_3 and PM pollution. In the morning, the development of thermal turbulence results in ABL expansion. Subsequently, pollutants from the residual layer and free atmosphere are entrained into the ABL. Several studies reported that entrainment

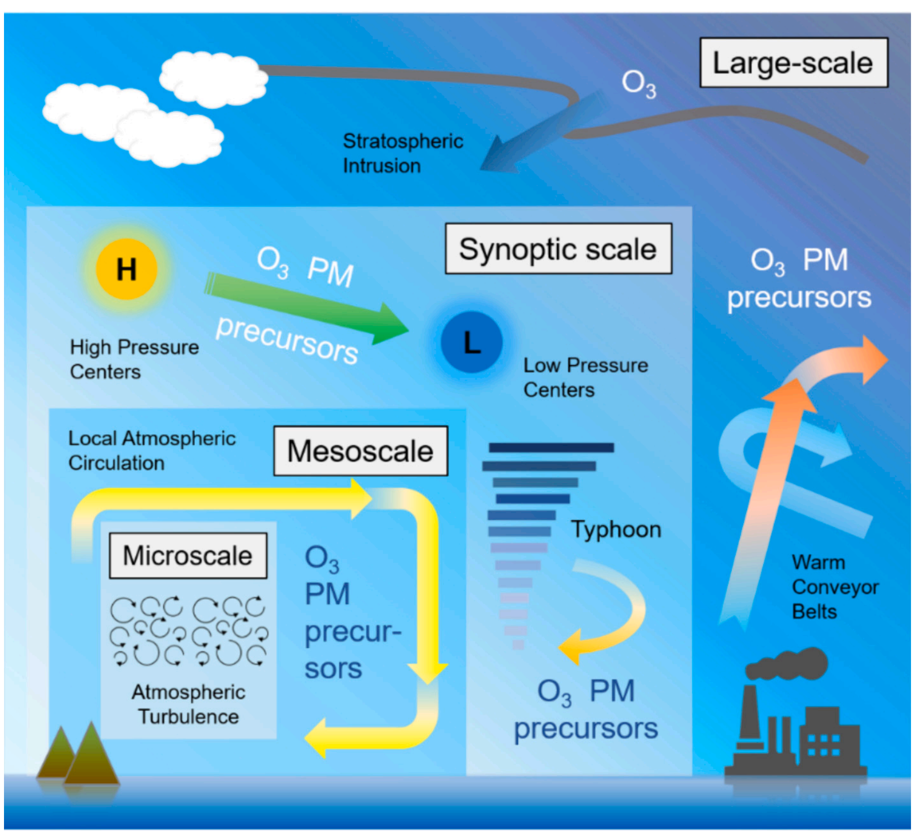

Fig. 1. Illustration of dynamic processes on multiple scales related to the CRT of O_3, PM and their precursors. Dynamic processes displayed in this figure include: atmospheric turbulence on the microscale; local atmospheric circulation on the mesoscale; high/low pressure centers (denoted in the figure as "H" and "L", respectively) and typhoon as typical systems on the synoptic scale; warm conveyor belts and stratospheric intrusion on the large scale.

contributes to the rapid increase of O_3 concentrations after sunrise (Morris et al., 2010; Kaser et al., 2017; Hu et al., 2018; Xu et al., 2018; Zhu et al., 2020a; Qu et al., 2023), and near-ground PM and some of its components (e.g., nitrate and secondary organic aerosol (SOA)) may be considerably contributed by this process as well (Curci et al., 2015; Janssen et al., 2017; Jin et al., 2021; Lampilahti et al., 2021; Tan et al., 2021; Jin et al., 2022). In the afternoon, turbulence is weakened with the progressive decrease in solar radiation, leading to shallower ABL. At night, the ABL becomes stable due to weak turbulence. These afternoon and night-time changes may not directly result in notable variations of pollutant concentrations (Qu et al., 2023), but enhance the accumulation of primary pollutants from near-ground emission and secondary PM from chemical production. It should be noted that pollutants can be rapidly transported downwards or upwards at night by the low-level jet, which is a fast-moving stream in the lower troposphere that is linked to night-time O_3 pollution and PM dissipation within the ABL (Hu et al., 2013; Klein et al., 2014; Miao et al., 2019; Wei et al., 2023; Wu et al., 2023).

2.1.2. *Local atmospheric circulations*

Sea (lake)-land breeze, mountain-valley breeze and urban heat island circulation drive mesoscale pollutant transport, especially when the circulations on the synoptic and large scales are relatively weak. Under their effects, pollutants are re-distributed, and pollutant concentrations may increase or decrease at a specific site. Some examples of such effects on regional O_3 and PM pollution are presented below. Sea or lake breezes can bring high-level O_3 above the sea or lake back to the land areas, resulting in late O_3 peak times and abnormally high O_3 in the afternoon or at night in coastal regions (Oh et al., 2006; Martins et al., 2012; Zhang et al., 2017a; Finardi et al., 2018; Zhao et al., 2022). Valley breezes may bring pollutants out of cities at lower altitudes during the daytime, thus diluting the pollution in urban regions. However, at night, mountain breezes may transport these pollutants back, exacerbating PM pollution or extending the duration of O_3 pollution (Miao et al., 2015; Bei et al., 2017; Bei et al., 2018). It is also possible that under valley breezes, pollutants are lifted to a height of 2–3 km and then transported over longer distances, leading to pollution over larger areas (Chen et al., 2009). Urban heat island enhances the transport of polluted urban air masses upwards and the transport of clean air masses from the outskirts, and thus it may reduce pollutant concentrations in cities to some extent (Huang et al., 2019a; Kang et al., 2022). Moreover, dynamically-driven mesoscale processes, such as mountain waves, foehn winds and lee waves, may also be linked to the complex processes of pollutant transport in mountainous regions (Steyn et al., 2013).

2.1.3. *Synoptic systems*

Typical synoptic systems include cyclones, anticyclones and fronts. When there is no significant increase or reduction of emissions, the changes in synoptic systems often drive the short-term evolution of regional O_3 and PM pollution. This is also the reason why pollution episodes often last for 3–8 days (Zhao et al., 2015a; Li et al., 2017b; Zhou et al., 2018a), consistent with the typical influencing periods of synoptic systems. The effect of synoptic systems leading to pollution is generally linked to meteorological conditions that favor the production and/or accumulation of pollutants (e.g., higher temperature, stronger radiation and lower wind speeds (stagnation) for O_3 pollution; higher humidity, shallower ABL and lower wind speeds (stagnation) for PM pollution (Chen et al., 2020; Nguyen et al., 2022)). For example, under uniform pressure fields or anti-cyclones, the precursors of O_3 and PM accumulate locally and react to produce O_3 and PM; thus, air pollution is likely to occur when these synoptic systems dominate (Hsu and Cheng, 2016; Yu et al., 2017; Lin et al., 2019; Xie et al., 2019; Graham et al., 2020; Xiao et al., 2021; Shi et al., 2022; Yan et al., 2023). However, enhanced transport of pollutants due to the changes in wind speed and/or transport pathway could be another cause of pollution. For relatively clean regions, some specific settings of pressure fields (e.g., low pressure near the clean region and high pressure near the polluted region) may result in the transport of pollutants from upwind polluted regions, thus triggering pollution in the receptor regions (Wu et al., 2022). Under fast-moving cold fronts, $PM_{2.5}$ can be transported over hundreds or thousands of kilometers, resulting in the rapid development of haze in the downwind regions (Huang et al., 2020; Kang et al., 2021; Zhang et al., 2021a). O_3- or PM-enriched air may be transported downwards by the downdrafts behind the cold front, contributing to near-ground air pollution (Kunz and Speth, 1997; Carmichael et al., 1998; Sun et al., 2023); after the passing of cold fronts, PM pollution may continue due to the effect of weak winds (Zhang et al., 2022a; Sun et al., 2023). Some synoptic systems may considerably influence the production, accumulation and transport of pollutants at the same time. A typical example is the periphery of typhoons (tropical cyclones) that frequently lead to O_3 pollution in southeast China (Chen et al., 2018a; Deng et al., 2019; Qu et al., 2021a). It enhances the horizontal transport of O_3 and precursors from central and even northern China as well as downward O_3 transport under downdrafts (Qu et al., 2021a). Changes in wind fields indicate varying accumulation conditions for O_3. Besides, higher temperatures, lower cloud covers and stronger solar radiation enhance O_3 production, also contributing to severer O_3 pollution under the typhoon periphery. It should be noted that the role of synoptic systems in O_3 and PM pollution is regionally specific. Even for the same region, differences may also exist between the effect of synoptic systems in various seasons, different stages of pollution and for O_3 and $PM_{2.5}$ pollution.

2.1.4. *Large-scale transport processes*

Last but not least, O_3 and PM can also be transported on the large and global scales. Driven by systems like warm conveyor belts (WCB; strongly rising airflows in extratropical cyclones) and deep convection, near-ground pollutants can be brought into the middle and/or upper troposphere (Cooper et al., 2004; Ding et al., 2009; Bozem et al., 2017). Without notable influence from the ground, air movements in the middle and upper troposphere are rapid and follow the global atmospheric circulation patterns, thus enabling the inter-continental transport of pollutants (National Research Council, 2010). Specifically, westerlies lead to the eastward inter-continental transport of pollutants in the mid-latitude areas. Examples of such a process in the northern hemisphere include the transport of O_3 and PM from Europe and Africa to Asia (Dong et al., 2018; Han et al., 2018a; Ni et al., 2018), the transport of O_3, peroxyacetyl nitrates (PANs), dust and anthropogenic $PM_{2.5}$ from East Asia to the west coast of North America (Jaffe et al., 1999; Liu et al., 2003; Uno et al., 2009; Guo et al., 2017), and the transport of wildfire plumes from North America to Europe (Forster et al., 2001; Ancellet et al., 2016; Markowicz et al., 2016). Another important large-scale process related to O_3 transport is stratospheric intrusion (SI), which is one of the main processes of stratosphere-troposphere exchange (Stohl et al., 2003). Since O_3 levels are higher in the stratosphere, the intrusion of O_3-enriched air may notably contribute to high O_3 levels in the middle and upper troposphere or even near the ground (Lu et al., 2019b; Tarasick et al., 2019; Roux et al., 2020; Daskalakis et al., 2022).

2.2. *Chemical processes during atmospheric transport*

As displayed in Fig. 2, pollutants still undergo active transformation through chemical reactions during transport, which are outlined in the following text.

2.2.1. *O_3 production*

During transport, O_3 is formed in the presence of VOCs and NO_x. Ongoing O_3 production along the transport partially explains why O_3 pollution in the downwind regions can be more pronounced than near the emission sources (Xu et al., 2011; Ge et al., 2012). If the transport process continues without the injection of new emissions, O_3 production is likely to slow down, and tends to become NO_x-limited (Robinson et al.,

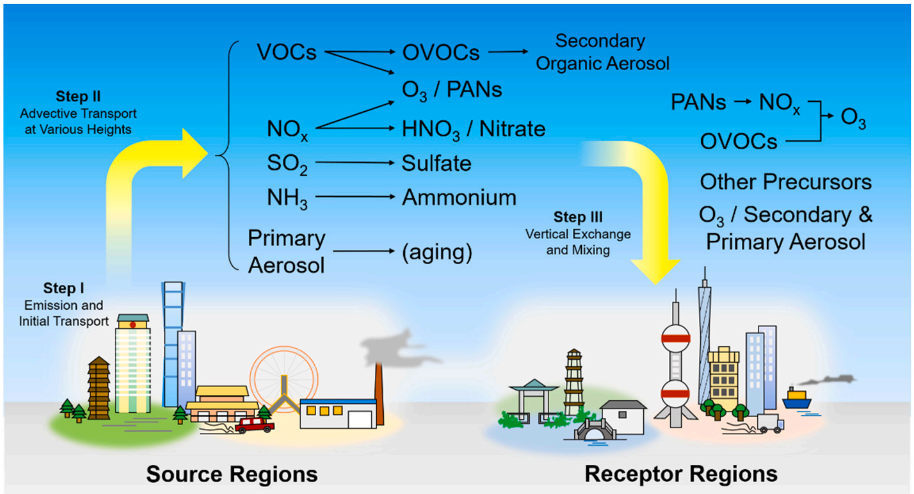

Fig. 2. Illustration of three steps in the cross-regional transport and chemical processes of O_3, PM and their precursors during transport. VOCs, volatile organic compounds; OVOCs, oxygenated VOCs; PANs, peroxyacetyl nitrates.

2021; Schneider and Abbatt, 2022). Besides O_3, PANs can also be produced from the reactions between VOCs and NO_x during transport.

2.2.2. PM formation

Secondary PM components, including particulate nitrate, ammonium, sulfate and secondary organic aerosol (SOA), can also be notably produced from their corresponding precursors in the transported air parcels. Specifically, NO_x is oxidized through multiple reaction pathways (e.g., OH oxidation and N_2O_5 hydrolysis) to produce HNO_3, which partially partitions into the aerosol phase as particulate nitrate. By reacting with acid gases, NH_3 is transformed into particulate ammonium. Considering the influence of environmental conditions (e.g., temperature and humidity) on the equilibrium between gas-phase HNO_3, NH_3, and particle-phase nitrate, ammonium, the relative proportions of these species are likely to change during transport (Sasaki et al., 1988; Itahashi et al., 2017). SO_2 is oxidized into particulate sulfate during transport, but often at a slower rate than NO_x to produce particulate nitrate (Rodhe et al., 1972; Altshuller and Linthurst, 1984; Sasaki et al., 1988; Stuart, 1988). Bae et al. (2017) and Du et al. (2020a) found that most particulate sulfate in Seoul and the NCP was derived from SO_2 oxidation during transport, highlighting its potentially important contribution to regional PM pollution. Besides, VOCs can react with oxidants (e.g., OH, NO_3, O_3) to generate oxygenated VOCs (OVOCs), and the low-volatility products of these reactions can be further involved in SOA formation (Apel et al., 2010; National Research Council, 2010). Primary PM components, e.g., black carbon (BC), sea salt and mineral dust aerosols, are relatively stable in comparison to secondary PM. However, it is possible that their microphysical properties, such as mixing state and hygroscopicity, gradually change due to the aging processes during transport (Zhang et al., 2021a). Primary organic carbon (POC), due to its semi- or low volatility, is also known to evolve in the aging processes during transport (Robinson et al., 2007).

It should be noted that the above chemical processes do not occur in the same stages of transport. Normally, the longer the atmospheric lifetime of the precursor is, the more likely that its chemical transformation lasts for a longer time and even considerably influences the chemical production of O_3 and PM in the downwind regions (details given in Section 2.4). Due to the above O_3- and PM-related chemical processes, transported air parcels are normally aged, which can be characterized by high O_3 (or O_x, $O_3 + NO_2$) levels and/or the dominance of secondary components in $PM_{2.5}$ (Xu et al., 2011; Park et al., 2013; Han et al., 2015a). Moreover, chemical processes during transport may also lead to higher toxicity of pollutants and, thereby, increased health risks for humans (Wong et al., 2019).

2.3. Chemical/microphysical processes favoring atmospheric transport

For pollutants with short atmospheric lifetimes, reactions with oxidants limit their transport duration and distance. However, some chemical or microphysical transformations could enable them to be transported at longer distances. A typical example is the transport of short-lived NO_x in the form of PANs. As mentioned before, PANs are produced via the photochemical oxidation of VOCs with NO_x. PANs have relatively long lifetimes in cold conditions and thus they can be transported at high altitudes to the areas far from where they are produced (Altshuller, 1993). After reaching the receptor regions, PANs may release NO_x through photolysis and/or thermal decomposition, and thereby contribute to downwind O_3 production. This has been reported as the classic mechanism that NO_x emissions in East Asia contribute to O_3 pollution on the west coast of America (Hudman et al., 2004). Another example is the facilitation of SOA transport by trapping polycyclic aromatic hydrocarbons (PAHs) inside (Friedman et al., 2014; Shiraiwa et al., 2017; Shrivastava et al., 2017). The reason is that PAHs diminish SOA volatilization and reduce SOA loss during transport. At the same time, SOA effectively reduces the collision between PAHs and atmospheric oxidants, and thus promotes PAHs transport as well.

2.4. Influence of atmospheric transport on local chemistry

Atmospheric transport may considerably influence local O_3 and PM chemistry in the receptor regions, which, in previous publications, was defined as the indirect effect of transport (or indirect transport; Chen et al., 2014). Such an effect is linked to multiple processes, e.g., the removal of transported O_3 by local NO_x emissions as well as the facilitation/hindrance of O_3 and PM production through the import of precursors and other related species. By interacting with local meteorology, the transport of PM also possibly influences the chemical processes of O_3 and PM. More descriptions of transport influence on local O_3 and PM chemistry are presented as follows.

2.4.1. Influence of atmospheric transport on local O_3 chemistry

O_3 pollution can be aggravated by local O_3 production contributed by O_3 precursors (including NO_x, PANs, hydrocarbons and OVOCs) transported from upwind regions (Wang et al., 2022a; Qu et al., 2023). However, indirect transport may lead to O_3 reduction in some urban areas, which suggests a greater influence from the titration of transported O_3 by locally emitted NO_x (Wu et al., 2017a).

2.4.2. Influence of atmospheric transport on local PM chemistry

Local PM chemistry is affected by indirect transport through more diverse mechanisms than O_3. Among $PM_{2.5}$ components, particulate nitrate is normally most contributed by indirect transport (Chen et al., 2014; Uranishi et al., 2018; Qu et al., 2021b; Sun et al., 2022a; Xu et al., 2023). The related mechanisms could be the reaction between transported HNO_3 with locally emitted NH_3 (Uranishi et al., 2018), the enhancement of nitrate production through the transport of more hygroscopic $PM_{2.5}$ components (Seo et al., 2020), or N_2O_5 hydrolysis between transported O_3 and local NO_x (Qu et al., 2021b). For particulate sulfate, dust transport potentially enhances its production through heterogeneous reactions of SO_2 on dust surfaces (Wang et al., 2017a). Moreover, since high humidity often enhances the production of secondary PM, the transport of water vapor may notably contribute to PM pollution in relatively dry regions (Wu et al., 2017b). Transported PM also influences local meteorology: It affects incoming radiation and heat transfer in the atmosphere, resulting in shallower ABL, weakened turbulence and increasing humidity. By enhancing the accumulation and production of PM, these changes further aggravate PM pollution (Huang et al., 2020).

To summarize, the detailed processes linked to the indirect effect of atmospheric transport on O_3 and PM are diverse and complex. Since indirect transport describes the interactions of pollutants derived from different regions, understanding its contributions and detailed processes is important for regional collaborative measures to improve air quality.

3. Current knowledge on the contribution of cross-regional transport

The contribution of CRT to O_3 and PM quantitatively characterizes the impact of CRT on pollution in the region of interest, and it is informative for both short- and long-term air quality improvements. In this section, we summarize the reported results of the CRT contribution to O_3 and PM in China from the following three perspectives: 1) methods used for the identification of the CRT contribution, 2) CRT contributions to O_3 and $PM_{2.5}$ in the main regions of China, and 3) the spatiotemporal variations of the CRT contributions.

3.1. Methods to quantify the contribution of cross-regional transport

3.1.1. Methods based on observations

The CRT contributions to O_3 and PM are often estimated based on observed pollutant concentrations, mostly following these three approaches:

(1) **Calculations based on observations at multiple sites**: It is generally recognized that pollutant levels at background sites mostly emanate from CRT, whereas these at urban sites are additionally contributed by local emissions. Therefore, pollutant concentrations at the aforementioned two types of sites can yield the local and CRT contributions. This method has been widely applied in relevant studies (e.g., Theodosi et al., 2011; Nie et al., 2013; Pikridas et al., 2018; Chatoutsidou et al., 2019; Vrekoussis et al., 2022). Based on data at multiple sites spread across a region, transport contributions from different distance ranges can be identified (Wang et al., 2020b). However, as pointed out by Ge et al. (2018), emissions within a region could also contribute to pollution in remote areas, and thus the CRT (local) contributions calculated using the above method have to be considered as their maximum (minimum) values in theory.

(2) **Decomposition using statistical methods**: By applying statistical methods, the data series of pollutant concentrations can be directly decomposed into contributions from multiple sources. The methods reported in relevant studies include wavelet analysis (Sabaliauskas et al., 2014), Fourier transform (Tchepel and Borrego, 2010), principal component analysis (PCA; Langford et al., 2009; Liang et al., 2018) and non-negative matrix factor decomposition (NMF; Luo et al., 2018; Yim et al., 2019). The first two methods decompose the series of pollutant concentrations into results with different frequencies, and these with high and low frequencies separately indicate the local and CRT contributions to pollutants. Based on observations at multiple sites within a region, PCA and NMF separate the series of pollutant concentrations into the contributions from components characterized with different spatiotemporal variations, which can further correspond to local and transport contributions.

(3) **Quantifications by combining pollutant concentrations with backward trajectories**: Under the influence of air masses transported from various directions, indicated by clustered backward trajectories (explanations given in Section 4.1), mean pollutant concentrations often differ, suggesting contributions from distinct sources to pollutant levels. For urban sites, the local contribution is estimated as the lowest value among the mean concentrations under different types of trajectories, assuming that it reflects conditions when the influence of transport is minimal and can be ignored; the increments of concentrations under other trajectories are viewed as the transport contributions. On the contrary, for background sites, the lowest mean concentration represents the background level of pollutants, and the increments of concentrations under other trajectories indicate transport contributions from different regions. Examples of its applications include Lv et al. (2015a), Wang et al. (2015a), Wang et al. (2018a), Zhang et al. (2018a), Wang et al. (2019a) and Wang et al. (2020c).

The observation-based methods enable an easy estimation of the CRT contributions to pollutants. They are also suitable for exploring long-term changes in the transport or local contributions to pollutants based on multi-year observational datasets. However, the results of these methods often fail to determine the specific sources affecting the pollutant levels, including CRT, thus limiting their usage in the practice of emission control.

3.1.2. Methods based on chemical transport models

Source apportionment based on chemical transport models (CTMs) identifies the contributions of pollutants derived from specific regions and/or sectors. Specifically, the contributions from emissions outside the studied region (including the global background contributions) can be viewed as the CRT contributions to pollutants. Three types of methods are often applied in CTMs-based source apportionment, namely, the Brute Force Method (BFM), the tagged method and the Decoupled Direct Method (DDM). Here, the fundaments of the three methods are introduced as follows. More detailed methodology, pros and cons of these methods can be found in Clappier et al. (2017), Thunis et al. (2019), Liu et al. (2020a), and Mircea et al. (2020).

(1) **Brute Force Method:** BFM is a widely used method in source apportionment. By subtracting simulation results with zeroed concerned emissions from the base simulation with all emissions considered, the contributions of the concerned emissions can be quantified. Also, they can be identified by subtracting simulation results without all emissions from simulations accounting only for the studied emissions. These two types of BFM are separately defined as the top-down and bottom-up BFM (Clappier et al., 2017). Since no additional module is needed for relevant simulations, BFM is applicable for nearly all CTMs to quantify the CRT contributions to pollutants. However, because BFM needs to run the model multiple times, its computation cost is higher than that of other methods. Another drawback is that the non-linearity between O_3, secondary PM and their precursors leads to non-additivity of the BFM results, which means that the sum of all

identified contributions differs from the concentration of O_3 and PM in the base case (Clappier et al., 2017; Thunis et al., 2019).

(2) **The tagged method:** By tagging O_3, PM and their precursors in simulations, the contributions from various sources (including the initial/boundary conditions and emissions from different regions and/or sectors) to pollutant levels can be directly provided from the model output. As the bulk species, tagged species undergo the same processes in the atmosphere, such as transport, deposition and chemical reactions. In each calculation step of these processes, the model directly attributes the change in pollutant level to source contributions. Typical examples of the tagged module include the Ozone and Particulate Matter Source Apportionment Technology (OSAT/PSAT) in the CAMx model (Yarwood et al., 2007; Yarwood and Koo, 2015), the Integrated Source Apportionment Method (ISAM) in the CMAQ model (Kwok et al., 2015), and the tagging techniques in the NAQPMS, WRF-Chem and LOTOS-EUROS models (Li et al., 2008; Kranenburg et al., 2013; Lupaşcu and Butler, 2019). Owing to its relatively low computation cost, the tagged method is efficient for source apportionment. However, given that pollutant levels cannot be negative, the contributions by applying the tagged methods are always greater than or equal to zero. Thus, the negative response of secondary pollutant to their precursors cannot be reproduced by this method (Itahashi et al., 2015; Clappier et al., 2017; Chatani et al., 2020; Fang et al., 2021).

(3) **Decoupled Direct Method:** DDM computes the first-order sensitivity (denoted as $S_{i,j}^{(1)}$) of pollutant concentration (c) to specific emissions ($E_{i,j}$; i, j separately indicate the regional and sector sources of emissions), written as (Napelenok et al., 2006):

$$S_{i,j}^{(1)} = \left.\frac{\partial c}{\partial E_{i,j}}\right|_{E_{i,j(0)}} \times E_{i,j(0)} \tag{1}$$

where $E_{i,j(0)}$ is the initial value of $E_{i,j}$ without any perturbation. To better describe the non-linear response of pollutant levels to emissions, the updated Higher-order Decoupled Direct Method (HDDM) also quantifies the secondary-order sensitivity term ($S_{i,j}^{(2)}$; Hakami et al., 2003):

$$S_{i,j}^{(2)} = \left.\frac{\partial^2 c}{\partial {E_{i,j}}^2}\right|_{E_{i,j(0)}} \times {E_{i,j(0)}}^2 \tag{2}$$

If $E_{i,j}$ changes by $\Delta\varepsilon$ relative to $E_{i,j(0)}$, according to Taylor-series expansion, the perturbed pollutant concentration (c_{pert}) can be estimated as follows (Itahashi et al., 2015; Hong et al., 2017):

$$c_{pert} \approx c_0 + \Delta\varepsilon \times S_{i,j}^{(1)} \left(+ \frac{1}{2} \times \Delta\varepsilon^2 \times S_{i,j}^{(2)} \right) \tag{3}$$

where c_0 is the unperturbed pollutant concentration. The difference between c_0 and c_{pert} can be viewed as the contributions of $E_{i,j}$ to pollutant. DDM and HDDM are also efficient source apportionment methods. However, uncertainty still exists in the estimation of source contributions to O_3 and secondary PM, even for HDDM which better captures the non-linear response of these pollutants to precursors (Itahashi et al., 2015; Huang et al., 2017).

In theory, for primary PM, the results by applying the above three methods should be consistent (Clappier et al., 2017; Thunis et al., 2019). But for O_3 and secondary PM, due to their non-linear responses to precursor emissions, discrepancies often exist when different methods are used (Clappier et al., 2017; Thunis et al., 2019). The comparisons of source contributions to O_3 and PM quantified by different CTMs-based methods were reported in some studies, such as Itahashi et al. (2015), Chatani et al. (2020) and Fang et al. (2021). Despite of disparities in the values of estimated contributions, the major regional sources for O_3 and PM in the targeted regions are overall similar when using different methods, making these results still applicable for air quality management.

CTMs-based methods quantify contributions to O_3 and PM from sources with clear definitions. These results are supportive of making effective strategies to improve regional air quality. They also consider the influence of non-transport processes, including chemistry and deposition, on O_3 and PM. However, the uncertainty of emission inventories, meteorological fields, chemical mechanisms and other parameterizations in CTMs may lead to considerable inaccuracy in results, which needs to be further improved. For O_3, different choices of chemical boundary conditions in regional CTMs simulations could cause notable differences in the background contribution as well as the CRT contribution, and thus should be of concern (Yan et al., 2021a; Li et al., 2023a; Zhu et al., 2024a).

3.2. Contribution of cross-regional transport to O_3 and $PM_{2.5}$ in the main regions of China

The contributions of CRT to O_3 and $PM_{2.5}$ have been reported in several main regions of China. Here, we only summarize CTMs-based estimates of the CRT contribution to the level of these two pollutants, which is clearly defined as the total contribution (in percentage) of sources outside the region of interest or the region where the receptor cities/sites are located. Thus, the CRT contribution includes the contribution from emissions outside the studied region and the global background, while the contribution from emissions within the studied region is hereafter called the local contribution. The CRT contributions over one or more months and mostly from papers published after 2015 are summarized and discussed in this section.

Fig. 3 displays the reported CRT contributions to the O_3 level in the three major regions of China (NCP, YRD, PRD). Due to the diverse selections of the model, source apportionment method, receptor region, studied period as well as O_3 level metric, the reported CRT contributions to O_3 show large differences. For all the three regions, the lowest reported CRT contributions are <40 %, whereas the highest values exceed 80 %. However, over 70 % of the results report that the CRT contributions to O_3 levels exceed 50 %, suggesting that CRT is the main contributor to O_3 pollution in these regions. For the seasons with the most severe O_3 pollution (summer for the NCP and YRD, summer and autumn for the PRD; Wang et al., 2022b), many studies found higher CRT contributions to O_3 than the corresponding local contributions. As part of the CRT contribution, O_3 transported from other regions in China is a considerable O_3 source in all the three regions (Shen et al., 2022). Therefore, to effectively alleviate O_3 pollution, it is required to collectively reduce O_3 precursor emissions in China.

The reported CRT contributions to $PM_{2.5}$ concentrations in multiple regions of China (NCP, YRD, PRD, SCB and others) are summarized in Fig. 4. Similar to the results of O_3, the CRT contributions to $PM_{2.5}$ vary notably between studies, ranging from ~10 % to ~70 %. Nearly half of the results indicate that the CRT contributions to $PM_{2.5}$ are close to or higher than 50 %, suggesting the important role of CRT in $PM_{2.5}$ pollution in China. However, these contributions could be less significant, and they are even lower than 20 % in some cases (e.g., Wang et al., 2014a; Qiao et al., 2019). In winter when most severe $PM_{2.5}$ pollution generally occurs (Zhai et al., 2019), the mean CRT contributions to $PM_{2.5}$ are lower than 30 % in the NCP, lower than 40 % in the SCB, close to 50 % in the YRD and higher than 60 % in the PRD. The results of the CRT contributions to $PM_{2.5}$ suggest that for some regions, reducing upwind emissions is more effective in decreasing $PM_{2.5}$ levels than reducing local emissions.

Moreover, the CRT contributions differ among $PM_{2.5}$ components. In general, the CRT contributions to $PM_{2.5}$ sulfate and nitrate are higher than those to other components in China (Zhao et al., 2012; Chen et al.,

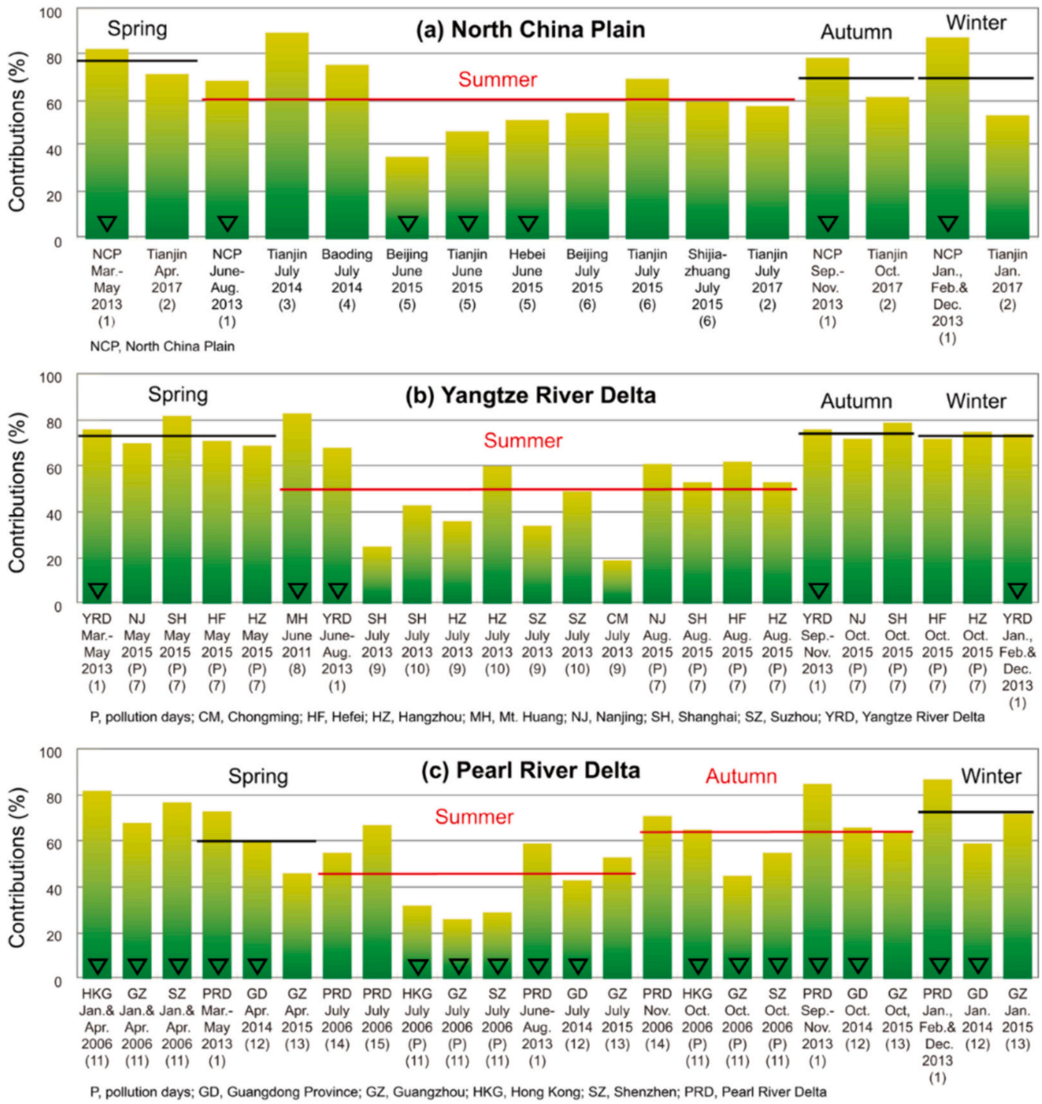

Fig. 3. Reported CRT contributions (in percentage, %) to O_3 levels in the three major regions of China: (a) North China Plain; (b) Yangtze River Delta; (c) Pearl River Delta. The lines indicate the mean CRT contributions during the corresponding seasons. Seasons with the most severe O_3 pollution in these regions are marked in red. The inverted triangles in the bars indicate that the O_3 level metric is the daily mean concentration; for the results without inverted triangles, the O_3 level metric corresponds to the daytime mean or maximum 8-h mean concentration. The references noted in the figure are as follows: (1) Gao et al., 2020; (2) Li et al., 2019b; (3) Hong et al., 2017; (4) Huang et al., 2018a; (5) Han et al., 2018b; (6) Wang et al., 2017b; (7) Li et al., 2019c; (8) Gao et al., 2017; (9) Li et al., 2016; (10) Li et al., 2015a; (11) Li et al., 2013; (12) Shen et al., 2017; (13) Yang et al., 2019; (14) Li et al., 2012a; (15) Li, 2013.

2017; Wu et al., 2017c; Wen et al., 2018; Chang et al., 2019; Du et al., 2019). High CRT contributions to $PM_{2.5}$ sulfate and nitrate were found in the PRD (> 60 % and > 80 %, respectively; Ying et al., 2014; Li et al., 2019d) compared to other regions. On the contrary, $PM_{2.5}$ sulfate in the NCP is overall less affected by CRT (< 40 %; Ying et al., 2014; Li et al., 2019d), which may be attributed to more intense local anthropogenic emissions and higher local production of $PM_{2.5}$ sulfate there.

3.3. Variations of cross-regional transport contribution to O_3 and $PM_{2.5}$

3.3.1. Spatial variations

(1) **Horizontal variation**: According to the results summarized in the last section (Fig. 3–4), the CRT contributions to O_3 and $PM_{2.5}$ levels vary between different regions. In summer, the CRT contributions to O_3 and $PM_{2.5}$ are both higher in North China (NCP) compared to South China (YRD and PRD) (Li et al., 2019d; Gao et al., 2020). It might be attributed to the influence of summer monsoon (characterized by south winds) and, thereby, increasing contributions from emissions in the upwind regions of East China to these two pollutants in North China. In winter, by contrast, the CRT contributions to $PM_{2.5}$ are higher in South China than in North China (Li et al., 2019d). This geographic pattern agrees well with the change in prevailing wind direction: Under winter monsoon (characterized by north winds), $PM_{2.5}$ emanating from emissions in East China is more likely to be transported southwards and leads to higher CRT contributions in South China. Moreover, the CRT contributions vary among provinces, cities or sites within the region. High CRT contributions are normally expected if the studied spot is located in the downwind of non-local emissions, or distant from local emission hotspots.

(2) **Vertical variation:** In the vertical direction, generally, the CRT contributions to O_3 and $PM_{2.5}$ increase with altitude (Li et al., 2017c; Ni et al., 2018; Han et al., 2019; Huang et al., 2020; Cheng et al., 2022). Relatively lower CRT contributions (or higher local contributions) near the ground are attributed to more densely distributed local pollutant emissions and slower transport due to lower wind speeds.

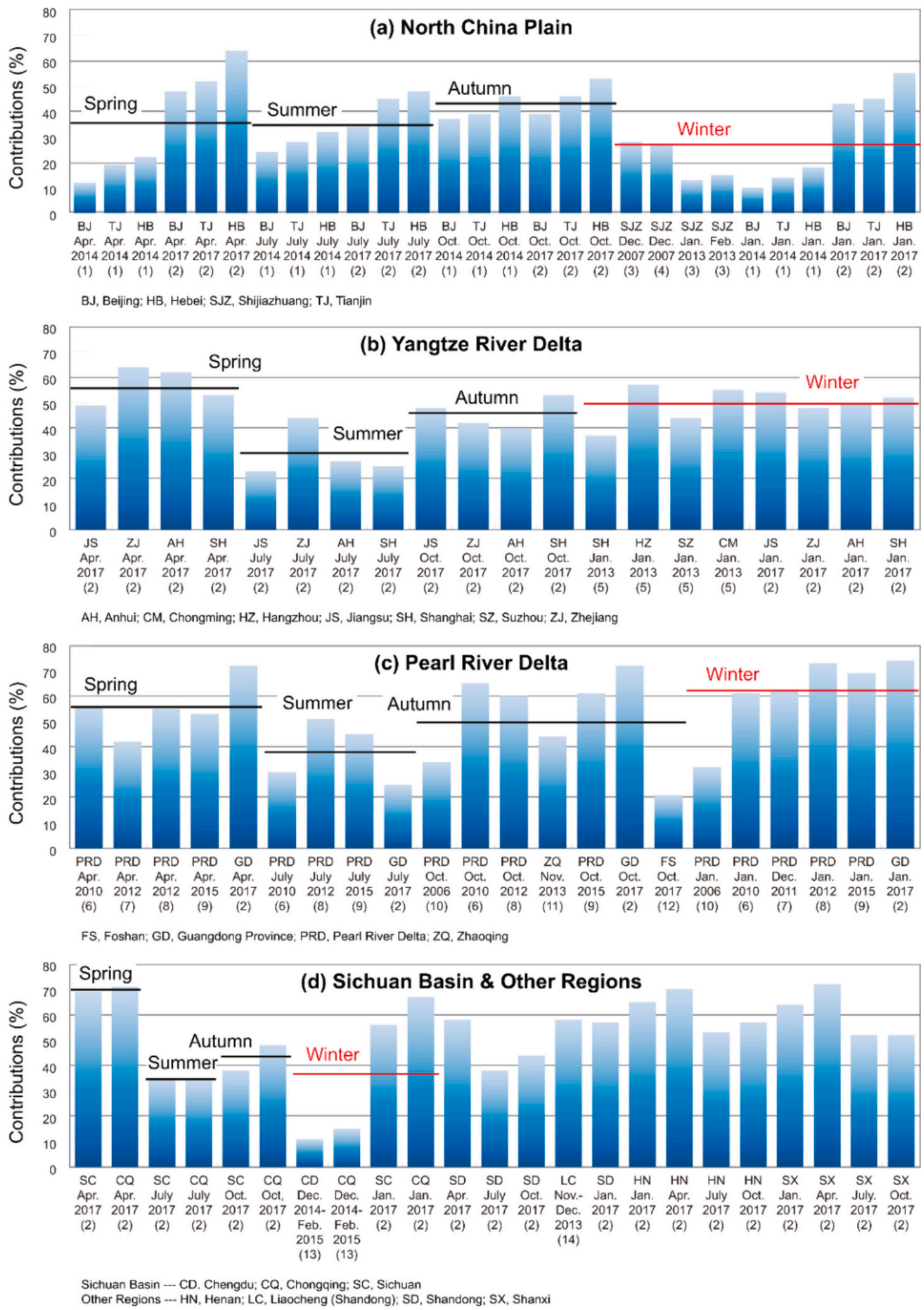

Fig. 4. Reported CRT contributions (in percentage, %) to $PM_{2.5}$ concentrations in multiple major regions of China: (a) North China Plain; (b) Yangtze River Delta; (c) Pearl River Delta; (d) Sichuan Basin and other regions. The lines indicate the mean CRT contributions during the corresponding seasons (in Fig. 4d, only the seasonal-mean results in Sichuan Basin are shown). Seasons with the most severe $PM_{2.5}$ pollution in the region are marked in red. The references noted in the figure are as follows: (1) Chang et al., 2019; (2) Li et al., 2019d; (3) Wang et al., 2014a; (4) Zhao et al., 2012; (5) Li et al., 2015b; (6) Hou et al., 2019; (7) Wu et al., 2013; (8) Yin et al., 2017; (9) Lu et al., 2019c; (10) Lv et al., 2015b; (11) Pan, 2015; (12) Shen et al., 2019; (13) Qiao et al., 2019; (14) Chen et al., 2017.

3.3.2. *Temporal variations*

(1) **Diurnal and seasonal variations:** In general, within a day, the CRT contributions (in percentage) to O_3 reach the minimum at noon and the maximum at late night (Li et al., 2012a; Li et al., 2016; Gao et al., 2017; Huang et al., 2018a; Li et al., 2019c; Yang et al., 2019; Gao et al., 2020). During the day, O_3 is produced through photochemistry mostly from local emissions, thus leading to increasing (decreasing) local (CRT) contributions to O_3. Higher CRT contributions to O_3 at night can be explained by local O_3 being titrated by NO, depleted by unsaturated VOCs, deposited and transported outwards while transport still bringing non-local O_3 into the region. As displayed in Fig. 3, the CRT contributions to O_3 in different regions are overall lower in summer than in other seasons. This is linked to more active photochemistry in summer, which enhances O_3 production from local emissions.

In contrast, the CRT contributions to $PM_{2.5}$ do not show a consistent diurnal variation in different periods and regions, and thus are rarely discussed in relevant publications. The seasonal variation of the CRT contributions to $PM_{2.5}$ also differs in the main regions of China (Fig. 4). For the NCP, Chang et al. (2019)

concluded that the seasonal-mean CRT contributions to $PM_{2.5}$ maximize in autumn and minimize in winter, whereas Li et al. (2019d) reported that they are higher in spring and slightly lower in summer. In the YRD, PRD and SCB, the CRT contributions to $PM_{2.5}$ tend to be lower in summer and higher in the other seasons (Yin et al., 2017; Hou et al., 2019; Li et al., 2019d; Lu et al., 2019c).

(2) **Short-term changes:** Short-term changes here refer to the changes in the CRT contributions to O_3 and $PM_{2.5}$ on the time scale of several days. Normally, short-term change is driven by the transition of synoptic systems. As mentioned in Section 2.1, different synoptic systems feature with distinct wind speeds, transport pathways and/or other meteorological factors, and thereby have a distinct influence on the transport of pollutants. The variations of CRT contributions to O_3 and $PM_{2.5}$ under multiple synoptic weather patterns (SWPs) have been reported in several regions of China (Shu et al., 2020; Yan et al., 2021b; Shi et al., 2022; Hu et al., 2022a; Yan et al., 2023). For instance, the CRT contributions to O_3 are overall higher in the YRD under the SWPs characterized by northeasterly and northerly winds (Shu et al., 2020). Two SWPs named the Northeast Ridge and the Southeast High are associated with O_3 pollution in Xi'an (located in the GZB), but relatively higher CRT contributions to O_3 can be found under the former due to increased contribution from emissions in Henan Province (Yan et al., 2021b). The rapid change in pollutant emissions may also result in short-term change in the CRT contributions. One typical example is the change in the CRT contributions to pollutants during the COVID-19 lockdown in 2020 that was characterized by notably reduced anthropogenic emissions. During that period, $PM_{2.5}$ mass contributed by CRT decreased by 20–30 % in the YRD (Ma et al., 2021).

(3) **Long-term changes:** We focus here on the changes in the CRT contributions to O_3 and $PM_{2.5}$ over multiple years. For instance, Wang et al. (2015a) reported an increasing trend of the CRT contributions to $PM_{2.5}$ in Beijing over 2005–2010. In Hong Kong, a similar trend of contributions outside Hong Kong (including the CRT contributions) to coarse PM (or PM_{10}) was found over 2001–2013 (Luo et al., 2018). The changes in meteorological/climate conditions and pollutant emissions could have a marked impact on the long-term variations of transport contributions. Yim et al. (2019) found that transport contributions to $PM_{2.5}$ in Hong Kong are lower in the El Niño years and higher in the La Niña years. Dong et al. (2020) reported a decreased CRT contribution to $PM_{2.5}$ in the NCP during 2014–2017, which accounts for 28 % of the total $PM_{2.5}$ decrease. While the role of regional emission reduction is evident in lowering transport contributions to $PM_{2.5}$, the effect of meteorological changes is also considerable. Overall, long-term change in the CRT contributions to O_3 and PM ($PM_{2.5}$) is insufficiently explored in China. More systematic studies are needed to identify the historical, recent and future characteristics and influencing factors of long-term changes in the CRT contributions in different regions of China and to provide practical suggestions for effective air quality improvement.

4. Current knowledge on the detailed process of cross-regional transport

This section focuses on how O_3, PM and their precursors are transported from the source regions to the receptor regions. Various observational and modeling studies have illustrated the complex CRT processes of pollutants in China. Currently, the transport pathways of pollutants influencing air quality in the main regions of China have been well identified. Researchers have also gained valuable knowledge about the characteristics of detailed CRT processes in China: Some studies investigated one or two important steps of the process, including emission and initial transport, advective transport, as well as vertical exchange and mixing of pollutants (Fig. 2), while others provided a comprehensive illustration of the complete CRT processes of pollutants. In the following text, we summarize the methods used and the main findings on the above contents in relevant research.

4.1. Methods used in the studies of pollutant transport process

Observation of pollutant levels is essential to understanding the detailed CRT processes of O_3, PM and their precursors. Besides, meteorological parameters related to the CRT processes are often simultaneously measured. Since CRT occurs on multiple spatiotemporal scales and both horizontally and vertically, traditional in-situ observations at a single site can hardly meet the requirement to study CRT processes. Recent progress in building *the space and ground integrated three-dimensional observational system* (Fig. 5) in China has enabled explorations of dynamic and chemical processes of pollutants during CRT (Zhao et al., 2021a; Zhou et al., 2023a). Table 1 summarizes the spatiotemporal scales, advantages and disadvantages of various observational techniques in this system (Visconti et al., 2007; Liu et al., 2021a). There is no perfect method capable of illustrating the CRT processes of pollutants on all spatiotemporal scales. Based on instruments or sensors on unmanned aerial vehicles (UAV), balloons (e.g. ozonesonde), tethered airships and aircraft, the distributions of targeted pollutants can be measured at high spatiotemporal resolutions. This is the reason why these mobile measurement techniques are widely applied in field campaigns, although they are often costly and may be restricted by air traffic regulations. The disadvantages of these techniques limit their current applications in long-term routine measurement. Spectroscopic techniques, such as multi-axis differential optical absorption spectroscopy (MAX-DOAS), ground-based lidar and satellite-based remote sensing, can be used to track pollutant transport on large scales and during long periods. Along with the improvement in precision and applicability (Liu et al., 2019a; Liu et al., 2022a) and the usage of geostationary satellites (Kim et al., 2020), these techniques are becoming powerful tools to document and understand CRT processes of pollutants.

Backward trajectories can be used to track the movement of air parcels reaching the receptor regions. Some of the trajectory models (e.g., Hysplit (Draxler and Rolph, 2003)) are publicly available and have a simple interface, thus they are easy to access and operate. These advantages explain why trajectory-based methods have been widely used in the explorations of CRT processes (Sun et al., 2017). By clustering trajectories over long time spans, the general transport pathways influencing regional air pollution can be identified (Sirois and Bottenheim, 1995). By combining trajectories with the measurements of pollutant concentrations, functions including the Potential Source Contribution Function (PSCF; Ashbaugh et al., 1985) and the Concentration-Weighted Trajectory (CWT; Seibert et al., 1994) have been applied to determine the distribution of emission sources contributing to pollution in the targeted areas. Although trajectory-based methods are useful, they only provide an outline of CRT processes under large-scale atmospheric movements, and often fail to capture the details on the dynamic and chemical drivers of these processes. Additionally, the choice of receptor locations and starting times may influence the representativeness of the results.

Global/regional CTMs are currently irreplaceable to explore the complete CRT processes of pollutants and their multiple effects. They are applied to: (1) illustrate three-dimensional structures of pollutant transport; (2) explain in detail how pollutants are transported into the region (are pollutants transported through near-ground horizontal transport or vertical exchange through the ABL top? In the form of pollutants themselves or the precursors?) by using the Process Analysis or alike modules in CTMs and/or simulation of sensitivity scenarios (Chen et al., 2014; Qu et al., 2021b; Qu et al., 2023); (3) identify the CRT processes of pollutants originated from different regions and/or

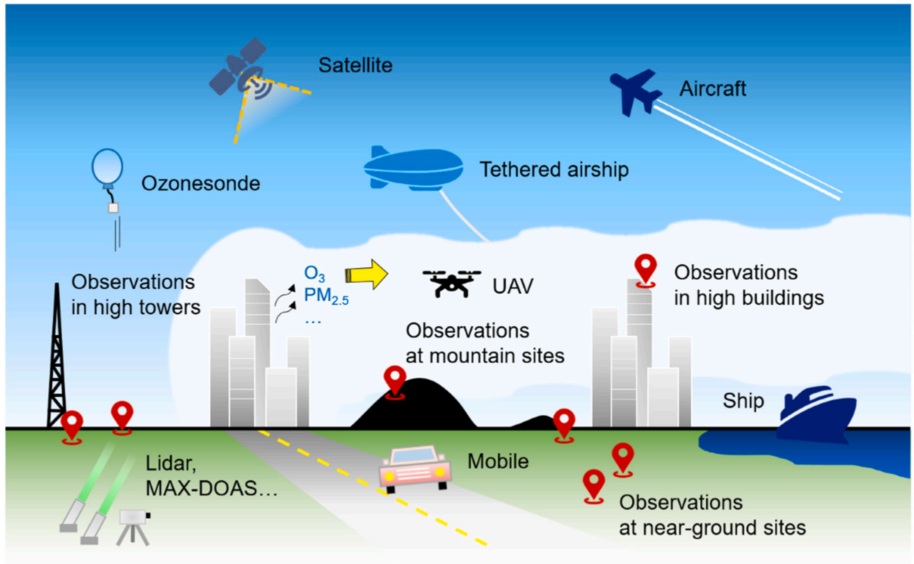

Fig. 5. Illustration of *the space and ground integrated three-dimensional observational system* in China to study the detailed CRT processes of pollutants. UAV, unmanned aerial vehicle; MAX-DOAS, multi-axis differential optical absorption spectroscopy.

emission sectors (Zhang et al., 2019a; Zhang et al., 2019b; Lin et al., 2021); (4) study the dynamic and chemical processes affecting pollutants during transport by combining with trajectory-based methods (Fairlie et al., 2009; Itahashi et al., 2017; Jeon et al., 2018); (5) explore the influence of transported pollutants on local meteorology and chemistry (Huang et al., 2020); and (6) quantify the detrimental effect of transported pollutants on human health, crops and ecosystems. However, CRT processes in CTMs are associated with uncertainties due to inaccurate descriptions of emission, meteorology, chemical and dynamic processes. An ongoing effort is needed to further improve emission inventories, meteorological fields, chemical mechanisms and other parameterizations that are applied in CTMs based on systematic evaluations of model performance with observational datasets.

It should be noted that the application of multiple observational methods or a combination of observational methods with models (including trajectory models, box models and global/regional CTMs) is promising for CRT-related studies. By taking advantage of the strength of each method, a comprehensive understanding of the CRT processes of pollutants can be obtained.

To quantitatively reveal the characteristics of pollutant transport, some studies estimated and analyzed the transport fluxes of pollutants through the interfaces between the targeted region and other regions. These fluxes are defined as the amounts of pollutants being transported across the interface during a unit time. For horizontal transport, the flux of the pollutant i (F_i) during a unit time dt is calculated as (Yang et al., 2012; Chang et al., 2018):

$$F_i = uc_iSdt \tag{4}$$

where u is wind speed perpendicular to the interface; c_i is the concentration of the pollutant i in the transported air parcels; S indicates the area of the interface. F_i can be calculated based on observational and/or modeling results, and its high values indicate the main CRT process contributing to regional pollution under specific settings of meteorology and emissions. To illustrate CRT processes of pollutants originating from different regions or/and emission sectors, some studies used source contributions replacing c_i in Eq. (4) to quantify their fluxes (Zhang et al., 2019a; Zhang et al., 2019b; Lin et al., 2021; Sulaymon et al., 2021; Zhang et al., 2022b). In contrast, vertical exchange fluxes through the ABL top were seldom reported in China (Jin et al., 2021; Jin et al., 2022; Qu et al., 2023). Systematic investigations are required to understand the general characteristics and variations of vertical exchange fluxes of pollutants, their contributions to the pollutant budgets, as well as their role in long-distance or intercontinental pollutant transport.

4.2. *Atmospheric transport pathways influencing air quality in the main regions of China*

Based on previous publications, we summarize the atmospheric transport pathways of pollutants influencing air quality in eight main regions of China (the NCP, the YRD, the PRD, the SCB, the GZB, the Middle Areas of the Yangtze River (Middle-YR), Northeast China (NE-China) and Northwest China (NW-China)), as displayed in Fig. 6. These transport pathways were mostly identified by using backward trajectories. According to the main sources of pollutants in the transported air parcels, they can be classified as these of dust, anthropogenic pollutants, biomass-burning plumes and marine parcels. These transport pathways and their potential influences on O_3 and PM pollution in the eight regions are briefly introduced below.

(1) **North China Plain (NCP):** Three transport pathways, including the northwest, southwest and southeast pathways, are potentially associated with O_3 and PM pollution in the NCP (Zhu et al., 2011; Lv et al., 2015a; Liang et al., 2016; Ren et al., 2016; Wang et al., 2016; Zhou et al., 2016; Wang et al., 2018a; Hao et al., 2019). Through the northwest pathway, dust originating from deserts and/or semi-deserts in Northwest China and the Mongolian areas can be transported into the NCP in spring (Wang et al., 2004; Zhang et al., 2013). In winter, transport through this pathway likely leads to the dissipation of pollutants (Pu et al., 2015). Transport through the southwest pathway is often linked to the development of severe haze in the NCP (Zheng et al., 2015; Wu et al., 2017b). Under its influence, PM and its precursors accumulate at the foot of mountains, where secondary PM is rapidly produced due to increasing humidity, weakened turbulence, reduced ABL heights as well as higher levels of precursors (Wang et al., 2010; Wang et al., 2015b; Wu et al., 2017b). Through the southeast pathway, anthropogenic pollutants derived from regions to the south of the NCP (including Shandong, Henan and the YRD) are transported northwards, contributing to summertime O_3 pollution and wintertime $PM_{2.5}$ pollution in the NCP (Zhang et al., 2015b; Chen et al., 2017; Li et al., 2017d).

Table 1
Comparisons between observational techniques used to investigate the detailed CRT processes of pollutants. DOAS, differential optical absorption spectroscopy; MAX-DOAS, multi-axis differential optical absorption spectroscopy; UAV, unmanned aerial vehicle. "iD" (i = 0–3) in the columns of spatial dimension means that the method can provide an i-dimensional dataset for further analyses. ABL, atmospheric boundary layer; FT, free troposphere. For the duration, "short" indicates that the method is normally used in field campaigns that last for several days or months, and "long" indicates that the method can be used for routine measurements that last for years.

Observational method		Spatial dimension	Height range	Duration	Advantages	Disadvantages
Site-based observations	Near-ground site	0D	Near-ground	Short / Long	Controllable observational conditions; capability to measure many species	Spatial representativeness may be lacking; disturbance from nearby emissions
	Tower / High building site	0D / 1D	Low ABL	Short / Long		
	Mountain site	0D	ABL / Low FT	Short / Long		
	Ground-based spectroscopic methods (DOAS, MAX-DOAS, Lidar, etc.)	1D / 2D	Troposphere	Short / Long	Good coverage and resolution in the vertical direction; high temporal and spatial continuity	Capability to measure only limited species; the existence of blind spot (lidar); uncertainties caused by retrieval algorithms
Mobile observations	Car / Bike / Ship	1D / 2D / 3D	Near-ground / Troposphere	Short	High flexibility; reasonable spatial coverage	Limited temporal coverage; disturbance from the nearby emissions
	UAV	3D	ABL	Short	High flexibility; lower costs than other mobile observations	Capability to measure only limited species; short duration; sensors need calibration; flight restrictions in some regions; inability to measure in bad weather conditions (e.g., storms)
	Tethered airship	1D	ABL	Short	High flexibility; capability to measure many species	High costs; inability to measure in bad weather conditions
	Balloon (e.g., ozonesonde)	1D	Troposphere / Low stratosphere	Short	Good coverage and resolution in the vertical direction	Capability to measure only limited species; high cost
	Aircraft (including civil aviation)	1D	Troposphere / Low stratosphere	Short / Long	Good coverage and resolution in vertical directions; high temporal and spatial continuity; suitability for both online and offline measurements	High cost; limited temporal coverage; flight restrictions in some regions
	Satellite-based remote sensing	2D / 3D	Troposphere / Stratosphere	Long	High spatial coverage	Low spatiotemporal resolution; uncertainties caused by sensors, retrieval algorithms, etc.

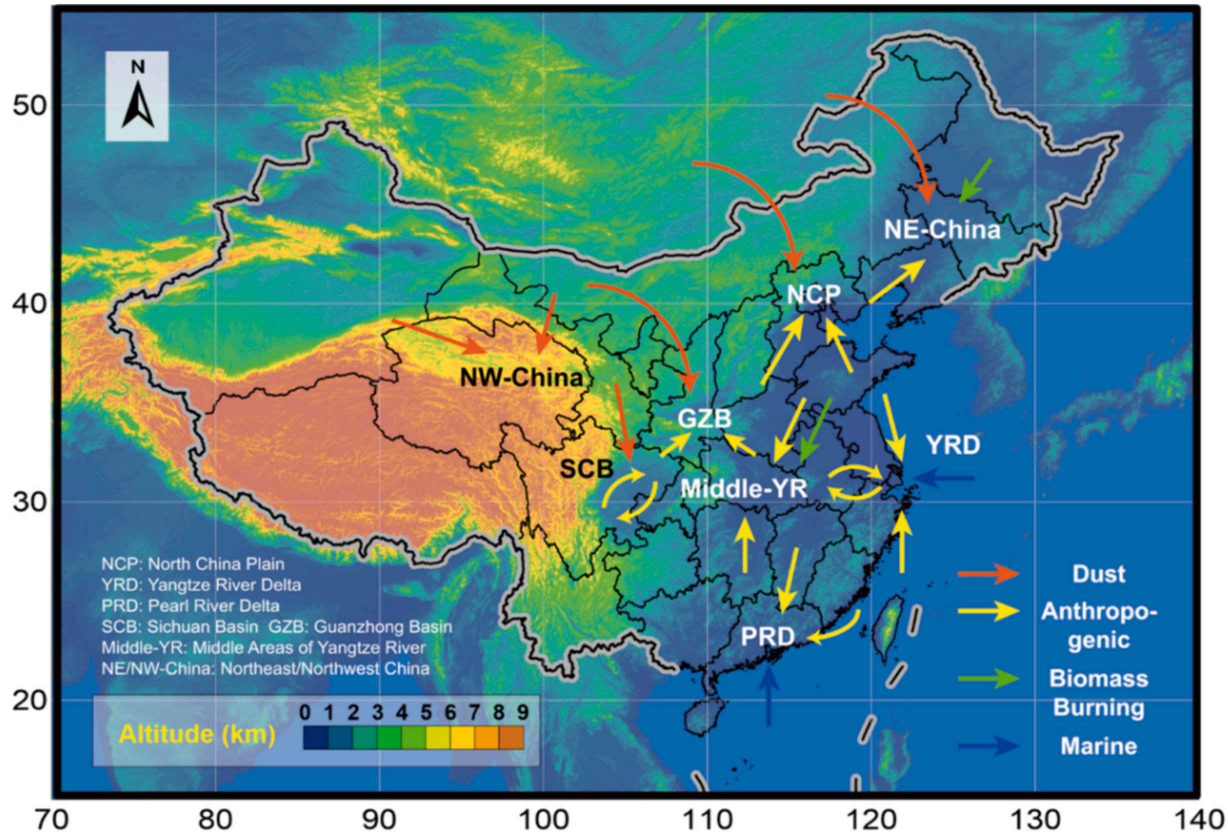

Fig. 6. Illustration of the atmospheric transport pathways of pollutants influencing air quality in the eight main regions of China. Transport pathways are classified based on the main sources of pollutants in the transported air parcels.

(2) **Yangtze River Delta (YRD):** The seasonal variation of East Asian monsoon explains the difference between the main transport pathways for the YRD in the four seasons (Li et al., 2012b; Kong et al., 2013; Li et al., 2015c; Lv et al., 2015a; Wang et al., 2015b; Zhao et al., 2015b; Zhou et al., 2016; Ding et al., 2017; Zhang et al., 2017b; Huang et al., 2018b; Yao et al., 2019). When winter monsoon that is characterized by frequent northerly winds prevails, polluted air parcels from regions including the NCP and Shandong are likely to be transported southwards to the YRD, triggering severe PM pollution. In contrast, summer monsoon may bring pollutants derived from anthropogenic emissions in Southeast China and/or shipping emissions near the coast northwards to the YRD. During the transition periods between the two monsoons, namely, in spring and autumn, the east pathway may also influence the region. Through short-distance transport in the west-east direction, pollutants emitted or produced in the west YRD and Middle-YR may influence air quality in the coastal area of the YRD, and pollutants contributed by emissions of the east YRD are also possibly transported westwards (Kong et al., 2013; Shi et al., 2018; Yao et al., 2019).

(3) **Pearl River Delta (PRD):** Three main transport pathways of pollutants were identified for the PRD: the continental, coastal and marine pathways (Lee et al., 2007; Zheng et al., 2010; Cheng et al., 2013; Nie et al., 2013; Lv et al., 2015a; Zhou et al., 2016; Li et al., 2018a). Through the continental pathway, air masses travel above land areas to the north of the PRD and cross over the Nanling Mountains to reach the PRD; whereas through the coastal pathway, air masses travel southwards to the PRD nearly following the southeast coastline of China. The division of air masses through these two pathways indicates the hindrance of the Wuyi Mountain (located between two pathways) to pollutant CRT (Li et al., 2018a). The continental and coastal pathways influence the PRD mostly in autumn and winter, potentially contributing to both O_3 and PM pollution (Lee et al., 2007; Zheng et al., 2010; Li et al., 2023b). Most appearing in spring and summer, the marine pathway is linked to the northwards transport of air parcels from the South China Sea. It is normally associated with relatively good air quality in the PRD. However, the increasing trends of O_3 and PM levels under its influence suggest the growing contributions from pollutant emissions from Southeast Asia (Wang et al., 2019b; Zeng et al., 2023), which should be further concerned.

(4) **Sichuan Basin (SCB):** The basin topography determines that pollutants in the SCB are often transported slowly within the region along short paths during pollution (Lv et al., 2015a; Zhou et al., 2016; Liao et al., 2017; Chen et al., 2018b; Wang et al., 2018b; Peng et al., 2019). Dust, which is transported from Northwest China southwards along the edges of the Tibetan Plateau, may also contribute to PM pollution in the SCB (Chen et al., 2018b). Good air quality is expected here when northeast winds with relatively high speeds (~1.5 m/s) prevail (Du et al., 2020b).

(5) **Guanzhong Basin (GZB):** Similar as the SCB, O_3 and PM pollution in the GZB are mainly influenced by short-distance transport (Li et al., 2006; Wang et al., 2012a; Wang et al., 2020d; Song et al., 2021). It is often hard to identify the primary transport directions related to O_3 and PM pollution in the GZB, because westward, northward and eastward transport could all considerably contribute to pollution. In addition, transport from the northwest may bring dust into the GZB, triggering or aggravating PM pollution here (Wang et al., 2014b).

(6) **Middle Areas of the Yangtze River (Middle-YR):** The Middle-YR, including three provinces (Hubei, Hunan and Jiangxi), is located in Central China. Due to the seasonal variations of

prevailing winds, the main transport pathways in the Middle-YR differ between seasons (Zhang et al., 2014; Xiong et al., 2017; Zhao et al., 2017; Yuan et al., 2018; Huang et al., 2019b; Huang et al., 2019c; Zhang et al., 2020a). The northwest pathway mainly influences the region in spring, and it may be linked to long-range dust transport. Under summer monsoon, transport through the south pathway affects the region more frequently, but it is less likely to trigger severe pollution. In autumn and winter, through the northeast and east pathways, pollutants contributed by anthropogenic and/or crop residue burning emissions in North China may be transported to the region, resulting in the occurrence of PM pollution here.

(7) **Northeast China (NE-China):** Dust transport may result in severe PM pollution in NE-China in spring as well (Ma et al., 2017). In autumn and winter, pollutants contributed by coal and/or crop residue burning emissions are possibly transported within the region or from other neighboring regions, contributing to extremely severe $PM_{2.5}$ pollution (Ma et al., 2017; Zhao et al., 2019a). Moreover, when the southwest pathway dominates, pollutants from the NCP and the Bohai Rim region are likely transported to NE-China (Meng et al., 2020; Zhao et al., 2020; Liu et al., 2021b).

(8) **Northwest China (NW-China):** Since NW-China is close to dust sources (including the Gobi Desert, the Taklamakan Desert, etc.), dust transport has a notable impact on air quality in this region, and its pathways correspond well with the relative spatial distribution of dust sources (Xin et al., 2016; Guan et al., 2019). In summer, pollutants may be transported to the region from the east, but they normally do not lead to severe pollution in NW-China (Xin et al., 2016; Guan et al., 2019).

4.3. Main findings on the detailed cross-regional transport processes in China

Generally, the detailed CRT processes from the source region to the receptor region can be decomposed into three steps (Fig. 2): (1) emission and initial transport; (2) advective transport at various heights, which mostly occurs horizontally; (3) vertical exchange through the ABL top (entrainment and detrainment) and vertical mixing within the ABL. Some studies focused on one or two of these steps based on observational and/or modeling results and revealed their influence on regional pollution. Moreover, researchers also tried to illustrate in detail the complete process of CRT and to quantify the effects of pollutant transport on local meteorology and chemistry of secondary pollutants. In this section, the main findings regarding the above detailed processes of CRT in China are reviewed.

4.3.1. Steps of the CRT processes

4.3.1.1. Emission and initial transport. The characteristics of emission and initial transport from emission sources are fundamental for evaluating their potential influence on regional air quality. Besides developing bottom-up emission inventory, observational methods, especially car-based mobile observations, are also applied to quantify the emissions of specific sources. In brief, assuming the conservation of pollutant masses within a small region, emissions can be estimated by using the net transport fluxes measured around the targeted source (Ibrahim et al., 2010). Based on mobile MAX-DOAS measurements and WRF modeling, Tan et al. (2020) quantified the emissions of SO_2 and NO_2 from two power plants in North China. By using similar methods, Cheng et al. (2020a) and Tan et al. (2019) estimated NO_x emissions in Beijing and Hefei, respectively, and compared these results with corresponding city-level emissions in the Multi-resolution Emission Inventory for China (MEIC).

4.3.1.2. Advective transport at various heights. Near-ground advective transport of plumes with high-level O_3 and PM leads to the rapid growth of pollutant concentrations in the receptor sites. Since transport takes time, such an increase often shows temporal lags between different cities/sites (Lv et al., 2017a; Chen et al., 2019; Gu et al., 2020; Huang et al., 2020). This temporal-lagging phenomenon can be used as an indicator of the influence of CRT on pollution. Through complex network analysis, the general patterns of pollutant transport can be identified based on measurements at multiple sites, where more detailed routes of CRT processes are revealed (Wang et al., 2017c; Wang et al., 2021; Ying et al., 2022; Wang et al., 2022c).

Apparently, advective transport of pollutants is not only confined to the near-ground height. Observations of the vertical structures of pollutants can reveal the effects of CRT at different heights. In the lower troposphere, O_3 levels may increase, decrease, or remain stable with height (Ding et al., 2008; Wang et al., 2012b; Ma et al., 2013; Li et al., 2017e; Su et al., 2017; Wang et al., 2017d; Chi et al., 2018; Zhang et al., 2019c; Zhao et al., 2019b; Li et al., 2020; Yan et al., 2021c; Li et al., 2022; Liu et al., 2022b), whereas PM concentrations (indicated by the scattering coefficient in some studies) generally reduce with height, especially during severe haze periods (Peng et al., 2015; Liu et al., 2017; Li et al., 2018b; Wang et al., 2018c; Fan et al., 2019; Li et al., 2019e; Lu et al., 2019d; Xu et al., 2019). However, observations in China also reported the existence of one or more elevated pollution layers at various altitudes ranging from ~500 m to ~5 km, characterized by notably higher levels of O_3 or PM compared to nearby altitudes (Guo et al., 2016; Li and Han, 2016; Heese et al., 2017; Han et al., 2018c; Liu et al., 2018; Lyu et al., 2018; Tian et al., 2018; Lu et al., 2019e; Cheng et al., 2020b; Liu et al., 2020b; Lv et al., 2020; Popovici et al., 2020; Pu et al., 2020; Sun et al., 2020; Xiang et al., 2020; Zhang et al., 2020b; Zhang et al., 2021b; Zhao et al., 2021b). CRT is considered as one of the processes leading to the formation of these layers. For PM, its chemical composition varies with altitude, which may indicate different influences of transport on various PM components. Higher proportions of secondary components (especially sulfate) and higher OC/EC ratios in $PM_{2.5}$ were found in the middle and/or high ABL than those near the surface, suggesting that secondary PM is more easily transported or is formed during transport (Chan et al., 2005; Sun et al., 2015; Hua et al., 2016; Sun et al., 2016; Zhou et al., 2018b; Liu et al., 2019b; Tian et al., 2019; Zhou et al., 2020; Sun et al., 2022b). However, as an important type of primary PM, dust was observed to be transported at altitudes higher than 2 km in the NCP, YRD and GZB (Han et al., 2015b; Lv et al., 2016; Sun et al., 2018a; Zhou et al., 2018c; Zhou et al., 2018d; Sheng et al., 2019; Wang et al., 2020e; He et al., 2021a; Tao et al., 2021). Dust may be directly transported downwards and contribute to poor air quality in regions along the transport pathway (Lv et al., 2020; Wang et al., 2020f), but it can also aggravate near-ground PM pollution indirectly by weakening solar radiation and enhancing local stagnation (Zhou et al., 2018c; Wang et al., 2020e; Liang et al., 2022).

Advective horizontal transport of pollutants at various heights can be quantitatively characterized by their fluxes (Eq. (4)). Some studies investigated the vertical structures of $PM_{2.5}$ fluxes in different regions of China based on mobile observations and/or model simulations. Since in most cases, $PM_{2.5}$ concentration is reduced with height while wind speed increases with height, peak $PM_{2.5}$ transport fluxes were often found in the middle/high ABL or low FT, at heights ranging from ~0.5 km to ~1.5 km (Chang et al., 2018; Zhang et al., 2019a; Zhang et al., 2019b; Lin et al., 2021; Liu et al., 2019c; Xiang et al., 2020; Ji et al., 2021; Sulaymon et al., 2021; Yu et al., 2021; Zhang et al., 2022b; Song et al., 2023). Thus, transport in these layers is more efficient than near-ground transport for $PM_{2.5}$. High values of $PM_{2.5}$ fluxes through the interfaces surrounding the region are used to indicate the main directions and heights of $PM_{2.5}$ transport. For instance, based on $PM_{2.5}$ transport fluxes calculated using mobile observations, Lv et al. (2017b) and Xiang et al. (2020) verified the major impact of CRT through the southwest pathway and at altitudes between ~0.5 km and ~1.5 km for $PM_{2.5}$ pollution in

Beijing. For more detailed routes of CRT, the results of regional source apportionment can be used in the flux calculations: high fluxes contributed by specific regions or cities indicate that they tend to be close to the transport route (Zhang et al., 2019a; Zhang et al., 2022b).

If the CRT process is restrained, pollutants accumulate locally, thereby resulting in persistent pollution over specific areas. The mechanisms behind the restraining effect on pollutant transport were explored in some regions of China, and found to be related to the influence of topography and/or synoptic systems. When the prevailing synoptic system for the region changes into a more stable type, e.g., high pressure, the CRT process slows down or even stops (Wang et al., 2022d). In the NCP and SCB, the blocking of air flow by high mountains or plateau terrain not only restrains pollutant transport, but also aggravates pollution through the formation of vertical vortexes, which leads to downdrafts and shallower ABL (Zhang et al., 2018b; Zhang et al., 2019d; Shu et al., 2021; Hu et al., 2022b). Besides the effect of terrain topography, confronting warm air was reported also to block the southward transport of $PM_{2.5}$ in the Middle-YR (Hu et al., 2021; Bai et al., 2022).

Since in the lower troposphere, vertical wind speed is normally lower than horizontal wind speed by over an order of magnitude, vertical transport through advection is less important for regional pollution in most cases. However, under some weather systems such as WCB and deep convection, near-ground pollutants may be rapidly brought to the high-latitude atmosphere through advection. For instance, Ding et al. (2009) reported high-level O_3, SO_2 and CO at the height of ~2 km in an aircraft-based measurement in NE-China, suggesting the impact of WCB. For O_3, SI is an important vertical process in CRT. Under its influence, O_3 levels increase in the middle/upper troposphere, accompanied by high potential vorticity, low humidity and low CO levels (Chen et al., 2021; Zhang et al., 2021b; Zhao et al., 2021b; Ma et al., 2022; Zhan and Xie, 2022; Chen et al., 2024). Recent studies also reported near-ground O_3 pollution episodes in both North and South China with considerable contributions from the stratosphere (Ni et al., 2019; Wang et al., 2020g; Chen et al., 2021; Lu et al., 2021; Zhao et al., 2021b; Zhao et al., 2021c; Chen et al., 2022) which range from 5 % to 32 % in percentage (Wang et al., 2020h; Meng et al., 2022a; Meng et al., 2022b). The dynamic drivers of O_3 SI during these episodes include typhoon periphery, cut-off low, mid-latitude cyclones and high-level jets (Li et al., 2014; Lu et al., 2021; Zhao et al., 2021b; Chen et al., 2022; Meng et al., 2022b; Yang et al., 2022).

4.3.1.3. Vertical exchange and mixing. Through vertical exchange, pollutants in the FT and/or residual layer are entrained into the ABL and involved in the ABL mixing, thereby contributing to pollution near the ground. Since pollutant transport at the heights of about 0.5–1.5 km is overall more efficient, pollutants contributed by non-local sources are more likely to be transported in the lower FT or middle/high ABL. As the process to bring these pollutants downwards to the surface, vertical exchange and mixing often act as essential steps in the CRT process. For near-ground O_3, vertical exchange has been reported to drive a rapid increase in O_3 concentration within a few hours after sunrise (Hu et al., 2018; Xu et al., 2018; Yang et al., 2021a; Hu et al., 2022b; Qu et al., 2023). By using the estimation methods based on O_3 profile observations or one-dimensional ABL models, the contributions of this process to daytime O_3 were quantified within the range of 20–65 % in the NCP and PRD (Qi et al., 2018; Zhao et al., 2019b; Zhu et al., 2020a; He et al., 2021b; Liu et al., 2022b). Similarly, $PM_{2.5}$ can also be transported into the targeted regions through vertical exchange, leading to the occurrence of regional haze or aggravating the pollution (Sun et al., 2013; Qin et al., 2016; Han et al., 2018d; Quan et al., 2020; Yang et al., 2021b; Zhou et al., 2023b; Jia et al., 2024). However, quantified contributions of vertical exchange to $PM_{2.5}$ were less reported compared with those to O_3 in China (Jin et al., 2021; Jin et al., 2022). In addition, precursors transported downwards into the ABL may also considerably contribute to the local chemical production of pollutants. For instance, SO_2 transported from the residual layer was found to drive new particle formation in Beijing despite high condensation sink during severe haze (Wang et al., 2023).

Although the ABL is normally stable at night, O_3–rich air in the residual layer can still be exchanged into the ABL, leading to rapid increase in nighttime O_3 levels, or "nocturnal O_3 enhancement" (He et al., 2023; An et al., 2024). The atmospheric dynamic processes related to this phenomenon include low-level jets, convective storms, local circulation, front, foehn winds, etc. (Zhu et al., 2020b; He et al., 2022; An et al., 2024; Zhu et al., 2024b). For more detailed information, readers are suggested to refer to the recent review paper by An et al. (2024).

O_3 pollution is under the notable influence of ABL mixing during the daytime. This process brings O_3 precursors emitted near the surface to the middle/high ABL, where O_3 is found to be more rapidly produced (Han et al., 2020). The newly produced O_3 is then transported downwards to the ground. This has been reported as the main process related to O_3 pollution in the NCP (Tang et al., 2017) and might also be applicable to other urban regions in China. The sensitivity of O_3 production to precursors (NO_x and VOCs) varies with height, following changes in VOC/NO_x ratio. As concluded by Tang et al. (2017) and Chi et al. (2018), O_3 production in the lower ABL of urban regions tends to be VOC-limited, while at higher altitudes, it is more likely to be NO_x-limited. The intensity of ABL mixing is also related to the severity of O_3 pollution. In the NCP, more severe O_3 pollution tends to occur in the strongly convective ABL with the daytime maximum ABL height of about 1–2 km, rather than in the forced or weakly convective ABL with higher or lower heights (Tang et al., 2021; Zhang et al., 2023). PM originating from near-ground emissions can be transported upwards through ABL mixing as well. However, differences exist among PM components: The transport of PM components with lower volatility (e.g., BC, particulate sulfate and low-volatile organics) is more efficient than that of semi-volatility PM components (e.g., particulate nitrate and semi-volatile organics) due to their weak evaporation into the gas phase (Liu et al., 2021c).

4.3.2. Complete CRT processes

By exploring the details in the complete CRT processes of pollutants, we can better understand how CRT influences the occurrence and development of regional O_3 and PM pollution. In relevant studies, the contents to be explored may include:

- The detailed routes of CRT from the source regions to the receptor regions
- The meteorological factors driving CRT at its different stages
- The main chemical reactions linked to pollutant production during CRT
- The effects of CRT on local meteorology, chemistry, and feedbacks on O_3 and PM pollution

Most studies in China tried to explore the complete CRT processes of pollutants during one or several pollution episodes. Xu et al. (2018) studied the CRT processes linked to O_3 pollution in the YRD: During two episodes, O_3 separately contributed by crop residue burning in North China and anthropogenic emissions in the surrounding cities was transported in layers above the ABL and then entrained into the region, triggering pollution. Zheng et al. (2021) simulated an O_3 pollution episode in the southeast coastal regions of China, and results show that O_3 precursors from the NCP, NE-China, Japan and Korea were firstly transported to the East China Sea, where O_3 was rapidly produced; afterwards, air masses with high-level O_3 were transported southwards to the studied region. Miao et al. (2017) revealed the CRT processes of $PM_{2.5}$ during an episode in Beijing: $PM_{2.5}$ contributed by provinces to its south was initially transported northwards to the Bohai Sea under the high-pressure system, and then sea breezes brought these pollutants westwards to Beijing. In another haze episode in the NCP, Sun et al. (2018b) found that $PM_{2.5}$ was transported uphill under east winds, leading to the formation of multi-layer temperature inversions; afterwards, along with the effect of south winds and vortex before the mountain, the ABL maintained stable, and $PM_{2.5}$ pollution further aggravated. According to Zhou et al. (2018c), CRT played an important role in a persistent $PM_{2.5}$ pollution episode in the YRD: Dust was transported from NW-China by cold fronts, and also, biomass-burning plumes were transported from Southeast Asia by westerlies. These pollutants not only were well mixed with local-emitted pollutants, resulting in severe regional pollution, but further promoted their accumulation through the interactions with local meteorology. Wang et al. (2017f) investigated two rapid, long-distance CRT processes linked to $PM_{2.5}$ pollution in Xiamen, South China: Under the influence of cold surges, wind speeds in East China exceeded 20 m/s and $PM_{2.5}$ was transported from the NCP to Xiamen (in a distance of ~2000 km) within only two days. The above reported results demonstrate the complexity of CRT processes in China.

To develop effective pollution control strategies, studies on the complete CRT processes of pollutants during only a limited number of pollution episodes may not be sufficient. Instead, it is required to systematically summarize the common characteristics of CRT processes during frequently occurring and/or the most severe episodes in the regions of interest. A typical example of such studies is by Huang et al. (2020), which introduced the mechanism of $PM_{2.5}$ CRT influencing the YRD by summarizing the characteristics of $PM_{2.5}$ CRT processes during 17 episodes: $PM_{2.5}$ contributed by emissions in the YRD was transported northwards when weak anticyclone prevails, and then accumulated in the NCP, where high humidity and stable ABL further enhanced the production of secondary $PM_{2.5}$. Afterwards, cold fronts brought these pollutants southwards back to the YRD. Based on this mechanism, earlier emission reductions and regional cooperation are essential to alleviate haze pollution in both the YRD and the NCP. From this example, we can learn that: (1) The interaction between CRT, local meteorology and chemistry may play an important role in the process of air pollution. As summarized in Section 2.4, various forms of interaction between CRT and local chemistry have been found, but further evaluation on their contribution to regional air pollution is still needed. (2) Such studies on the complete CRT processes of pollutants are practical to support air pollution control. However, relevant reports are limited.

With the ability to provide results with satisfying spatiotemporal resolutions and coverages, global/regional CTMs were applied in all of the aforementioned studies on the complete CRT processes of pollutants. The results of three-dimensional observations and other models (e.g., the trajectory model) often act as important supplements leading to solid conclusions. The above choices of methods are also applicable to relevant studies in the near future.

5. Conclusions and prospects

O_3 and PM pollution is not only contributed by local emissions and chemical transformations, but is often notably influenced by cross-regional transport (CRT). The detailed processes of pollutant CRT are complex due to the multi-scale nature of dynamic processes and the intricate chemical transformations affecting pollutants during/after transport. China, which is still impacted by occasional severe O_3 and PM pollution, aims to further improve its air quality through regional cooperation in emission reduction. Therefore, it is essential to systematically understand the effects of CRT on O_3 and PM pollution in different regions of China. Based on numerous publications that were mostly reported after 2017, this paper has reviewed the methodologies adopted for relevant studies and the current knowledge on this topic. The key outcomes are summarized as follows:

(1) **Contribution of CRT to regional O_3 and PM pollution**:
- The CRT contribution (in proportion) can be quantified by using observation-based methods or CTMs-based source apportionment methods. The former enables an easy estimation of the CRT contribution, while the latter quantifies the CRT contribution from well-defined sources.
- According to recent studies in the main regions of China including the NCP, YRD and PRD, the CRT contribution to O_3 and $PM_{2.5}$ spans a wide range of values (40–80 % for O_3 and 10–70 % for $PM_{2.5}$). However, the atmospheric concentrations of both pollutants were often mostly contributed by CRT (sources outside the studied region) rather than by local emissions, highlighting the importance of CRT for regional air pollution. Among the $PM_{2.5}$ components, sulfate and nitrate often feature with relatively higher CRT contributions.
- The contribution of CRT to pollutants varies between regions, and generally increases with altitude. The CRT contribution to O_3 tends to be lower at noon and in summer, indicating the effect of enhanced O_3 production from local emissions. In contrast, the diurnal and seasonal variations of the CRT contribution to $PM_{2.5}$ are not consistent among different periods and regions. Short- and long-term changes in the CRT contributions have been increasingly explored, and they are mainly attributed to changes in meteorological (climatic) conditions and/or emissions.

(2) **Detailed CRT processes of O_3, PM and their precursors**:
- Trajectory-based methods have been widely used to identify transport pathways and potential source regions related to O_3 and PM pollution. Based on backward trajectories, the main transport pathways influencing air pollution in the main regions of China have been identified, as shown in Fig. 6.
- The three-dimensional observational system integrating multiple techniques (Fig. 5) is under rapid development in China, and it has started to play an essential role in the investigation of complex CRT processes. For example, mobile observations using spectroscopic techniques (e.g. MAX-DOAS) have been applied to quantify the emissions from specific sources and to illustrate the main transport pathways and heights of pollutants in the NCP. However, global/regional CTMs are currently irreplaceable in illustrating the complete CRT processes of pollutants as well as their impact on local meteorology and chemistry.
- When CRT occurs near the surface, its influence can be seen from the sudden increase in pollutant levels in the site-based measurements. Normally, however, CRT occurs at different heights, and may lead to the formation of elevated polluted layers at the altitudes of 0.5–5 km as indicated by the observations. According to the reported results of transport fluxes, horizontal transport of pollutants is generally more efficient in the middle/high ABL or low FT, at the altitudes of 0.5–1.5 km, than near the ground.
- Specific settings of topography and/or weather systems can restrict atmospheric transport, resulting in the accumulation of pollutants and thereby severe air pollution in some areas.
- Some larger-scale processes of pollutant transport in China have also been reported, including the upward transport of anthropogenic pollutants through the WCB and the downward intrusion of stratospheric O_3.
- Vertical exchange and mixing are likely to be an important step in the CRT of pollutants, and many studies have found their notable contribution to near-ground O_3 and $PM_{2.5}$ pollution. Vertical mixing, especially in the convective ABL, enhances O_3 production and thus exacerbates O_3 pollution.
- The complete CRT processes during one or more pollution episodes have been investigated in some studies, where the detailed transport routes, meteorological driving factors, chemical reactions during transport and the interactions of transport with local meteorology and chemistry were revealed. However, there was limited research to summarize the general

characteristics of typical CRT processes of pollutants based on the systematic analyses of multiple episodes.

For future studies on the effects of CRT on O_3 and PM pollution in China, we suggest the following aspects to be strengthened:

(1) **More comparisons and collaborations between observation and modeling are required.** An increasing number of studies have attempted to use both observations and model results to investigate the CRT of O_3, PM and their precursors in China. This allows researchers to take advantage of the accuracy of observations and the high spatiotemporal coverage of model results. However, as a prerequisite for consolidating the findings, the comparison between observations and model results needs to be strengthened. In addition to commonly measured meteorological parameters and pollutant concentrations, we recommend including in the comparisons more variables related to atmospheric dynamics (e.g., ABL height), chemistry (e.g., mixing ratios of atmospheric oxidants and intermediates) and pollutant budgets (e.g., horizontal/vertical transport and deposition fluxes of O_3 and PM). A satisfactory performance of the model results ensures both accurate quantification of the CRT contribution and realistic representation of the CRT processes. To provide sufficient data for model validation, it is necessary to conduct short-term campaigns targeting a larger number of atmospheric pollutants and dynamic parameters, both near and distant from the emission sources, as well as long-term measurements by the three-dimensional observing system. On the other hand, the model results can provide useful information for the design of the observations, including the duration, the location for site-based observations and the route for mobile observations.

(2) **Short- and long-term variations of the CRT contributions to O_3 and $PM_{2.5}$ in different regions of China need more systematic investigations.** Most publications have focused on the CRT contributions to O_3 and $PM_{2.5}$ in the NCP, YRD and PRD. Thus, additional studies are needed for other regions. Even for these three major regions of China, there are still open questions about how the changes in anthropogenic and natural emissions, meteorological and climatic factors lead to the short- and long-term variations of the CRT contributions. Systematic research on this topic will enable us to better understand how the above factors influence regional air quality, and will provide support for more successful pollution control. It should also be noted that more accurate emission inventories with high spatial and temporal resolution in China are essential for related CTM-based studies. Thus, continuous efforts are required for their development. In addition, studies on the potential change in the CRT contributions in future scenarios are also warranted for long-term air quality improvement.

(3) **A better understanding of the detailed chemical processes in the air parcels during transport is needed.** As shown in Fig. 2, various chemical processes of pollutants take place in the transported air parcels. The duration of CRT is usually long enough to ensure the occurrence of fast and slow chemistry, so that both can contribute significantly to downwind pollution. However, it is still not fully understood how different pollutants are produced and transformed at different stages of transport and how precursors, oxidants and meteorological conditions influence these processes. Specifically, there is a need to better understand the transformations of O_x, reactive nitrogen (NO_y; mainly including NO_x, HONO, NO_3, N_2O_5, HNO_3, particulate nitrate and organic nitrates), VOCs and heterogeneous reactions on the aerosol surfaces during transport, and their potential influence on regional O_3 and PM pollution. Results on this topic have been reported in other countries and/or regions. For example, detailed chemical processes in the wildfire plumes in the U.S. have been reported by analyzing aircraft-based observations (Decker et al., 2021; Xu et al., 2021; Peng et al., 2022). As anthropogenic and biogenic emissions are widely distributed in East China, newly emitted pollutants may be added to the air parcels at any stage of transport, potentially making the chemical processes of pollutants during transport more active and complex. Although relative studies are scarce, with advances in observational and modeling techniques, more in-depth investigations of the chemistry of pollutants during transport in China are expected in the near future.

(4) **Studies are proposed to further understand how vertical exchange and mixing affect regional pollution.** Vertical exchange and mixing are likely to become an important step in the CRT. However, reports on their contributions to ground-level O_3 and PM pollution in China have been limited overall. Studies should be conducted to quantify these contributions using ABL models or global/regional CTMs, and to identify their spatiotemporal variations and influencing factors in different regions of China. Given the dominant role of vertical exchange in the hourly mass changes of pollutants in the ABL, it is also worth exploring the influences of these vertical processes on the characteristics of regional O_3 and PM pollution, including pollutant sources, sensitivities, and reaction pathways (Qu et al., 2023). Moreover, ABL parameterizations nowadays often show remarkable differences in describing the stable ABL, especially during severe haze periods (Jia and Zhang, 2020). More efforts are needed not only to further improve ABL parameterizations and better represent subgrid processes within the stable ABL, but also to better characterize its influence on regional pollution.

(5) **Based on relative conclusions, emission reduction strategies in practice should be evaluated through scenario simulations.** Emission reductions aim to effectively and efficiently alleviate air pollution and limit its detrimental effects on humans and other living organisms. In practice, there are more questions to be answered, including when and where to reduce emissions, and which species and/or sources to focus on. Conclusions from transport-related studies are indicative of the design of feasible emission reduction strategies, but whether they work out well should be further assessed based on the results of scenario simulations. In addition, emission reduction would become more complex if cooperation between several regions and other objectives (e.g., high economic efficiency and low inequality) were taken into account. Therefore, tools are needed for such a task of multi-objective optimization. The results can provide suggestions for both short-term air pollution reduction and long-term air quality improvement.

Abbreviations

ABL	Atmospheric boundary layers
BC	Black carbon
BFM	Brute Force Method
CRT	Cross-regional transport
CTMs	Chemical transport models
DDM	Decoupled Direct Method
DOAS	Differential optical absorption spectroscopy
FT	Free troposphere
GZB	Guanzhong Basin
HDDM	Higher-order Decoupled Direct Method
MAX-DOAS	Multi-axis differential optical absorption spectroscopy
MEIC	Multi-resolution Emission Inventory for China
Middle-YR	Middle areas of the Yangtze River
NCP	North China Plain
NE/NW-China	Northeast/Northwest China
NMF	Non-negative matrix factor decomposition
NO_x	Nitrogen oxides; $= NO + NO_2$

NO_y	Reactive nitrogen; $= NO_x + HONO + NO_3 + N_2O_5 + HNO_3 +$ particulate nitrate + organic nitrates + …
O_x	Odd oxygen; $= O_3 + NO_2$
OVOCs	Oxidized volatile organic compounds
PAHs	Polycyclic aromatic hydrocarbons
PANs	Peroxyacyl nitrates
PCA	Principal component analysis
PM	Particulate matters
$PM_{2.5}$	Fine particulate matters (diameters ≤2.5 μm)
PM_{10}	Coarse particulate matters (diameters ≤10 μm)
POC	Primary organic carbon
PRD	Pearl River Delta
SCB	Sichuan Basin
SI	Stratospheric intrusion
SOA	Secondary organic aerosol
SWPs	Synoptic weather patterns
UAV	Unmanned aerial vehicle
VOCs	Volatile organic compounds
WCB	Warm conveyor belt
YRD	Yangtze River Delta

CRediT authorship contribution statement

Kun Qu: Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Yu Yan:** Writing – review & editing, Writing – original draft, Investigation. **Xuesong Wang:** Writing – review & editing, Supervision, Project administration, Investigation, Conceptualization. **Xipeng Jin:** Writing – review & editing, Investigation. **Mihalis Vrekoussis:** Writing – review & editing. **Maria Kanakidou:** Writing – review & editing. **Guy P. Brasseur:** Writing – review & editing. **Tingkun Lin:** Investigation. **Teng Xiao:** Investigation. **Xuhui Cai:** Writing – review & editing, Conceptualization. **Limin Zeng:** Writing – review & editing, Conceptualization. **Yuanhang Zhang:** Writing – review & editing, Supervision, Conceptualization.

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.

Data availability

No data was used for the research described in the article.

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