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Research Paper

Impacts of Indian summer monsoon and stratospheric intrusion on air pollutants in the inland Tibetan Plateau

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ABSTRACT

Air pollutants can be transported to the pristine regions such as the Tibetan Plateau, by monsoon and stratospheric intrusion. The Tibetan Plateau region has limited local anthropogenic emissions, while this region is influenced strongly by transport of heavy emissions mainly from South Asia. We conducted a comprehensive study on various air pollutants ($PM_{2.5}$, total gaseous mercury, and surface ozone) at Nam Co Station in the inland Tibetan Plateau. Monthly mean $PM_{2.5}$ concentration at Nam Co peaked in April before monsoon season, and decreased during the whole monsoon season (June–September). Monthly mean total gaseous mercury concentrations at Nam Co peaked in July and were in high levels during monsoon season. The Indian summer monsoon acted as a facilitator for transporting gaseous pollutants (total gaseous mercury) but a suppressor for particulate pollutants ($PM_{2.5}$) during the monsoon season. Different from both $PM_{2.5}$ and total gaseous mercury variabilities, surface ozone concentrations at Nam Co are primarily attributed to stratospheric intrusion of ozone and peaked in May. The effects of the Indian summer monsoon and stratospheric intrusion on air pollutants in the inland Tibetan Plateau are complex and require further studies.

1. Introduction

In recent years, there is a growing body of studies that provide increasing evidence for the transport of atmospheric pollutants from South Asia, one of the most polluted regions in the world, across the Himalayas to the Tibetan Plateau, which could adversely impact a relatively pristine environment (e.g., Lüthi et al., 2015; Dong et al., 2017; Lin et al., 2019; Jiao et al., 2021). The Tibetan Plateau is a region with several ecosystems sensitive to regional and global changes, minimal human activities, and supposedly pristine environment (Yao, 2019; Zhao et al., 2019). The increasing atmospheric pollutants in the Tibetan Plateau is linked to a multitude of adverse impacts on human health, ecosystems such as the cryosphere (also known as the "Third Pole" due to the sheer size of its cryosphere), and weather and climate change (Kang et al., 2019). Previous studies have explored and documented the extent of atmospheric pollutants over the Tibetan Plateau and the mechanisms and pathways of their transport, in particular the *trans*-Himalayan transport of atmospheric aerosols, from various source regions to the Tibetan Plateau (Lüthi et al., 2015; Lin et al., 2019; Kang et al., 2019; Li et al., 2020). The dynamics of the Indian summer monsoon (ISM) is considered as an important driver that facilitates the transport of atmospheric/air pollution into the Tibetan Plateau (Sheng et al., 2013; Li et al., 2016, 2020; Kang et al., 2019). In addition to the ISM, stratospheric intrusion can lead to high levels of surface ozone in the Tibetan Plateau and the Himalayan mountain region (Cristofanelli et al., 2010), a region with a rel-

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atively cleaner atmospheric environment than its surrounding areas. However, the role of the ISM and stratospheric intrusion in influencing concentrations of various air pollutants over the inland Tibetan Plateau remains uncertain due to a lack of concurrent observations and integrated analysis of various air pollutants. In this study, we explore hitherto unavailable distinct seasonal characteristics, sources regions and driving mechanisms of three key air pollutants. Using ground-based simultaneous observations at a typical background site (Nam Co) in the inland Tibetan Plateau, data acquired with the CALIPSO (The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) observations and the CAM-Chem (Community Atmosphere Model with Chemistry) model simulations, we argue that the ISM and the stratospheric intrusion have various distinctly different effects on air pollutants in the Tibetan Plateau. Therefore we highlight the complex sources and mechanisms of air pollutants in the inland Tibetan Plateau.

2. Materials and methods

Simultaneous observation of $PM_{2.5}$ (particulate matter with an aerodynamic diameter equal to or less than 2.5 μm), TGM (total gaseous mercury), a pollutant with increasing global interest, and O_3 (surface ozone) were performed at Nam Co Station in the inland Tibetan Plateau (Fig. 1) for a period of 3 years from January 1st, 2012 to December 31st, 2014 (Supplementary Data, Fig. S1). These three air pollutants with distinct characteristics can represent different types of air pollutants in terms of their sources, transport, and transformation processes and thus they were used in this study to investigate the factors that controlled their temporal variations. The $PM_{2.5}$, TGM, and O_3 were measured by a Tapered Element Oscillating Microbalance (model RP 1400, TEOM, Thermo Environmental Instruments, USA), a Tekran automated ambient mercury analyzer (model 2537B, Tekran Instruments Corp., Canada), and an ozone analyzer (model 49i, Thermo Environmental Instruments, USA), respectively. The details of the observation site and methodology are described in the supplement (Supplementary Data).

3. Results and discussion

The monthly mean $PM_{2.5}$ concentration at Nam Co peaked in April (in the pre-monsoon season) which is before the summer rainy monsoon season (June–September) (Fig. 2a). During the pre-monsoon season, emissions from various anthropogenic activities (e.g., agro-residue burning) and natural sources (e.g., forest fires) in South Asia are also at their peak. The prevailing meteorological conditions over South Asia and the Tibetan Plateau during the pre-monsoon season favor transport of large amounts of emissions from South Asia and Southeast Asia to the Tibetan Plateau (Lüthi et al., 2015; Lin et al., 2019; Kang et al., 2019; Li et al., 2020). During monsoon season, the monthly mean $PM_{2.5}$ concentrations were dramatically decreased and the daily mean $PM_{2.5}$ concentrations at the Nam Co Station during January 2012–December 2014 were negatively correlated (correlation coefficient, $r = -0.33$) (Fig. 2b) with the ISM index, which is derived by averaging the negative value of the outgoing longwave radiation over the Bay of Bengal-Indian region (10°N–25°N, 70°E–100°E) (Wang and Fan, 1999). In contrast to $PM_{2.5}$, the TGM daily concentrations were positively correlated with the ISM index ($r = 0.54$) (Fig. 2b), and the monthly mean TGM concentrations were higher during the entire monsoon season with the highest value in the month of July (Fig. 2a), indicating that the meteorological conditions during summer monsoon were conducive to transport of TGM to the Tibetan Plateau. It was also high in April, indicating the dry weather in pre-monsoon favored transport of both TGM and $PM_{2.5}$ together to the Tibetan Plateau. Unlike $PM_{2.5}$ and TGM, the monthly mean surface ozone mixing ratio was maximum in the month of May (Fig. 2a) and it was not correlated with the ISM index ($r = 0.01$)

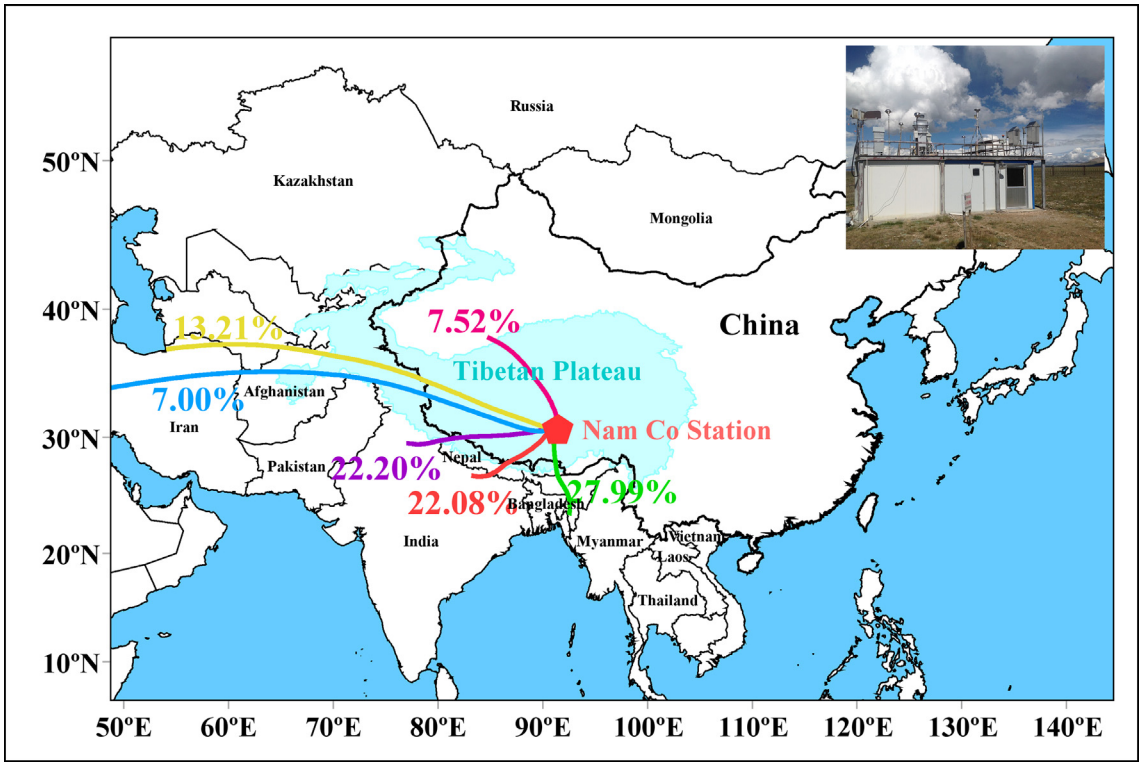

Fig. 1. Location of Nam Co Station in the Tibetan Plateau (indicated by cyan contour) and the pollution monitoring building shown in the inset.

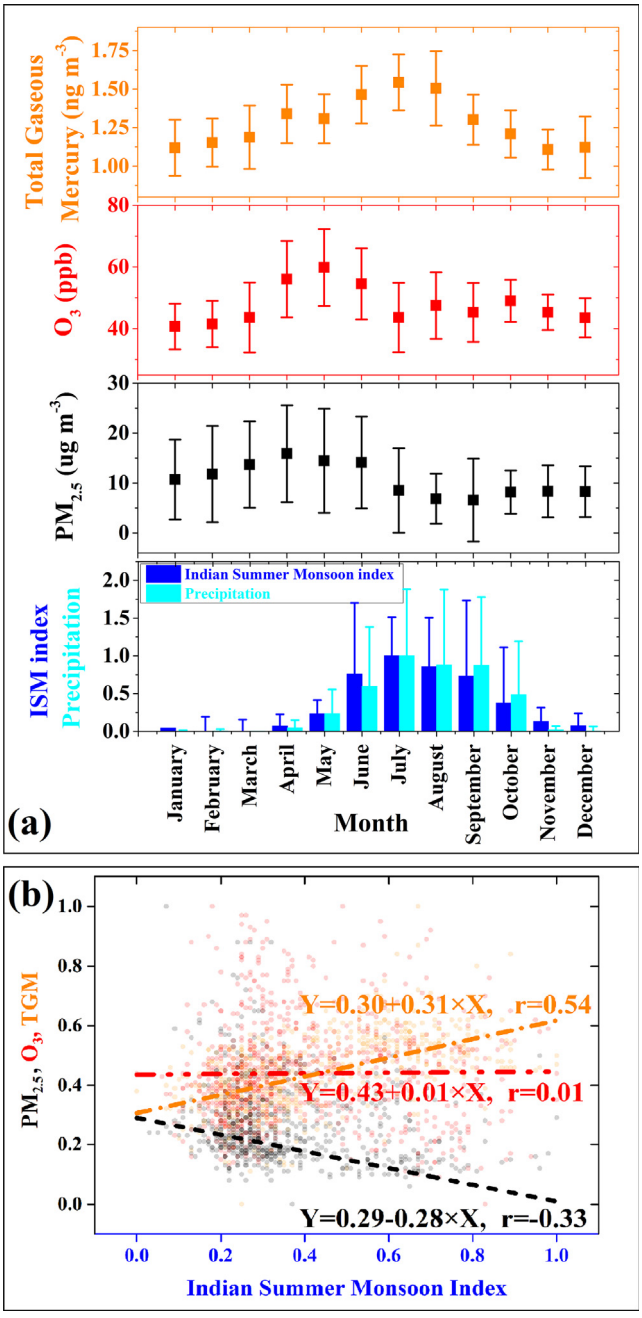

Fig. 2. Monthly mean variations of hourly mean total gaseous mercury (TGM), surface ozone, $PM_{2.5}$, normalized levels of Indian summer monsoon (ISM) index, and normalized levels of precipitation at Nam Co Station during 2012–2014 (a). Relations between daily mean levels of normalized surface ozone (in red), TGM (in orange), and $PM_{2.5}$ (in black) at Nam Co Station and the Indian summer monsoon index from January 2012 to December 2014 (b), $p < 0.05$.

(Fig. 2b), suggesting a different mechanism that mostly controlled the surface ozone.

$PM_{2.5}$, TGM, and surface ozone showed different seasonal variations due to their different sources and atmospheric processing, transport and removal mechanisms. The ISM and stratospheric intrusion are two key mechanisms that play different roles in regulating the transport of these air pollutants over the Tibetan Plateau. During a period of 3 years the majority (ca. 73%) of air masses, before arriving at the Nam Co site, originated and/or passed over major heavily polluted source regions in South Asia, in particular the Indo-Gangetic Plain. The remaining 20% and 7% originated in western Asia (Arabian Peninsula) and western China/Central Asia (Fig. 1), indicating clearly that South Asia is a major source region of atmospheric pollutants to even the inland Tibetan Plateau. The episodes of high $PM_{2.5}$ at Nam Co occurred mostly during the pre-monsoon season (Liu et al., 2017) and they were mostly associated with the air masses from South Asia (Fig. 3a and b). The CALIPSO data analysis (Supplementary Data, Fig. S2) and the CAM-Chem model simulations (Fig. 3c and d) show that South Asia had extremely high levels of natural dust and pollution-dust mixture during pre-monsoon season. During the ISM season the frequent and heavy precipitations in the region suppress emissions from some sources (e.g., forest fires) and wash out $PM_{2.5}$ from atmosphere, while at the same time some major seasonal sources in South Asia like the brick production are absent. Interestingly, in contrast to $PM_{2.5}$, the TGM correlated strongly positively with the ISM index. This is because TGM (especially elemental mercury) is only sparingly water-soluble (while $PM_{2.5}$ is washed out by rains and clouds) and thus can be transported over a long distance from South Asia to the inland Tibetan Plateau remote from man-made sources during the monsoon season (Yin et al., 2018). The eastern Indo-Gangetic Plain is identified as the largest potential source region of TGM to Nam Co (Fig. 4), and the air masses passed through eastern Tibetan Plateau potentially contributed to high levels. Our study provides an important insight into TGM over Tibetan Plateau showing that maximum transport of TGM from South Asia to Tibetan Plateau occurs during the ISM season. The ISM index had a very weak positive correlation with surface ozone variation at Nam Co, indicating that ISM does not play any significant role in regulating surface ozone over the inland Tibetan Plateau. It was reported in a previous study that the surface ozone at Nam Co is strongly influenced by the intrusion of stratospheric ozone into the planetary boundary layer (Yin et al., 2017). The ratio O_3S/O_3 (where O_3S is a tracer used in the CAM-Chem model that represents the amount of ozone in the troposphere that came from the stratosphere) was frequently higher than 0.8 at Nam Co during the episodes with high surface ozone (Supplementary Data, Fig. S3), suggesting that more than 80% of surface ozone mixing ratio during peak episodes can be attributed to the stratospheric ozone intrusion. The correlation coefficient between surface ozone and O_3S in April and May 2012 was 0.5, indicating that stratospheric ozone is a major contributor to tropospheric ozone during the spring episodes. The evidence of the stratospheric intrusion process was also corroborated in the meridional cross-section analysis of potential vorticity, zonal winds, ozone mass mixing ratio, and O_3S/O_3 ratio (Fig. 5).

It is clear that the ambient concentrations of key pollutants (e.g., $PM_{2.5}$, TGM and surface ozone considered in this study) over Nam Co region, which is representative of the inland Tibetan Plateau with limited local anthropogenic emissions, is influenced strongly by transport of regional emissions, mainly from heavily polluted source regions in South Asia. It is found that the ISM strongly mediates the sources, transport and characteristics of $PM_{2.5}$ and TGM, and consequently the Tibetan Plateau experiences their completely different seasonal variations, whereas surface ozone is primarily attributed to stratospheric intrusion of ozone. The ISM acts as a facilitator for transport of gaseous pollutants but a suppressor for particulate pollutants during the monsoon season. The impacts of air pollutants, including three pollutants considered in this study, on air quality, climate and ecosystems in the Tibetan Plateau, a region with sensitive ecosystems, are

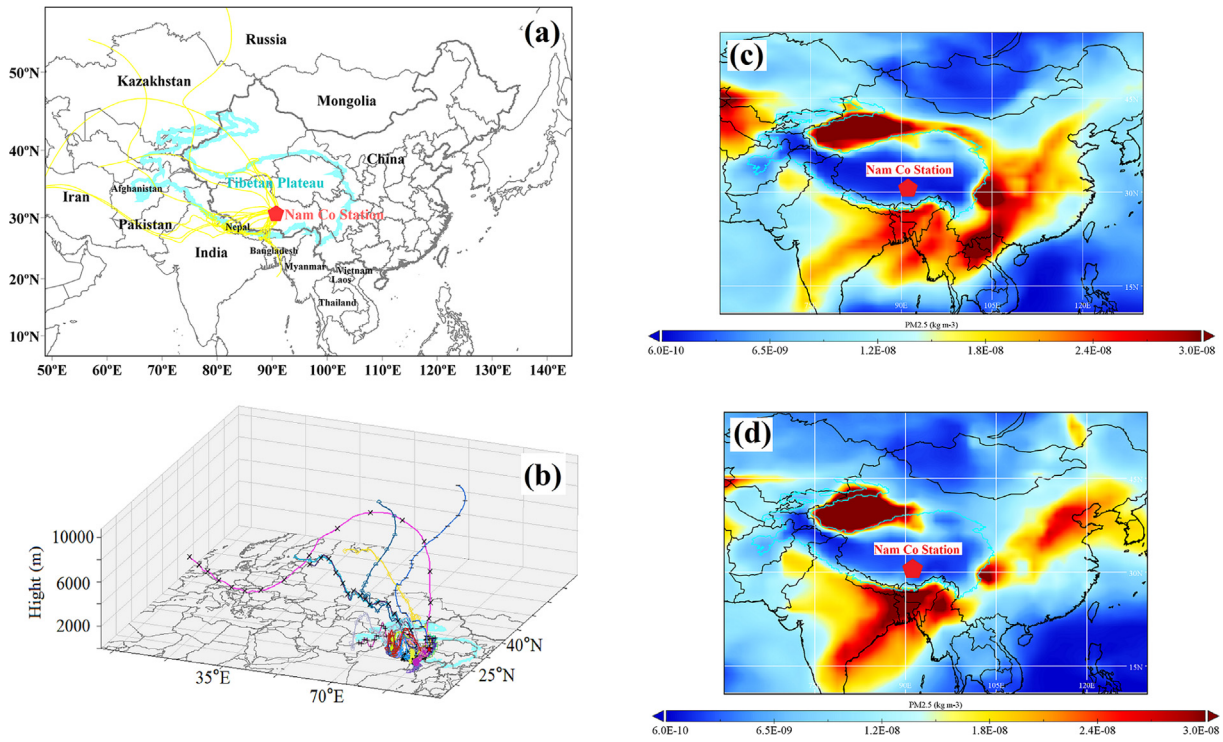

Fig. 3. Backward trajectories arriving Nam Co Station (a) and 3D view of backward trajectories when hourly mean $PM_{2.5}$ concentrations higher than 36.1 μg m^{-3} (mean + 3 × standard deviation during 2012–2014) at Nam Co Station in April and May in 2012 (selected for the high episodes analysis of $PM_{2.5}$) (b). Monthly mean concentrations of $PM_{2.5}$ at 857.5 hPa in April (c) and May (d) 2012 from CAM-chem. Nam Co Station shown by red pentagon.

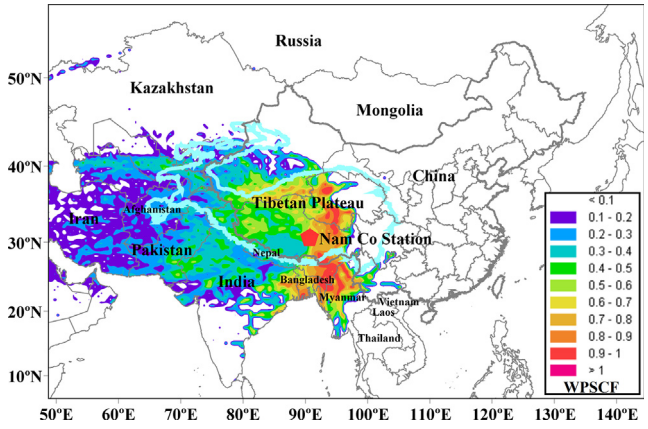

Fig. 4. Results of the Potential Source Contribution Function (PSCF) analysis investigating potential source regions to total gaseous mercury (TGM) at Nam Co Station during 2012–2014. Extent of the Tibetan Plateau shown in cyan. Nam Co Station is shown by a red pentagon. WPSCF = weighted PSCF, and high WPSCF value cells are indicative of areas of 'high potential' contributions for the TGM at Nam Co Station.

highly uncertain. It is clear from our analysis that the sources and mechanisms of individual air pollutants in the inland Tibetan Plateau are complex. Our study provides a scientific basis in calling for further follow-up studies of this complex and interactive system, in particular extensive chemistry-transport model simulations, to understand implications of changing concentrations of atmospheric pollutants over this region, role of the governing mechanisms, reduce the uncertainties in impact assessment, and to assess the appropriateness and values of various atmospheric environment protection policies.

Author contributions

Xiufeng Yin, Shichang Kang, and Qianggong Zhang designed the research and wrote the original draft. Benjamin de Foy, Dipesh Rupakheti and Maheswar Rupakheti contributed to paper writing. Xiufeng Yin, Zhiyuan Cong, Xin Wan, and Guoshuai Zhang contributed to data processing and analysis of results.

Declaration of Competing Interest

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

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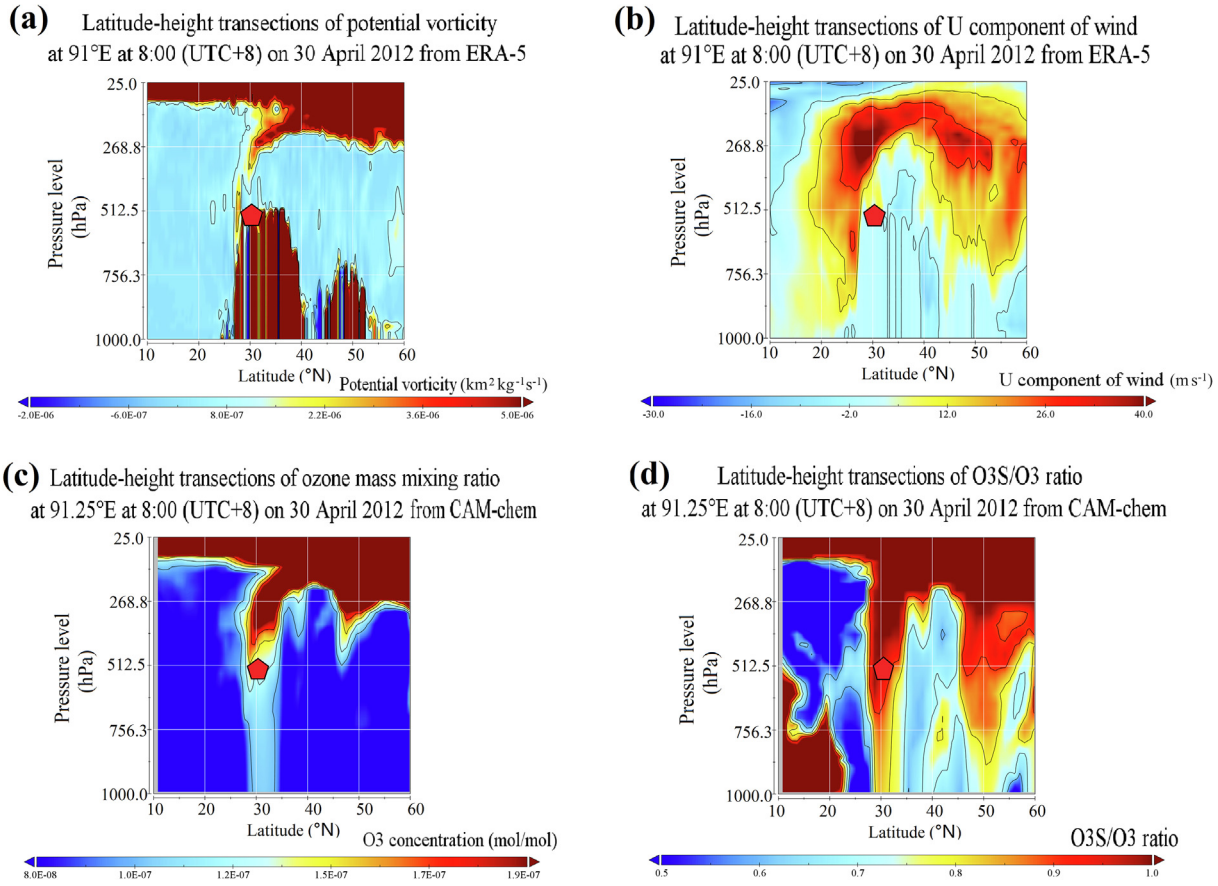

Fig. 5. Meridional cross-section at 91°/91.25°E (~over Nam Co Station) at 8:00 (UTC + 8) on 30th April 2012, derived from ERA5 reanalysis data and CAM-chem simulation, including potential vorticity (a), zonal winds (U component of wind) (b), ozone mass mixing ratio (c), and O_3S/O_3 ratio (d). Nam Co Station shown by red pentagon.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.gsf.2021.101255.

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