Influences of stratospheric intrusions to high summer surface ozone over a heavily industrialized region in northern China

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Abstract

The stratospheric contribution to tropospheric ozone has long been a topic of much debate over the past few decades. In this study, we leveraged multiple datasets from surface, sounding and satellite observations to reanalysis datasets, along with a global chemical transport model (Global Nested Air Quality Prediction Modelling System, GNAQPMS) to investigate the impact of a stratospheric-to-tropospheric transport (STT) event characterized by long duration and wide range in the summer on surface high ozone episodes over heavily industrialized regions in northern China. In 14–18 August 2019, the ERA5 reanalysis datasets showed a potential vorticity (PV) tongue and a deep, upper-level trough penetrate towards 35\textdegree N over the North China Plain (NCP), indicating the occurrence of a stratospheric intrusion. From Atmospheric Infrared Sounder (AIRS) measurements, we found that the ozone-rich, stratospheric air mass had been injected into the lower altitudes. The GNAQPMS generally captured the featured layers, although there was a slight underestimation in the low troposphere. The averaged magnitudes of stratospheric contribution (O3S) and percentage (O3F) simulated by GNAQPMS were 3–20 µg m\textsuperscript{-3} and 6%–20%, respectively, while the Whole Atmosphere Community Climate Model (WACCM) indicated a higher stratospheric contribution by 3–5 µg m\textsuperscript{-3}. Through this study, we give our opinions on the controversial topic of a more thorough understanding of the influence of natural processes apart from anthropogenic emissions, even in a heavily polluted region during summer.

1. Introduction

Despite stringent emission control [1, 2] in recent years, ozone pollution has become an emerging concern in China [3]. Enhanced tropospheric ozone poses an increasing threat to human health [4], vegetation [5], and infrastructure [6]. As the third important greenhouse gas, O\textsubscript{3} potentially affects climate change [7].

In contrast to ground-level ozone generated by tropospheric photochemical reactions involving volatile organic compounds and nitrogen oxides [8], stratospheric-to-tropospheric transport (STT) is not a reasonably controllable or preventable exceptional event, which plays a vital role in determining the tropospheric O\textsubscript{3} budget, accounting for 20%–30% of O\textsubscript{3} resources in the middle latitudes of the Northern Hemisphere [9]. From a global perspective, STT is governed by the Brewer–Dobson circulation [10–12], in which tropospheric air enters the stratosphere in the tropics, upwells within the tropical stratosphere, and then spreads poleward before descending.

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in the middle and high latitudes. STT is a common occurrence at mid- and high latitudes where several synoptic processes such as tropospheric folding [13], cut-off lows [14, 15], wave breaking, and extratropical cyclones are prone to happen. Owing to the conductive combination of higher O3 concentrations in the stratosphere [16] and more active air mass motion at mid- and high latitudes, the STT ozone commonly peaks in the Northern Hemisphere extratropical in late winter and spring.

STT has long been a controversial topic focused on the degree to which stratospheric ozone can reach the surface [17, 18]. Intensive researches [19, 20] have been conducted centred on the western United States. In contrast, in China, a major effort in STT research has been qualitatively researching dynamic mechanisms in the Tibetan Plateau and Northeast China where the pollution is not serious using multiple observations [14, 21–25] and Lagrange models such as HYSLIGHT [26–28] and CLaMS [29, 30]. Researches have revealed that the strong downdrafts of typhoon periphery will strengthen the STT and caused the positive ozone anomalous [31–34] in heavily polluted region. However, quantification of the contribution of STT to surface ozone, especially in regions with heavy emissions is more important, but the research is still limited. Wang et al [35] employed the MOZART to quantify the contribution of a stratospheric intrusion to surface ozone in Nanjing with the magnitude of about 10 ppbv in summertime. Lu et al [36] used the GEOS-Chem to diagnose the monthly averaged stratospheric source contribution to surface ozone, and the ozone enhancement from stratosphere in North China Plain (NCP) is about 8 ppbv in June, indicating that stratospheric contribution in NCP, even in summertime cannot be ignored. Zhang et al [37] studied the STT contribution using two global models of GEOS-Chem and GFDL-AM4, and the former simulated 25 ppbv lower ozone than the former, indicating large uncertainties [19, 38–40] in single model quantification. The previous researches have shown that importance of stratospheric intrusions to surface ozone over the NCP region in summertime is not well established, and the model performance in quantifying stratospheric contribution should be further evaluated.

To fill the knowledge gap and thus to provide scientific insight for effective pollution control measures in NCP [41–43], in this study, we combine two global CTMs (GNAQPMS and WACCM) with multiple datasets to comprehensively analyse an STT event characterized by long duration and wide range over the NCP region. We aimed to: (a) depict the causal relationship of stratospheric-tropospheric exchange, (b) quantify the stratospheric contribution to surface ozone, and (c) evaluate the applicability of GNAQPMS in modelling STT and provide insight into model improvements.

2. Materials and methods

2.1. Chemical transport model

Regional transport models have limited capabilities in simulating stratospheric-tropospheric exchange (STE) for their susceptibility to lateral boundary, and it is hard to distinguish whether the ozone transport to the region is from stratosphere. The O3 concentration and O3 contribution originating from stratospheric (O3S) were simulated in this study by GNAQPMS, a global chemical transport model developed by the Institute of Atmospheric Physics, Chinese Academy of Sciences. The meteorological data inputted into the GNAQPMS were simulated using the global version of the Weather Research and Forecasting model (GWRF-v4.0), because GWRF can provide fine resolution and useful diagnostic variables for GNAQPMS. This model [44, 45] is an extension of mesoscale weather research and forecasting (WRF) developed for GWRF applications. In this study, the GWRF model was driven by the US National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) data, with a horizontal resolution of 0.5° × 0.5°.

GNAQPMS is a global chemical transport model with terrain-following coordinates that includes detailed tropospheric O3-hydrocarbon gaseous chemistry (CBM-Z) [46], aqueous (RADM) chemistry [47], aerosol (ISORROPIAI1.7) chemistry [48], and heterogeneous chemistry [49]. For physical processes, GNAQPMS couples parameterisation of horizontal and vertical advection [50] with dry and wet deposition [51, 52]. An accurate radiative-transfer model (TUV version 4.5) with an eight-stream discrete ordinate solver was coupled online with GNAQPMS to provide photolysis rates [49]. The model has been utilized to evaluate mercury transport [53], and more recently the global distributions of gaseous pollutants [54], aerosol components [55, 56], and intercontinental transport [57].

Global anthropogenic emission data combine those for pollutants derived from the Emission Database for Global Atmospheric Research (EDGAR v5.0 http://edgar.jrc.ec.europa.eu/overview.php?v=50_AP) based on 2015 [58], non-methane volatile organic compounds (NMVOC) from EDGAR v4.3.2, and NH3 emissions from the Hemispheric transport of air pollution (HTAP v2.2) emission inventory for 2010 (data available at https://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123) [59], and the soil NOx emission is also contained to reduce the uncertainty of ozone simulation [60]. Biogenic emission data were obtained from the Model of Emission of Gases and Aerosols from Nature (MEGANv2.1; http://accent.aero.jussieu.fr/database_table_inventories.php) developed by the US National Center for Atmospheric Research (NCAR) [61]. The biomass burning emissions were calculated
from the daily global fire inventory from NCAR (FINN v1) [62] with a resolution of $0.1^\circ \times 0.1^\circ$ based on 2018. Climatic $1^\circ \times 1^\circ$ lightning emissions from the Global Emissions Inventory Activity (GEIA) were included in this study. We adopted emissions of CO, CHBr$_3$, CH$_3$I, and CH$_3$Br$_2$ from the CAMS-81 project with a resolution of $0.5^\circ$ [63].

We conducted simulations for August 2019 at global $0.5^\circ$ latitude by $0.5^\circ$ longitude resolution with a one-month spin-up; vertically, 40 layers were created from the model bottom to the model top of 20 km. To rationally depict stratospheric chemical behaviour in the model, we employed the relaxation to monthly averaged climatology from the Model for Ozone and Related Chemical Tracers (MOZART v2.4), with 2.8° resolution [64]. To rationally depict the tropopause height, we replaced the old thermal-defined method with the potential vorticity (PV)-defined method, in which a PV below 1.6 defines the troposphere [65–67]. An online source-receptor relationship module, based on a tagged tracer approach coupled to GNAQPMS was employed to quantitatively track stratospheric O$_3$ contribution [68].

The WACCM is a comprehensive numerical model, which is being run in real-time with the configuration of CESM2. The simulation was preformed using the specified dynamical configuration, and the meteorological data inputted into the WACCM were simulated using the NASA/GMAO GEOS-5. WACCM is a ‘high top’ model extends from the surface to approximately $140$ km. It has 66 vertical levels and horizontal resolution of $1.98$ latitude by $2.58$ longitude. The chemistry module of WACCM is based on the version 3 of the Model for Ozone and Related Chemical Tracers [69], containing 59 species and 217 gas-phase chemical reactions. WACCM explicitly represents the radiative transfer of carbon dioxide, methane, nitrous oxide and two halogens (CFC11 and CFC12). Different from GNAQPMS focused on troposphere chemistry, the WACCM does not include a detailed representation of tropospheric chemistry beyond methane and CO oxidation. WACCM has superior performance in simulating stratospheric ozone, for it has complex stratospheric chemistry and a representation of quasi-biennial oscillation (QBO) [70] and nonorographic gravity waves [71]. WACCM used O$_3$ tracer to tag the stratospheric ozone contribution [72, 73]. WACCM has been widely used to simulate atmospheric composition and coupling processes between the stratosphere and troposphere, to reduce uncertainty, the WACCM output (www.acom.ucar.edu/wacccm/DATA) was also used to evaluate the stratospheric contribution against GNAQPMS in this study [74, 75].

2.2. Observations
We compiled an ensemble of surface observations, sounding observations, reanalysis datasets and satellite observations to evaluate model performance and analyse STT events.

2.2.1. CENEMC
Hourly surface pollutant observations for the simulation period were obtained from the China National Environmental Monitoring Centre (CENEMC, www. cnenm.cn). We used CENEMC to evaluate the model performance at surface layer. In addition, six observation sites were selected: EEDS (109.81°E, 39.59°N), BT (110.03°E, 40.55°N), HHHT (111.66°E, 40.85°N), DT (114.66°E, 40.13°N), BJ (116.23°E, 39.93°N), TL (121.67°E, 40.02°N). More details were provided in figure S1.

2.2.2. Sounding
The upper-air sounding data was accessed from the Department of Atmospheric Science, University of Wyoming (http://weather.uwyo.edu/upperair/ sounding.html) at two stations (Ulaan-Baator, 106.52°E, 47.55°N; and Dalanzadgad, 104.25°E, 43.35°N) (figure S1). We used the sounding data to test the vertical distribution of meteorological fields.

2.2.3. ERA5
The fifth generation of ECMWF atmospheric reanalysis (ERA5) is the newest generation of reanalysis datasets with enhanced temporal and spatial resolution, allowing for a detailed evolution of weather systems. Comparison with radiosonde and PILOT data prior to assimilation showed an improved fit for both ozone and meteorological variables [76]. We collected the variables of PV, geopotential height, u-component of wind, and v-component of wind from ERA5 datasets for 2015–2020 to diagnose STE occurrences.

2.3.4. NCEP
Additionally, we used the variables of air temperature, vertical velocity ($\omega$, in Pascal/s) and tropopause height collected from the US NCEP Global Forecasting System Final Analysis (FNL, www.psl.noaa.gov/data/gridded/data.ncep.reanalysis.html) to provide more observed synoptic conditions favourable for STE events.

2.3.5. AIRS
The Atmospheric Infrared Sounder (AIRS) is a grating spectrometer ($R = 1200$) aboard the second Earth Observing System (EOS) polar-orbiting platform, EOS Aqua [77]. We used O$_3$ retrieved from AIRS Level 3 (https://disc.gsfc.nasa.gov/datasets/AIRS3STD_7.0/summary) binned into $1^\circ \times 1^\circ$ to provide a large-scale view of horizontal and vertical ozone structures during the STE occurrence. The AIRS ozone retrievals has been widely used to estimate vertical structure of ozone in STT, and the high bias of which in the troposphere has been reduced
by using tropopause-based reference climatologies in version5 [78].

3. Results and discussion

3.1. Synoptic conditions conducive to STT occurrences

We examined the dominant meteorological conditions favourable for STT occurrences during 14–19 August 2019. As shown in figure 1(a), the climatological tropopause height was approximately 150 hPa over the NCP; moreover, it was abnormally low during the period of 14–19 August. In the case of adiabatic and frictionless, the PV value of the air mass is conserved, so PV is a suitable tracer for diagnosing STT occurrence. Compared to climatological PV, the PV pattern during 14–19 August displayed a hook-shaped stream of air extending from northern China to the Yangtze River Delta. Its magnitude reached 12 PVU in the mass centre, which is a typical feature for an STT event [79]; this structure was captured by GWRF (figure S2). Figure 1(b) depicted the development and intensification of air circulation. The strong anticyclone centre clearly corresponded to the South Asian High, accompanied by an upper-level trough centred in the NCP. On 14 August, the upper-level trough was shadowed and weak. The temperature field lagged the height field, which promoted the intensification of the upper-level trough. On 17 August, the upper-level trough penetrated the southernmost part of China, facilitating the descent of stratospheric air masses into the lower layer. On 19 August, the upper-level trough withdrew from China, ending the STT event. Additionally, we obtained more direct information from the vertical distribution of relative humidity (RH) and air temperature, as shown in figures S3 and S4. Simulations and observations showed strong agreement, especially for air temperature, providing confidence for the following analysis. During 14–16 August, the soundings detected a sudden decrease in RH in the layer of 400–600 hPa, which deepened on 15 August, indicating that the dry air mass originated in the stratosphere.

In 6–16 August 2019, a typhoon KROS A (http://agora.ex.nii.ac.jp/digitaltyphoon/summary/wnp/s/201910.html.en) generated over western North Pacific at 06:00 on August and remained active during August 8–14, which facilitated the STT process. A typhoon developed in the western Pacific typically can extend to the lower stratospheric at heights of about 10–18 km, with strong vertical circulations. Researches have shown that positive ozone anomalies are likely to occur when the region is under the control of typhoon periphery (about 600–1700 km away) with strong downdrafts. From figure S4, as typhoon KROS A moved northwest from August 13 to August 14, the tropical cyclone gradually weakened and finally strengthened the cyclone controlling the STT, and a positive ozone anomalies appeared on the surface of its periphery.

3.2. Vertical distribution of ozone during STT occurrences

Figure 2(a) illustrated the ozone horizontal distribution retrieved from the AIRS at 300 hPa. On 14 August, the ozone concentration over the NCP was approximately 95 ppb at 300 hPa. A tongue with high O3 exceeding 110 ppbv stretched to 30° N on 15 August. Consistent with the development of the upper-level trough, the ozone tongue gradually moved eastward. The magnitude of the O3 mixing ratio centred in the ozone tongue reached 140 ppb over the NCP on 16–18 August 2019. From the AIRS measurements, we concluded that the air mass containing rich ozone from the stratosphere was indeed injected into the troposphere; however, it was uncertain whether the stratospheric air mass reached the surface. To gain insights into the deep descent of stratospheric O3, four cross sections along the ozone tongues on 15–18 August were selected. From the zonal winds depicted in figure 2(b), we observed that the downward motion of air was within the ozone tongue. A steep concentration gradient was found at an altitude of 200 hPa from AIRS measurements, indicating the positions of the upper stratosphere and lower troposphere (UTLS). A belt of high ozone concentration with a magnitude of approximately 120 ppb can be seen from the AIRS measurement; it stretched to the lower troposphere.

3.3. Evaluation of GNAQPMS

Before quantifying the stratospheric contribution, we evaluated the horizontal and vertical performance of GNAQPMS. Figure S6 shows the horizontal distribution of GNAQPMS simulated O3 against CNEMC observations. GNAQPMS showed good spatial correlations with CNEMC observations, and the magnitude of correlation coefficient was over 0.8. However, a slight underestimation (with the NMB of about −15%) was also found over the NCP. This underestimation can be largely attributed to uncertainties in emissions inventories [60, 80] and photochemical treatment [81]. Specifically, soil NOx emissions (1 TgN yr⁻¹) in EDGAR are much smaller than the magnitude of current bottom-up or top-down estimates (3–20 TgN yr⁻¹) [82], which may explain the underestimation of ozone simulation. In addition, the CNEMC observation sites are mostly located in urban region with high ozone levels, while the model output are averaged with rural sites and urban sites.

Because we lacked radar observations, we conducted vertical evaluations of GNPQMS simulations against AIRS observations. As shown in figure 2(b), GNAQPMS captured the vertical structure of ozone
during STT events. The altitude of the UTLS simulated by GNAPMS was lower than that of the AIRS measurements. An enhanced ozone belt was also observed in the GNAPMS simulations, which also stretched to the lower troposphere, but the magnitude of ozone was smaller than that from AIRS measurements, especially in the regions of 600 hPa and 800 hPa. This underestimation of the ozone vertical distribution can be partly attributed to the lower vertical resolution of AIRS measurements. Downward flux of ozone in GNAQPMS is mainly controlled by the vertical wind velocity simulated by GWRF, so the GWRF errors may caused the underestimation of GNAQPMS. In addition, the simplified stratospheric scheme in GNAQPMS also leads to the underestimation.

3.4. Contribution of stratospheric intrusions

From the thermal and dynamical analysis above, we can conclude that an STT event characterized by long duration and wide range occurred in the NCP during the period 14–19 August 2019. Next, we quantified the stratospheric contributions to surface ozone. From figure 3, the enhanced O3S with a magnitude of approximately 20 µg m⁻³ simulated by GNAQPMS was located at 40° N-45° N, 105° E-115° E on 14 August. On 15 August, the GNAQPMS simulated O3S was approximately 15 µg m⁻³, and the O3S of the
NCP region was within the range of 3–10 µg m\(^{-3}\) on 16–18 August. In contrast, the stratospheric contribution simulated by the WACCM was smaller than that simulated by the GNAPMS on 14 and 15 August. During 16–18 August, O3S simulated by WACCM was within the range of 6–15 µg m\(^{-3}\), which was larger than that of GNAPMS by 3–5 µg m\(^{-3}\). Overall, the stratospheric contribution quantified by GNAPMS is smaller than that of WACCM. It can be partly attributed to the difference of the tagging method, because GNAPMS used the tagged method while WACCM used the O3 tracer \[72, 73\]. And researches have shown that the O3S tracer gives a greater estimate of stratospheric ozone than the tagged ozone \[39\]. From the analysis of synoptic conditions discussed in section 3.1, we found that the strongest upper-level trough existed on 18 August, while the largest stratospheric contribution simulated by GNAPMS and WACCM occurred on 14 and 15 August, indicating an inconsistency between the meteorological and chemical conditions. This inconsistency can be partly attributed to the terrain height, because the stratospheric air mass will first influence the western NCP with higher terrain height, where the stratospheric ozone is easier to reach surface.

We then calculated the relative contribution from the stratosphere to surface ozone (denoted by O3F). On 14 and 15 August, O3F simulated by GANQPMS reached 20%, while the relative stratospheric contribution was approximately 12% in the NCP during 17–18 August. In contrast, O3F simulated by WACCM was approximately 18% more than the NCP during STT occurrence. In contrast to the O3S horizontal distribution, the O3F simulated by GNAPMS captured the STT pattern, indicating that GNAPMS reasonably reproduced its dynamic process.

To further analyse the evolution of ozone concentration and O3S, we selected six stations from west to east. The GNAPMS generally captured the diurnal variation of ozone (figure 4); however, the simulated ozone concentration was lower than that of the observation. The biases have been conducted between the simulations and measurements, and we found that the GNAPMS have comparable performance within period with stratospheric intrusions and without stratospheric intrusions. Since O3 is more abundant in the stratosphere than in the troposphere, CO is directly emitted or chemically formed in the troposphere \[83\]. We next performed a correlation analysis between O3 and CO to identify the possible origins of surface O3 during STT occurrence \[84, 85\].
Figure 4. Time series of observed O\textsubscript{3} concentrations, GNAQPMS simulated O\textsubscript{3} concentrations and O\textsubscript{3}S (Left panel, (a1)–(f1)) at six stations at local times. Blue shading highlights the period of stratospheric intrusions. The scatter plots of O\textsubscript{3} and CO in six stations (Right panel, (a2)–(f2)), and the dots in purple/green denotes the period with/without STT. The MB\textsubscript{in} and MB\textsubscript{out} denotes the bias between the simulation and observations period with/without stratospheric ozone. Note that the magnitude of O\textsubscript{3}S and O\textsubscript{3}F is averaged during the shaded period. The stratospheric influence period was selected based on the enhanced O\textsubscript{3}S in surface.

The negative correlation shown in figure 4(a2)–(f2) supports the hypothesis of a stratospheric origin of air mass. From the blue shading in figure 4, with the movement of upper-level trough from west to east, the stations in the west would be earlier affected by the stratospheric intrusion than the stations in the east. At the EEDS, BT, and HHHT sites, the peak magnitude of O\textsubscript{3}S reached 18 \(\mu\text{g m}^{-3}\), 11 \(\mu\text{g m}^{-3}\) and 20 \(\mu\text{g m}^{-3}\), respectively, with averaged stratospheric contributions of about 7%–10%. The stratospheric contribution peaked in August 15 in DT, with peaked O\textsubscript{3}S and O\textsubscript{3}F values of 8.98 \(\mu\text{g m}^{-3}\) and 14%, respectively. In BJ, the stratospheric contribution peaked on 16 August with a magnitude of 5 \(\mu\text{g m}^{-3}\). The stratospheric contribution to the surface ozone in TL is weak, and the averaged stratospheric contribution is approximately 3.18 \(\mu\text{g m}^{-3}\). In figure S7, we present more information about the stratospheric concentration of ozone in the boundary layer. When STT events occurred, the magnitude of O\textsubscript{3}S in the boundary layer was between 10 and 50 ppb, comparable to that reported by Banerjee \textit{et al} [86].

4. Conclusions

In this study, we analysed the STT process that occurred on 14–19 August 2019 over the NCP, which was characterized by a long duration and wide range. First, we analysed the synoptic conditions conducive to STT occurrence using multiple observations. The upper-level trough centred in the NCP region facilitated O\textsubscript{3}-enriched stratospheric air mass injection into the troposphere. Second, from the AIRS measurement, we found the ozone enhancement over the NCP to be 300 hPa, and the stratospheric air masses injected to the lower altitudes. Finally, we quantified the stratospheric contribution using GNAQPMS and evaluated its performance against WACCM. The stratospheric contribution quantified by GNAQPMS is within the range of 3–20 \(\mu\text{g m}^{-3}\), indicating large spatiotemporal variation.

The GNAQPMS generally captured the horizontal distribution and vertical structure of ozone illustrated by surface ozone observations and satellite measurements, and quantified the stratospheric contribution...
with a magnitude comparable to that of the WACCM. However, the GNAQPMS overestimated the O$_3$ mixing ratio in the UTLS region, which can be attributed to the lack of explicit depiction of stratospheric chemistry [87]. In addition, the method of quantifying stratospheric contributions also introduces uncertainties. Overall, accurately simulating stratospheric intrusion processes and rationally quantifying stratospheric contributions is still a challenging international subject for global atmospheric chemistry models [88–91]. This study is our first attempt to address this issue and provide insights for model improvements (e.g. grid configuration, tropopause definition, top boundary and tagged methodology); it provides a foundational step for future work.

Here, we show preliminary evidence of STT events during the summer over the NCP. The STT event that occurred on 14–19 August 2019 is not unique; we found another 24 STT events over the NCP in summer (listed in table S1). The most frequent STT events occurred in 2016 and 2019 with the value of eight, while it only occurred twice in other years in summer. The processes were selected based on ERA5 data and followed two principles: the pattern correlation with the horizontal of PV at 200 hPa in 14–19 August 2019 is larger than 0.6; the dynamical tropopause is lower than climatological values. As shown in figure S8, we conducted a composite analysis of 24 selected STT events, and positive PV anomalies and negative GPH anomalies were apparent over the NCP region. From figures S9(b) and (e), we can see positive westerly anomalies in lower latitudes while negative westerly anomalies in higher latitudes, indicating a southward shift of westerly jets. The southward movement of zonal winds deepens the cyclone and thus promoting the stratospheric air mass sinking. O$_3$-rich air mass injected into the troposphere and exerted positive ozone anomalies even in the lower troposphere (below 800 hPa, see in figure S9(f)). However, not all of the selected STT events reached the surface, whether they can be injected to the surface is complicated by multiple factors and more work is needed in the future. Apart from Qinghai-Tibet Plateau and northeast China, we identified the North China Plain as a potential ‘hotspot’ sensitive to STT.

Facing the increasingly serious ozone pollution, the Chinese government has taken plenty of measurements and designated air quality standard. As a not reasonably controllable or preventable exceptional event, the stratospheric contribution may in some degree dilute the positive effect of pollution control measures, and pose challenge for satisfying the ozone air quality threshold. We highlight the need to improve our understanding of natural processes such as episodic stratospheric intrusions, in addition to anthropogenic emissions, even in heavily polluted regions, and the air quality monitoring data influenced by such natural processes may be considered to be excluded from regulatory determinations related to violations of the air quality standard in the future.

**Data availability statement**

The data that support the findings of this study are available upon request from the authors.

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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