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#### **Environmental significance**

It is generally assumed that the main sources of atmospheric and glacial pollutants in the Third Pole region are transported from external locations. The main local emissions distribute in the Tibetan cities. Coronavirus Disease 2019 (COVID-19) accidently appeared in Tibet on August 7, 2022, and broke the 920 consecutive epidemic-free days. It provided a valuable opportunity for understanding how variations of urban air pollutants responded to the COVID-19 lockdown in the special highland cities of the third polar. Global research showed limited change in the total gaseous oxidant ( $O_x = NO_2 + O_3$ ) due to Lockdown 2020, as gains in O3 compensated for drops in NO2, including Tibetan urban cities. A slightly negative O3 anomaly and greater PM2.5 and NO2 reductions were found in the Lockdown 2022. The decrease of atmospheric oxidation smoothed the diurnal profiles of NO<sub>2</sub> and PM<sub>10</sub> and weakened those of O<sub>3</sub>, PM<sub>25</sub>, and PM<sub>2.5</sub>/CO. The random forest model-based weather normalization technique suggested that vehicle-related sources can contribute about 28% and 29% to PM<sub>2.5</sub> and NO<sub>2</sub>, respectively. Others were mainly from residential combustion emissions due to the weak industrialization in Tibet cities. This is the first study to understand the variations of air pollutant response to the large COVID-19 lockdown in Tibetan cities. The findings provide a reference for future air pollution management and urbanization in the Tibet.

#### 1 Introduction

Millions of people die every year from diseases caused by exposure to outdoor air pollution,<sup>1,2</sup> especially particulate matter with an aerodynamic diameter  $< 2.5 \ \mu m \ (PM_{2.5})$  and ozone  $(O_3)$ .<sup>3,4</sup> Surface ozone  $(O_3)$  pollution can absorb

ultraviolet rays and also impede plant growth and biomass accumulation, thereby further increasing global warming.<sup>5</sup> The Tibetan Plateau (TP), containing the most extensive glacial area outside of the Polar Regions, is known as the Asian Water Tower.<sup>6</sup> It is generally assumed that the main sources of atmospheric and glacial pollutants in the Third Pole region are transported from external locations (e.g., South Asia and Middle East) in regional areas.<sup>7-9</sup> Moreover, local pollutant emissions from residential burning activities and vehicles cannot be ignored with the economic development and urbanization on the TP.10-12

Many studies have applied various statistical models, remote sensing, and numerical models to decouple the nonlinear

homes. It provided a valuable opportunity for understanding how variations of urban air pollutants responded to the COVID-19 lockdown in the special highland cities of the third polar. Compared with the global COVID-19 restrictions in 2020, a slightly negative O<sub>3</sub> anomaly and greater PM<sub>2.5</sub> and NO<sub>2</sub> reductions were found. The study showed  $PM_{10}$  (-65%) <  $NO_2$  (-53%) <  $PM_{2.5}$  (-52%) <  $O_3$  (-15%) < CO (-11%) <  $SO_2$ (10%) in the Tibetan Lockdown 2022. The total gaseous oxidant ( $O_x = NO_2 + O_3$ ) decreased 18% in the Tibetan Lockdown 2022, but showed a limited change in the Lockdown 2020. Moreover, the diurnal profiles of NO2 and PM10 disappeared and the diurnal profiles of Ox, PM25, and PM25/CO became weakened. Via the random forest model-based weather normalization technique, we obtained the decoupled meteorological effects  $1.3 \pm 0.6$  times higher than the net emission reduction due to the Tibetan Lockdown 2022. Meanwhile, we found that the deweathered PM<sub>2.5</sub> was mainly from residential combustion emissions due to the weak industrialization in Tibet. Here we roughly estimated that vehiclerelated sources can contribute about 28% and 29% to PM25 and NO2 in urban cities of Tibet respectively. For eco-vulnerability protection and sustainable development in Tibet, more attention should be paid to

lockdown in cities of the Tibetan Plateau\*

Variations of air pollutant response to COVID-19

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Coronavirus Disease 2019 (COVID-19) accidentally appeared in Tibet on August 7, 2022, and broke the 920 consecutive epidemic-free days. The cities in Tibet completely kept lockdown to restrict the public to

reducing the more intensive local pollutant emissions from vehicles and residential combustion of urban cities.

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#### Paper

effects of the meteorological conditions and emissions on the air quality. These studies were the basis of the policies on ecovulnerability protection and regional sustainable development for Tibet.13-15 Short-term interventions on real-world air quality provide a unique opportunity to check and constrain these decoupling models. Stringent emission controls have proven effective for temporary improvement of air quality, such as the 2014 Asia-Pacific Economic Cooperation (APEC) meeting, the 2015 China Victory Day Parade and the 2022 Beijing Winter Olympics,16-18 when control strategies were planned in advance for the main emission sectors.<sup>19</sup> However, Coronavirus Disease 2019 (COVID-19) appeared as a sudden intervention event and has caused disastrous health and socioeconomic crises around the world.20 Meanwhile, diverse policies have been adopted by governments across the world, such as limiting public transportation, encouraging or mandating working from home, and enforcing closure of public services.21,22 Global researchers focused on the air quality change due to lockdown from February to May in 2020, and mostly in the urban agglomerates:<sup>23-25</sup> The total gaseous oxidant ( $O_x = NO_2 + O_3$ ) showed limited change as gains in O3 compensated for drops in NO<sub>2</sub>. Decreasing trends of PM<sub>2.5</sub> were observed in most cities, and short-term and unexpected fluctuations were accounted for the atmospheric oxidation enhancement due to reduced titration.<sup>26-28</sup> To the best of our knowledge, negative ozone anomalies due to COVID-19 lockdown were rare and only reported at two UK sites and a high mountain site in northern Italy.24,25

Atmospheric pressure that decreases with altitude affects lung physiology, so COVID-19 is likely more harmful to the public in Tibet (ave. 4000 m a.s.l.) and needs special consideration.29 Fortunately, lockdown policies throughout China effectively contained the transmission of the COVID-19 epidemic ("Lockdown 2020"), resulting in only one confirmed case in Tibet. An emission assessment report suggested that the Lockdown 2020 has less impact on the local daily lives and industrial production (e.g. energy, raw material processing and manufacturing) in Tibet.30 A telephone interview also reported that the transportation frequency in Tibet was also less affected.31 Studies on two remote high mountains on the TP supported the opinion that the decrease in air pollutant was exclusively associated with the emission reduction in the surrounding areas, such as South Asia.32,33 However, the COVID epidemic suddenly appeared in Tibet on August 7, 2022, and powerful control policies were implemented ("Tibetan Lockdown 2022"). We systematically analyzed and quantified the role of emission reductions and meteorological contribution for the air pollutants, i.e. NO2, O3, sulfur dioxide (SO2), carbon monoxide (CO), PM<sub>2.5</sub>, and PM<sub>10</sub> in Tibetan Lockdown 2022. This provides one chance to understand how air quality is influenced by the local emissions in Tibetan urban cities. Compared with Lockdown 2020, a slight negative ozone anomaly and greater PM2.5 and NO2 reductions were found in Tibetan urban areas due to the current lockdown. The findings provide a reference for future air pollution management and urbanization in Tibet.

## 2 Materials and methods

## 2.1 Data sources and preprocessing

Tibet has a weak industrial base, but is rich in hydropower resources.34 The Tibetan Statistical Department reported that the third industry accounted for 56% of the gross domestic product, much higher than the industrial production (9%) and construction (27%) in 2021. There were 1.34 million urban residents and 41.53 million tourists in 2021. Mining, the major industrial production sector, was located in the counties (i.e. Dazi, Mozhu Gongka, and Zhongba) instead of in the Tibetan urban areas. Water supply, medicine, and cement were mainly for the urban residential consumption (http:// www.xizang.gov.cn/zwgk/zfsj/ndtjgb/). About 5-25% of residential energy consumption was coal, while firewood and dung cake still accounted for 50-60% in Tibet.35 Through literature reviews, vehicle emissions and residential combustion emissions were the main sources for air pollutants in Tibetan urban cities.12,35,36

Fig. S1<sup>†</sup> shows the locations of seven major cities in Tibet, *i.e.* Lhasa, Xigaze, Nyingchi, Qamdo, Nagqu, Ngari, and Lhoka. The concentrations of six criteria air pollutants (*i.e.*, NO<sub>2</sub>, O<sub>3</sub>, SO<sub>2</sub>, CO, PM<sub>2.5</sub>, and PM<sub>10</sub>) were obtained from the China National Environmental Monitoring Center (CNEMC) (https:// air.cnemc.cn:18007/). Only 11 selected sites contained longterm observation data in Tibet. Six of them were further merged with the nearest meteorological observation site in the Integrated Surface Database (ISD) of NOAA (National Oceanic and Atmospheric Administration). These sites were located in the government departments in the urban center. The time resolution of ISD was 3 hours, which was obtained via the "worldmet" R package, including wind speed, wind direction, atmospheric temperature, visibility, dew point, relative humidity, and atmospheric pressure (represented by the sea level pressure). 72 hour back trajectories were calculated for each hour using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, with the NCEP/NCAR reanalysis data as input. The starting height was set as 100 m and then clustered into 6 clusters via the Euclidian distance by "openair" R package.26,37

## 2.2 Tibetan Lockdown 2022

China Central Television (CCTV) and Tibet Satellite TV reported that there was only one confirmed case in Tibet for 920 consecutive days, since the first appearance of COVID-19 in Wuhan. However, COVID-19 accidentally appeared in Tibet on August 7, 2022, from which the confirmed cases increased exponentially. Meanwhile, the migration index decreased sharply, suggesting that the peak of tourism in Tibet came to an abrupt end (Fig. 1) (Baidu Company, https://qianxi.baidu.com/ #/). All major cities implemented lockdown policies before August 14, including the highest epidemic emergency response, community lockdown, and traffic restrictions. The Tibetan government successively cut off all COVID-19 transmission chains in communities from August 26 to 31 in different cities. *Via* effective actions, Tibetan urban cities

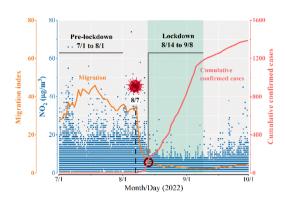


Fig. 1 Temporal variations in the mass concentration of NO<sub>2</sub> at observation sites, cumulative confirmed cases of COVID-19 (data from: National Health Commission of China) and migration index (100 times Baidu's migration scale index, reflecting the population moving into Tibet, https://gianxi.baidu.com/#/).

successively announced success in achieving "dynamic zero-COVID" conditions, and the growth rate of confirmed cases slowed down after September 8. Therefore, the period from July 1 to August 1 was defined as the "pre-lockdown" period and the period from August 14 to September 8 was the "lockdown" period (Fig. 1).

#### 2.3 Random forest model and weather normalization

This study assessed the percentage change  $(P_{2022})$  in the observed air pollution concentrations using eqn (1).

$$P_{2022} = \frac{C_{\rm ld} - C_{\rm pre}}{C_{\rm pre}} \times 100\%$$
(1)

where  $C_{\text{pre}}$  and  $C_{\text{ld}}$  are the average concentrations in the pre-/lockdown period, respectively.

Due to the influence of the Indian monsoon, the usual seasonal variation of aerosols in summer on the Tibetan Plateau cannot be ignored.<sup>7,8</sup> The percentage change  $P_{2019-2021}$  was used to build "business-as-usual" conditions (BAU) according to the same periods in 2019–2021. So the net percentage change  $P^*$  (*i.e.*, the change in air pollutant concentration arising from lockdown effects alone) was given as eqn (2).<sup>24,26</sup>

$$P^* = P_{2022} - P_{2019-2021} \tag{2}$$

The "rmweather" R package is a random forest model-based weather normalization technique (RF-deweather). Many studies applied it to investigate the variations of air pollutant response to the COVID-19 lockdown.<sup>26,38,39</sup> It is nonparametric with fewer limits from assumptions, including sample normality, independence, homoscedasticity, and adherence to other strict parametric assumptions.<sup>23,37,40</sup> 80% of original data were randomly selected to build the model and the other was used to evaluate the accuracy of the model. Through literature reviews, input factors for the model included time variables (*i.e.*, Unix time, Julian day, day of the week, and hour of the day), meteorological data and trajectory clusters in "Data Sources and Preprocessing".<sup>26,41,42</sup> The initial parameters were as follows: 300

decision trees (n\_trees = 300) and 1000 times to sample the database (n\_samples = 1000), and others were default in the "rmweather" R package. The statistics of the deweathered result and original data were based on the "openair" R package.<sup>43,44</sup> The average correlation coefficients between predicted values and observation values was 0.91. Details are in Table S1.<sup>†</sup>

*Via* RF-deweather, the percentage change, *P*, could be divided into two parts, meteorological effect ( $P_{met}$ ) and anthropogenic effect ( $P_{ant}$ ) as eqn (3).<sup>45</sup>

$$\begin{cases}
P_{\text{met}} = \frac{(C_{\text{ld,obs}} - C_{\text{ld,dew}}) - (C_{\text{pre,obs}} - C_{\text{pre,dew}})}{C_{\text{pre,obs}}} \times 100\% \\
P_{\text{ant}} = \frac{C_{\text{ld,dew}} - C_{\text{pre,dew}}}{C_{\text{pre,obs}}} \times 100\% \\
P_{\text{met}} + P_{\text{ant}} = P_{2022}
\end{cases}$$
(3)

where the subscripts "obs" and "dew" were further used to distinguish the results between observation and the deweathered. Meteorological conditions affect the air quality, positively or negatively, and the  $P_{\text{met}}$  is the net difference of  $(C_{\text{obs}} - C_{\text{dew}})$ between lockdown and pre-lockdown periods. The anthropogenic effect was acknowledged as the net effects of the local anthropogenic emission change.45,46 According to the emission scenarios in Lockdown 2020, vehicle-related emissions and industry emissions were controlled most.19 The sources of vehicle-related emissions included on-road vehicles and some off-road engines, i.e. agricultural or industrial equipment, locomotives and commercial generators.<sup>47</sup> SO<sub>2</sub> emitted mainly from coal burning.<sup>22,48</sup> Construction activities and mechanical grinding could emit coarse man-made fugitive dust.<sup>49</sup> Especially for NO<sub>2</sub>, O<sub>3</sub>, PM<sub>2.5</sub>, and CO, the change of the deweathered concentrations roughly represented the contribution of the vehicle-related emissions (Pveh) in Tibetan urban cities as eqn (4).41

$$P_{\rm veh} = \frac{C_{\rm pre,dew} - C_{\rm ld,dew}}{C_{\rm pre,dew}} \times 100\%$$
(4)

## 3 Results and discussion

#### 3.1 Overview of air pollutants and meteorological conditions

From July 1 to October 1, 2022, the daily average mass concentrations of NO<sub>2</sub>, O<sub>3</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, and CO were 6.1, 106.9, 4.2, 9.7, 7.3 and 349  $\mu$ g m<sup>-3</sup>, respectively, and all met the Ambient Air Quality Standards I in China (GB 3095—2012). 8 hour daily maximum O<sub>3</sub> (MDA8 O<sub>3</sub>) was 100.5, from 34 to 167  $\mu$ g m<sup>-3</sup>, up to the WHO Air Quality Guideline (AQG, 100  $\mu$ g m<sup>-3</sup>). So ozone is a non-negligible long term health risk substance to the public in Tibet.

Pre-lockdown 2022 and July to September in 2019, 2020, and 2021 could represent the usual summer conditions in Tibetan urban cities. Fig. 2 shows the diurnal variations of six air pollutants for an average of 11 sites. Similar results are summarized in Fig. S2† for the same periods in 2019, 2020 and 2021. Generally, the hourly concentrations of  $NO_2$ , CO,  $PM_{2.5}$ , and  $PM_{10}$  displayed a bimodal distribution in the usual

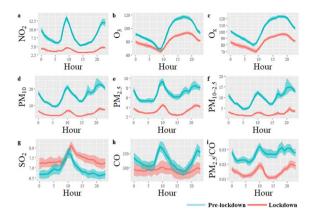


Fig. 2 Diurnal variations in air pollutants during the pre-lockdown and lockdown in 2022. Units:  $\mu g m^{-3}$  for NO<sub>2</sub> (a), O<sub>3</sub> (b), total gaseous oxidant (O<sub>x</sub> = NO<sub>2</sub> + O<sub>3</sub>) (c), PM<sub>10</sub> (d), PM<sub>2.5</sub> (e), coarse particles (PM<sub>10-2.5</sub> = PM<sub>10</sub>-PM<sub>2.5</sub>) (f), SO<sub>2</sub> (g) and CO (h); dimensionless for the rate of PM<sub>2.5</sub>/CO (i). Error bars represent the 95% confidence interval of the hourly mean *via* "openair" R-package.

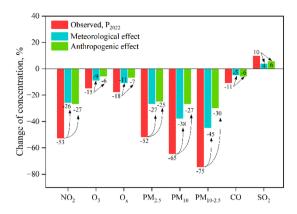
summer, with two peaks from 9:00 to 10:00 and 21:00 to 22:00 (Beijing time), and minima from 5:00 to 7:00 and 14:00 to 17:00, respectively. SO<sub>2</sub> only had a peak at 10:00. The diurnal variance of  $O_3$  was opposite and unimodal with a peak at 17:00. The result was similar to the typical diurnal cycle mode of six criteria air pollutants in Chinese plain cities. It was mainly ascribed to elevated traffic emissions in rush hours, and the combined effect from a raised boundary layer height and photochemical oxidation reactions.<sup>15,50,51</sup> The first valley point of NO<sub>2</sub> and the peak point of  $O_3$  were at about 6:00 and 20:00, respectively, when sun usually rose and set in summer. During the study periods, the variations of meteorological factors were limited, and the water vapor content in the air tended to slightly increase: RH and dew point increased, and air temperature slightly decreased during the lockdown (Fig. S3†).

## 3.2 Air quality change during the lockdown

According to the BAU conditions,  $P_{2022}$  was equivalent to  $P^*$  for all sites, suggesting that general changes of air pollutants cannot be fully attributable to the seasonal variation of meteorology (Fig. S4†). Fig. 3 shows that the mean change rates ( $P_{2022}$ ) were PM<sub>10</sub> (-65%) < NO<sub>2</sub> (-53%) < PM<sub>2.5</sub> (-52%) < O<sub>3</sub> (-15%) < CO (-11%) < SO<sub>2</sub> (10%) at Tibetan urban cities. Particulate matter and NO<sub>2</sub> both displayed the largest reduction due to the lockdown. Fig. 3 shows the meteorological effect ( $P_{met}$ ) and anthropogenic effect ( $P_{ant}$ ), and Table 1 lists the dewearthered concentrations and estimated  $P_{veh}$ .

**3.2.1 SO**<sub>2</sub> **and CO.** Our study shows that SO<sub>2</sub> slightly increased from 6.3 to 6.9  $\mu$ g m<sup>-3</sup>, mainly in the afternoon (Fig. 2g), and two peaks of CO almost disappeared and its diurnal profile had a 95% confidence interval (Fig. 2h). The average changes were small, about -11% and 10% for CO and SO<sub>2</sub>, respectively (Fig. 3). The variations were consistent with those (about ±20%) in other cities in East China during the COVID lockdown periods.<sup>52</sup> The reason for SO<sub>2</sub> increase was usually attributed to the power plants and raw material

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**Fig. 3** Change rate of concentrations due to the Tibetan Lockdown 2022, and results of the random forest model-based weather normalization, including assigned anthropogenic effects and meteorological effects.

Table 1 Dewearthered concentration and contribution of vehicle-related emission ( $P_{\rm veh}$ ) via random forest model-based weather normalization

| Concentration, $\mu g m^{-3}$ |      | СО  | $NO_2$ | $O_3$ | O <sub>x</sub> | PM <sub>2.5</sub> |
|-------------------------------|------|-----|--------|-------|----------------|-------------------|
| Pre-lockdown                  | Mean | 380 | 6.8    | 84    | 90             | 6.2               |
|                               | Std. | 150 | 2.7    | 16    | 18             | 1.2               |
| Lockdown                      | Mean | 360 | 4.8    | 78    | 83             | 4.5               |
|                               | Std. | 150 | 1.8    | 15    | 17             | 0.9               |
| Average $P_{\text{veh}}$ , %  | 6    | 29  | 7      | 8     | 28             |                   |

industry, as the main contributors to SO<sub>2</sub> emissions. They must operate for the electricity consumption of households and have uninterrupted industrial processes. Moreover, traffic reduction induced the disappearance of CO peaks from 370 to 300 µg m<sup>-3</sup> in rush hours and decreased CO from 320 to 290 µg m<sup>-3</sup> in nonrush hours in Tibetan urban areas (Fig. 2h). It was reported that household stoves in Tibet also emitted CO when burning firewood and dung cake, which could contribute 0.4–0.5 Mt, five times the transportation emission (~77 Kt) by the latest assessment.<sup>35,47</sup> After the RF-deweather process, we roughly estimated CO sources accounting for ~6% from vehicle-related emissions ( $P_{\rm veh}$ ), suggesting that residential combustion emissions dominated for CO in Tibetan cities (Table 1).

**3.2.2** NO<sub>2</sub>. The vehicle-related emission was the major source of NO<sub>2</sub> in cities.<sup>42,53</sup> According to the Multi-resolution Emission Inventory for China (MEIC), the vehicle-related emission accounted for 85% of total nitrogen oxide (NO<sub>x</sub> = NO + NO<sub>2</sub>) emission in Tibet, twice higher than the national average level at  $36.5 \pm 12.2\%$ .<sup>47</sup> During the Tibetan Lockdown 2022, an approximately 53% decrease rate of NO<sub>2</sub> in Tibet was observed. The result was higher than 42% during the Lockdown 2020 (Fig. 3 and 4). The transportation frequency in Tibet was less affected in Lockdown 2020.<sup>31</sup> The range of the NO<sub>2</sub> decrease rate around the world was only up to -50% in some severely affected cities (*e.g.*, Wuhan and Milan) due to the lockdown.<sup>23</sup> Fig. 2a shows that the diurnal fluctuation of NO<sub>2</sub> basically disappears and smoothly ranged from 3 to 5 µg m<sup>-3</sup>. The

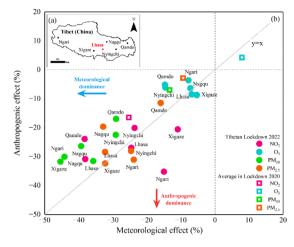


Fig. 4 Anthropogenic effects and meteorological effects on changes of air pollutants in Tibetan Lockdown 2022 for six cities. (a) Locations of the studied Tibetan cities. Lhoka was not included, because meteorological data were not accessible. (b) Open squares were the average changes for the Lockdown 2020, via the random forest model-based weather normalization.

diurnal profile was similar to the observation at the background site in the Tibetan Plateau (4.4  $\pm$  2.7  $\mu$ g m<sup>-3</sup>).<sup>54</sup> Table 1 shows that NO<sub>2</sub> sources accounted for  $\sim$ 29% from vehicle-related emissions (Pveh), via RF-deweather. These parts of emissions almost disappeared due to the lockdown and led to a -27%decrease in the observed concentration. The others were attributed to the conducive meteorological conditions, and Pmet was -26% (Fig. 3). We noticed  $[1 - P_{veh}]$  up to  $\sim 71\%$  and deweathered concentrations deceasing from 6.8 to 4.8  $\mu g \ m^{-3}$ due to the lockdown. The result reflected that the residential combustion emissions remained the high contribution in Tibetan cities during the lockdown.

3.2.3 Surface  $O_3$ . Surface  $O_3$  is a secondary pollutant produced by photochemical oxidation of volatile organic compounds (VOCs) and carbon monoxide (CO) in the presence of  $NO_x$ . In the presence of sunlight,  $O_3$  is chemically produced from the oxidation of VOCs in the presence of NO<sub>x</sub>. On the other hand, in the process of NO<sub>x</sub> titration, freshly emitted NO rapidly reacts with O<sub>3</sub> to form NO<sub>2</sub> (titration reaction). It was acknowledged that the surface O3 in Tibet could be affected more by natural sources (including biogenic sources, lightning and soil), stratospheric intrusion, and long-range transport.13,14,55 For example, Yin et al. (2017) proposed that stratospheric intrusion contributed 18.2  $\pm$  2.6% of the ozone variance at a Tibetan remote site, 21.10% in spring but only 0.41% in Autumn.<sup>56</sup> Tibetan Lockdown 2022 is an opportunity to generally evaluate the role of local anthropogenic pollutants in surface O<sub>3</sub>. Fig. 3 shows the weak negative ozone anomalies (about -15%) in Tibet due to the lockdown. Via RF-deweather, anthropogenic contributions accounted for  ${\sim}7\%$  from vehiclerelated emissions  $(P_{\text{veh}})$  in Table 1.

The diurnal profile did show decreasing trends of  $O_3$  and  $O_x$ (Fig. 2b and c). Before the lockdown, the pattern of daytime O<sub>3</sub> was similar to the typical parabola pattern and had a higher

range from the peak to the valley point. It was similar to the condition driven by the atmospheric photochemistry in polluted plain areas,<sup>15,51</sup> and different from the smooth O<sub>3</sub> profiles at the remote sites on the TP.54,56,57 NOr from local vehicle-related emission was likely more sufficient than VOCs from 6:00 to 10:00 in the usual summer morning, making a strong titration reaction, and  $O_x$  remained at about 80 µg m<sup>-3</sup> (Fig. 2c). During the lockdown, the  $O_3$  and  $O_r$  concentrations both decreased significantly, and the maximum declines were from 11:00 to 20:00, when NOx was no longer sufficient, resulting in a NO<sub>r</sub>-limited regime for surface O<sub>3</sub>. In addition, a broader platform-like diurnal profile of surface O3 was observed after sunrise, followed by a shorter climb (Fig. 3b and c). These were different from the same periods in 2019-2021 (Fig. S2<sup>†</sup>) and similar to observations in TP remote sites.<sup>56,57</sup> Therefore, we can conclude that O<sub>3</sub> in urban areas on the TP is strongly related to NO<sub>x</sub> anthropogenic emission and regional O<sub>3</sub> is mainly contributed by stratospheric intrusion and long-range transport.

3.2.4 PM<sub>10</sub> and PM<sub>2.5</sub>. The local epidemic in Tibet had a great impact on atmospheric particles. PM<sub>10</sub> and PM<sub>2.5</sub> both decreased up to -50%, due to the lockdown. Their decrease rates in Lockdown 2020 averaged -20% in China.27,28,58 PM10-2.5 was mainly dominated by man-made fugitive dust from construction activities, mechanical grinding and roads, natural dust from the ground, and biogenic sources.49 Fig. 3 shows that observed PM<sub>10-2.5</sub> decreased 75%, much higher than -52% in PM<sub>2.5</sub>. The machine learning method further calculated meteorological effects accounting for 45%, 38% and 27% of the decreases of PM<sub>10-2.5</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>, respectively. The anthropogenic effects were similar, 25–30% (Fig. 3). PM<sub>2.5</sub>/CO is a good tool to show the formation rate of secondary aerosols.26,27 We noticed that the diurnal variation almost disappeared for PM<sub>10</sub> and primary PM<sub>10-2.5</sub> during the lockdown, and the diurnal patterns of PM2.5 and PM2.5/CO just weakened but remained bimodal (Fig. 2). It suggested that the reduction of atmospheric oxidation capacity  $(O_x)$  weakened the formation processes of secondary aerosol in Tibetan urban cities due to the Tibetan Lockdown 2022. In Tibet, 38% of the primary PM<sub>2.5</sub> emission was set as the vehicle-related contribution in the MEIC.47 Zhao et al. (2022) reported that vehicle-related organic aerosol accounted for ~14% in fine particles in Lhasa.<sup>36</sup> In Table 1, we roughly estimated that 28% of PM<sub>2.5</sub> emissions were assigned to vehicle-related emissions. Our studies supplied a reference benchmark scene for the emission inventory update and revision: the emissions from residential combustion emissions (~72%) could still dominate the primary  $PM_{2.5}$  in Tibetan urban cities, and deweathered PM<sub>2.5</sub> concentrations varied from 6.2 to 4.5  $\mu$ g m<sup>-3</sup> in the studied cities with negligible traffic flux due to the lockdown.

#### 3.3 Comparison between lockdowns 2022 and 2020

During the Tibetan Lockdown 2022, meteorological conditions were conducive to the removal of air pollutants, *i.e.* PM<sub>10</sub>  $(-38\%) < PM_{2.5} (-27\%) < NO_2 (-26\%) < O_3 (-9\%)$  (Fig. 3). Different Tibetan cities had similar trends, and meteorological

effects were slightly larger (1.3  $\pm$  0.6) than anthropogenic effects (Fig. 4). Only PM<sub>2.5</sub> and NO<sub>2</sub> in a few cities were dominated by anthropogenic effects, such as NO<sub>2</sub> in Ngari and Xigaze cities. The result is likely related to the type and total number of local motor vehicles in different cities.

Tibet had a lockdown from January 26 to February 19 in 2020 and a pre-lockdown from January 1 to 20 in 2020.32,33 We found that anthropogenic effects were more important for the NO<sub>2</sub> decrease in 2022 than Lockdown 2020 (Fig. 4). PM<sub>2.5</sub> and PM<sub>10</sub> both decreased more in Tibetan Lockdown 2022, under more stringent traffic control and more conducive weather (Fig. 4). The changes of O<sub>3</sub> in 2022 and 2020 were in the opposite phase as shown in Fig. 4. Compared with 2022, the diurnal profiles of six criteria air pollutants and indices (O<sub>x</sub>, PM<sub>2.5</sub>/CO, and PM<sub>10-</sub>  $_{2.5}$ ) only weakened slightly in 2020, and smooth diurnal trends were not found (Fig. S5<sup>†</sup>). The result suggested that the pollutant reductions were much larger and then weakened the atmospheric oxidizing capacity during the Tibetan Lockdown 2022. Importantly,  $O_3$  compensated for the drops in  $NO_2$  (about 10  $\mu$ g m<sup>-3</sup>), and hourly O<sub>x</sub> rarely changed in 2020 (Fig. S5<sup>†</sup>). It suggested that decreases of NO<sub>x</sub> were not enough and NO<sub>x</sub> was still sufficient relative to VOCs in the Lockdown 2020. The titration reaction of NO and O3 was probably depressed, and thus promoted the elevation of the O<sub>3</sub> concentration, especially in the night. The net effects on the total oxidant  $(O_r = NO_2 + O_3)$ were limited. Other researchers reported similar results in Tibet during the Lockdown 2020 and estimated that O<sub>3</sub> increased about 20%.<sup>39,52</sup> Global studies also reported that the gains in O<sub>3</sub> compensated for drops in NO2 due to the Lockdown 2020.23,52 The negative ozone anomalies due to the Lockdown 2020 were rarely reported and only in Aberdeen city, UK ( $-36 \pm 8.6\%$ ), and a mountain site near urban areas in Italy (-16%).<sup>24,25</sup> They proposed that great NO<sub>2</sub> reductions in urban areas could decrease surface O<sub>3</sub> under a NO<sub>x</sub>-limited regime and even affect nearby mountains.<sup>24,25</sup> In Tibetan urban cities, the greater NO<sub>2</sub> reductions (-53%) also led to a negative ozone anomaly (-15%)(Fig. 3), as the COVID lockdown policy was more stringent in 2022 than those in Lockdown 2020. Considering the max hourly concentration of  $O_3$ , MDA8  $O_3$  decreased up to -20%, from 116 to 93  $\mu$ g m<sup>-3</sup>, and it met the AQG of WHO (Fig. S4<sup>†</sup>). Therefore, vehicle-related emission control was a potential way to reduce the public health risk from O<sub>3</sub> in Tibetan urban areas.

## 4 Conclusions

The sudden appearance of COVID on August 7, 2022, broke the 920-consecutive-day safe condition in Tibet. The rapid decline of  $NO_2$  possibly led to a  $NO_x$ -limited regime for surface  $O_3$ , in Tibetan urban areas, and a net negative anomaly of surface  $O_3$  was for the first time found followed by larger decreases of  $PM_{10}$  and  $PM_{2.5}$ . This was different from the air quality change in Lockdown 2020, when air pollutant declines were not large enough to offset the  $O_3$  enhancement by the weakened titration reaction in Tibet. Our study conducted diurnal variation analysis using "business-as-usual" analysis and random forest model-based weather normalization analysis during the lockdown 2022 in Tibet. Differences between lockdowns 2020 and

2022 did provide clear evidence that local anthropogenic emissions influenced the atmospheric environment in Tibetan cities, especially NO<sub>2</sub>,  $O_3$ , PM<sub>2.5</sub>, and PM<sub>10</sub>.

In summer, we proposed that the surface O<sub>3</sub> concentrations become smaller than 100  $\mu$ g m<sup>-3</sup> (the AQG of WHO) by reducing the vehicle-related emission and controlling NO<sub>2</sub> at 2.5–5  $\mu$ g m<sup>-3</sup>. For local anthropogenic emissions, the contributions of vehicle-related emissions were estimated via RF-deweather: NO2 (29%) > PM<sub>2.5</sub> (28%) > O<sub>3</sub> (7%) > CO (6%). The other emissions were mainly assigned to residential combustion emissions due to weak industrialization in Tibetan cities. For observation concentrations, meteorological effects were more conducive to the changes of air pollutants, and were  $1.3 \pm 0.6$  times higher than the net effects from emission changes during the Tibetan Lockdown 2022. This study serves as a general reference for future air pollution control in Tibetan cities. We should point that the monitoring sites are fewer in Tibet than those in Chinese plain cities, which might lead to certain uncertainty in this study. In the future, the construction of monitoring networks and detailed source apportionments could provide more accurate references for air pollution management and urbanization in Tibet. Meanwhile, the advances of public transport, railway transport, and new energy vehicle plan will significantly improve urban air quality through a series of great projects in the future. More attention should also be paid to reducing the more intensive local pollutant emissions from residential combustion emissions and vehicles in Tibetan urban cities.

## Author contributions

Xiyao Chen: writing – original draft, methodology, and visualization. Fan Zhang: methodology and writing – review & editing. Dianguo Zhang: resources and funding acquisition. Liang Xu: data curation and resources. Rui Liu: data curation and resources. Xiaomi Teng: data curation and resources. Xin Zhang: methodology and writing – review & editing. Shuo Wang: data curation and resources. Weijun Li: conceptualization, supervision, and funding acquisition.

## Conflicts of interest

There are no conflicts to declare.

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