Spatial and temporal changes of the ozone sensitivity in China based on satellite and ground-based observations

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Abstract. Ground-level ozone (O₃) pollution has been steadily getting worse in most parts of eastern China during the past 5 years. The non-linearity of O₃ formation with its precursors like nitrogen oxides (NOₓ = NO + NO₂) and volatile organic compounds (VOCs) are complicating effective O₃ abatement plans. The diagnosis from space-based observations, i.e. the ratio of formaldehyde (HCHO) columns to tropospheric NO₂ columns (HCHO/NO₂), has previously been proved to be highly consistent with our current understanding of surface O₃ chemistry. HCHO/NO₂ ratio thresholds distinguishing O₃ formation sensitivity depend on regions and O₃ chemistry interactions with aerosol. To shed more light on the current O₃ formation sensitivity over China, we have derived HCHO/NO₂ ratio thresholds by directly connecting satellite-based HCHO/NO₂ observations and ground-based O₃ measurements over the major Chinese cities in this study. We find that a VOC-limited regime occurs for HCHO/NO₂ < 2.3, and a NOₓ-limited regime occurs for HCHO/NO₂ > 4.2. The HCHO/NO₂ between 2.3 and 4.2 reflects the transition between the two regimes. Our method shows that the O₃ formation sensitivity tends to be VOC-limited over urban areas and NOₓ-limited over rural and remote areas in China. We find that there is a shift in some cities from the VOC-limited regime to the transitional regime that is associated with a rapid drop in anthropogenic NOₓ emissions and VOC emissions controls is essential for O₃ abatement plans.

1 Introduction

Ground-level ozone (O₃) is one of the major air pollutants that has negative impacts on human health and can result in eye and nose irritation, respiratory disease, and lung function impairment (Jerrett et al., 2009; Khaniabadi et al., 2017; Huang et al., 2018). Y. Tian et al. (2020) observed increased admissions for pneumonia associated with O₃ exposure, especially for elderly people. In addition, it also has important impacts on climate as a greenhouse gas by absorbing thermal radiation (Fishman et al., 1979; IPCC, 2014). Photochemical tropospheric O₃ is formed in a non-linear manner from O₃ precursors such as volatile organic compounds (VOCs) and nitrogen oxides (NOₓ = NO + NO₂) in the presence of sunlight (Crutzen, 1974; Jacob, 2000).

In 2008, China was found to be the largest contributor to Asian emissions of carbon monoxide (CO), NOₓ, non-methane volatile organic carbon (NMVOC), and methane (CH₄) (Kurokawa et al., 2013). Because of these large emissions of anthropogenic air pollutants, the Chinese State Council released the “Air Pollution Prevention and Action Plan” (APPAP) on September 2013, which has as a key task to prevent and control air pollution in China (Cai et al., 2017). Since then, critical emission control strategies have been carried out that are designed to reduce the concentrations of six environmental pollutants: sulfur dioxide (SO₂), nitrogen dioxide (NO₂), CO, O₃, and particulate mat-
ter (PM$_{2.5}$ and PM$_{10}$) (Zhang et al., 2016; Feng and Liao, 2016). During the past decade, the concentrations of many pollutants including SO$_2$, NO$_2$, CO, PM$_{2.5}$, and PM$_{10}$ have declined in most cities; however, O$_3$ concentrations showed an increasing trend (W. N. Wang et al., 2017; Z. Wang et al., 2019; Zeng et al., 2019). Therefore, reducing O$_3$ concentrations has become the focus of China’s next air quality control strategy (Cheng et al., 2018).

In terms of O$_3$ concentrations, the effectiveness of emissions control strategy depends on whether the photochemical regime of O$_3$ formation is a VOC-limited or NO$_x$-limited regime (Jin et al., 2020). In the VOC-limited (or NO$_x$-saturated) regime, VOC emission reductions reduce the chemical production of organic radicals (RO$_2$) which in turn lead to decreased cycling with NO$_x$ and consequently lower concentration of O$_3$ (Milford et al., 1989). In the NO$_x$-limited (or VOC-saturated) regime, NO$_x$ emission reductions reduce NO$_2$ photolysis, which is the primary source of free oxygen atoms. Therefore, in a NO$_x$-limited regime, NO$_x$ reductions reduce ambient O$_3$. In contrast, in a VOC-limited regime, NO$_x$ acts to reduce O$_3$ so a NO$_x$ decrease in emissions promotes O$_3$ production (Kleinman, 1994).

The observed photochemical indicators and observation-based models (OBMs) are the most commonly used tools to diagnose the O$_3$ formation sensitivity. O$_3$ production efficiency (OPE = $\Delta$O$_3$ / $\Delta$NO$_2$) and the H$_2$O$_2$ / NO$_2$ (or H$_2$O$_2$ / HNO$_3$) ratio are two widely used indicators to infer the O$_3$ formation regimes (Chou et al., 2011; Ding et al., 2013). T. Wang et al. (2017) concluded that lower OPE values (e.g. < 4) indicate a VOC-limited regime. In contrast, higher OPE values (e.g. > 7) indicate a NO$_x$-limited regime. OPE values in the medium range (e.g. 4 < OPE < 7) mark the transition between the two regimes. Another indicator of the O$_3$ formation sensitivity regime is the H$_2$O$_2$ / NO$_2$ ratio. Hammer et al. (2002) defined that, in the VOC-limited regime, lower H$_2$O$_2$ / NO$_2$ ratios would be expected and higher H$_2$O$_2$ / NO$_2$ ratios indicate the NO$_x$-limited regime. In the past decade, the observed photochemical indicators have been applied to identify the O$_3$ formation sensitivity in different periods and regions of China.

The OBM combines in situ field observations and chemical box modelling. It is built on widely used chemistry mechanisms (e.g. Master Chemical Mechanism (MCM), Carbon Bond, Regional Atmospheric Chemical Mechanism (RACM), Statewide Air Pollution Research Center mechanism (SAPRC)) and applied to the observed atmospheric conditions to simulate various atmospheric chemical processes, including the in situ O$_3$ production rate. However, ground-based measurements are often limited in time period and spatial extent. The OBM analysis requires measuring nitric oxide (NO) at sub-ppt levels and more than 50 different types of VOCs with high accuracy, which is difficult to achieve (T. Wang et al., 2017).

Satellite remote sensing provides an alternative way to investigate long time periods of O$_3$ formation sensitivity on large spatial scales. For over 2 decades, satellite-based spectrometers have provided continuous global observations on a daily basis for two species indicative of O$_3$ precursors, i.e. NO$_2$ for NO$_x$ (Martin et al., 2004; Lamsal et al., 2014) and formaldehyde (HCHO) for VOCs (Palmer et al., 2003; Fu et al., 2007). NO$_x$ can be approximated from satellite observations of NO$_2$ column because of the short lifetime of NO$_2$ and high ratio of NO$_2$ / NO$_x$ in the boundary layer (Duncan et al., 2010; Jin and Holloway, 2015). HCHO is an intermediate of the oxidation reaction of various VOCs in the atmosphere. The production of HCHO is approximately proportional to the summed rate of reactions of VOC with OH radicals (Sillman, 1995). Therefore, HCHO can be used as a tracer for VOCs in the absence of other VOC observations (Martin et al., 2004; Duncan et al., 2010). The O$_3$ formation sensitivity is defined by the ratio of HCHO to NO$_2$ (referred to as FNR) (Martin et al., 2004). Duncan et al. (2010) combined models and Ozone Monitoring Instrument (OMI) HCHO and NO$_2$ data to show certain ranges of FNR that can be useful for classifying a region into VOC-limited or NO$_x$-limited regime. An FNR smaller than 1 indicates the VOC-limited conditions, and an FNR higher than 2 indicates the NO$_x$-limited conditions. An FNR in the range of 1–2 should generally be considered indicative of the transitional regime. These FNR thresholds defined by Duncan et al. (2010) have been widely used for various regions (Choi and Souri, 2015; Jin and Holloway, 2015; Souri et al., 2017; Jeon et al., 2018) and with different satellite instruments (Choi et al., 2012).

However, these prior studies linked FNR with surface O$_3$ sensitivity in models (Martin et al., 2004; Duncan et al., 2010). Modelled and observed HCHO columns, NO$_2$ columns, and surface O$_3$ often disagree. Jin et al. (2017) found that the spatial and temporal correlations between the modelled and satellite-derived FNR vary over the used satellite instruments. Brown-Steiner et al. (2015) found persistent O$_3$ biases under all configurations of a global climate–chemistry model (GCCC) with detailed tropospheric chemistry. Although FNR thresholds defined by Duncan et al. (2010) have been used previously to investigate O$_3$-NO$_x$- VOC sensitivity in China (Witte et al., 2011; Tang et al., 2012; Jin and Holloway, 2015), their conclusions were based on the atmospheric situations in the United States and may not be suitable for the more complicated air pollution in China, concerning the different emission factors, sources, pollution levels, and climatology. For example, compared with the United States, most cities in China have higher aerosol levels (van Donkelaar et al., 2010; X. Li et al., 2019). Secondary aerosol production may become a large sink of radicals, which could shift O$_3$ production toward a VOC-limited regime under these FNR thresholds suited to the United States (Liu et al., 2012; K. Li et al., 2019). It is therefore useful to describe surface O$_3$ sensitivity using FNR thresholds derived entirely from satellite-observed FNR and ground-based measurements of O$_3$. In addition, Schroeder et al. (2017) using airborne measurements suggested that the
range and span of FNR marking the transitional regime varies regionally.

In this study, we assess whether space-based HCHO/NO$_2$ ratios capture the non-linearity of O$_3$ chemistry by matching satellite observations with ground-based O$_3$ measurements over major Chinese cities. Thresholds suited for China between space-based HCHO/NO$_2$ and the ground-based O$_3$ response patterns are derived from observations instead of model results. We focus on the spatial and temporal variability of O$_3$ formation sensitivity using our FNR thresholds on a nationwide scale and in typical cities from 2016 to 2019.

More recently, a new unique situation has occurred with the outbreak of the COVID-19 pandemic, which provided a unique opportunity to demonstrate our predicted effects on O$_3$ pollution in China. Efforts to halt the spread of COVID-19 have drastically reduced human activities worldwide (Siciliano et al., 2020; H. Tian et al., 2020). As a result of these restrictions, a significant reduction in primary air pollutant emissions, especially in the concentration of NO$_2$, has been noticed in China and several European and American countries (Tobias et al., 2020; Wang and Su, 2020; Bawens et al., 2020; Ding et al., 2020). By contrast, increasing O$_3$ concentrations during the same period were observed in densely populated metropolitan areas throughout the world (Siciliano et al., 2020; Zoran et al., 2020; Huang et al., 2020).

Section 2 describes the data and methods used in this study. Section 3 presents our derived FNR thresholds method and variations of O$_3$ formation sensitivity in China. In addition, impacts of the COVID-19 outbreak on O$_3$ levels are discussed. Finally, Sect. 4 gives a brief summary.

2 Data

2.1 Satellite data

We use the NO$_2$ and HCHO observations from the Ozone Monitoring Instrument (OMI) aboard the National Aeronautics and Space Administration (NASA) satellite Aura, which was launched in July 2004 (Levelt et al., 2006). In an ascending sun-synchronous polar orbit, OMI passes the Equator at about 13:40 LT (local time), providing global measurements of aerosol parameters, cloud, and various trace gases (NO$_2$ and HCHO among them) (Levelt et al., 2006). The high spatial resolution (13 km $\times$ 24 km at nadir) allows for observing fine details of atmospheric parameters (Jin and Holloway, 2015). OMI data are considered to be reliable and of good quality for the full mission thus far (Zara et al., 2018). In addition, the OMI overpass time is well suited to detect the O$_3$ formation sensitivity during the afternoon, when O$_3$ photo-chemical production peaks and when the boundary layer is high and the solar zenith angle is small, maximizing instrument sensitivity to HCHO and NO$_2$ in the lower troposphere (Jin et al., 2017).

We use the OMI tropospheric NO$_2$ and HCHO data products from the European Quality Assurance for Essential Climate Variables project (QA4ECV, http://www.qa4ecv.eu/, last access: 6 May 2021). NO$_2$ data are compiled by the Royal Netherlands Meteorological Institute (KNMI). The tropospheric NO$_2$ column density is defined as the vertically integrated number of NO$_2$ molecules between the Earth’s surface and the tropopause per unit area. We select QA4ECV NO$_2$ daily observations following the recommendations given in the product specification document (Boersma et al., 2011) for this data product: (1) no processing error, (2) less than 10% snow or ice coverage, (3) solar zenith angle less than 80°, and (4) cloud radiance fraction less than 50%. The QA4ECV NO$_2$ monthly datasets are processed with a spatial resolution of 0.125° $\times$ 0.125°. Boersma et al. (2018) reported the single-pixel uncertainties for the QA4ECV NO$_2$ columns are 35%–45% in the polluted regions; the monthly mean NO$_2$ columns are estimated to have an uncertainty of ±10%.

The OMI tropospheric HCHO data are retrieved by the Belgian Institute for Space Aeronomy (BIRA-IASB) (Smedt et al., 2017a). We select processing_quality_flags = 0 or > 255, providing a selection of observations that is considered optimal. Zara et al. (2018) found that the QA4ECV HCHO slant column densities (SCDs) have uncertainties of 8–12 $\times$ 10$^{15}$ molecule/cm$^2$ and a remarkably stable trend (increase < 1%/yr). The QA4ECV HCHO monthly datasets are available with a spatial resolution of 0.05° $\times$ 0.05°. Temporal averaging has been shown to reduce the HCHO measurements uncertainty and noise (Millet et al., 2008). We regrid the monthly OMI HCHO data (0.05° $\times$ 0.05°) to the same grid as for the monthly OMI NO$_2$ data (0.125° $\times$ 0.125°).

2.2 NO$_x$ emission

Emission inventories of air pollutants are important sources of information for policy makers and form essential input for air quality models. Bottom-up inventories are usually compiled from statistics on emitting activities and their typical emission factors but are sporadically updated (Li et al., 2017). Satellite-derived emission inventories have important advantages over bottom-up emission inventories: they are spatially consistent, have high temporal resolution, and provide up-to-date emission information (Mijling and van der A, 2012). In this study, we use monthly mean NO$_x$ surface emission estimates derived from OMI observations of tropospheric NO$_2$ columns (the QA4ECV product discussed in Sect. 2) by the Daily Emission estimation Constrained by Satellite Observations (DECSO) algorithm. Mijling and van der A (2012) for the first time developed DECSO (version 1) by calculating the sensitivity of concentration to emission based on a chemical transport model and using trajectory analysis to account for transport away from the source. Ding et al. (2015) improved DECSO (version 3) and demonstrated that it is able to detect the monthly change of NO$_x$ emis-
sions due to air quality regulations on a city level. The NO\textsubscript{x} emissions derived by the improved DECSO version 5 are in good agreement with other bottom-up anthropogenic emission inventories. In addition, the improved algorithm is able to better capture the seasonality of NO\textsubscript{x} emissions. The precision of monthly NO\textsubscript{x} emissions derived by DECSO version 5 for each grid cell is about 20\% (Ding et al., 2017).

Here, we use NO\textsubscript{x} emissions derived by the latest DECSO version 5.1a which provides monthly emissions for the last decade (2007–2020) (Ding et al., 2018). These datasets are available from https://www.temis.nl/emissions/region_asia/datapage.php (last access: 6 May 2021).

2.3 Ground-based observations

Since 2012, the Chinese government at various levels began to establish a national air quality monitoring network, which released real-time ground-level O\textsubscript{3} monitoring data to the public. By 2016, the establishment of more than 1000 sites was completed, covering more than 300 cities across the country. At each monitoring site, the concentration of O\textsubscript{3} is measured using the ultraviolet absorption spectrometry method and differential optical absorption spectroscopy; NO\textsubscript{2} is measured using the chemiluminescence method by a set of commercial instruments. The instrumental operation, maintenance, data assurance, and quality control were conducted based on the most recent revisions of China environmental protection standards (CMEE, 2013).

We use hourly O\textsubscript{3} and NO\textsubscript{2} concentrations (in standard conditions: 273 K, 101.325 kPa) from the network of ~1000 sites operated by the China Ministry of Ecology and Environment (CMEE) since 2016. CMEE revised the monitoring of pollutants to a new reference conditions (298 K, 101.325 kPa) since 1 September 2018 (CMEE, 2018). Daily ground-based O\textsubscript{3} and NO\textsubscript{2} observations are calculated from hourly observations at OMI overpass time (average of 13:00 and 14:00LT). In this study, we convert the gas concentrations before 1 September 2018 from the standard conditions to the reference conditions. The temperature dependence is according to Charles’s law (Eq. 1),

\[
\frac{V_{\text{std}}}{V_{\text{ref}}} = \frac{T_{\text{std}}}{T_{\text{ref}}},
\]

where \(V_{\text{std}}\) is the volume of a gas under standard conditions, \(V_{\text{ref}}\) is the volume of a gas under reference conditions, \(T_{\text{std}}\) (unit: K) is the thermodynamic temperature of standard conditions, and \(T_{\text{ref}}\) (unit: K) is the thermodynamic temperature of reference conditions. The gas concentration conversion follows

\[
\frac{C_{\text{std}}}{C_{\text{ref}}} = \frac{M/V_{\text{std}}}{M/V_{\text{ref}}} = \frac{V_{\text{ref}}}{V_{\text{std}}},
\]

where \(C_{\text{std}}\) is the gas concentration under standard conditions, and \(C_{\text{ref}}\) is the gas concentration under reference conditions.

Because the Chinese national air quality monitoring network stations are mostly located in the centre of cities or densely populated areas, which are usually the most polluted regions, we select the Naha station, located on the small island of Okinawa in Japan, as a location with a clean atmosphere. The hourly O\textsubscript{3} and NO\textsubscript{2} observations of Naha station are provided by the Japanese Atmospheric Environmental Regional Observation System (AEROS; http://soramame.taiki.go.jp/index.php, last access: 6 May 2021).

2.4 CLASS model

We simulate the non-linear relationship among O\textsubscript{3}, NO\textsubscript{2}, and HCHO using the Chemistry Land-surface Atmosphere Soil Slab model (CLASS). We performed a series of numerical experiments with the same dynamic and chemistry conditions listed in Table 1, but we modified only the concentrations of NO\textsubscript{2} and HCHO. The initial mixing ratios of chemical species are shown in Table S1 in the Supplement. The initial mixing ratio data are from van Stratum et al. (2012). All other species (except for molecular oxygen and nitrogen) are initialized at zero, and we modified only the concentrations of NO\textsubscript{2} and HCHO.

The CLASS model solves the diurnal evolution of dynamical variables (temperature, specific humidity, and wind) and chemical species over time in a well-mixed convective atmospheric boundary layer (ABL) in which entrainment and boundary layer growth are considered (Vilà-Guerau de Arellano et al., 2011; van Heerwaarden et al., 2010). All these variables are assumed to be constant with height due to intense turbulent mixing driven by convection (van Heerwaarden et al., 2010). The surface is assumed to be homogeneous in this box model. Chemistry is represented by a chemical scheme based on 27 reactions that control O\textsubscript{3} formation described by van Stratum et al. (2012), with O\textsubscript{3}, NO\textsubscript{x}, and isoprene as the most important species. This simplified chemical scheme is able to represent the evolution of chemical species in semirural areas (Janssen et al., 2012; van Stratum et al., 2012). This chemical scheme is able to represent the evolution of the O\textsubscript{3}–NO\textsubscript{x}–VOC–HO\textsubscript{2} cycle in semirural areas (Vilà-Guerau de Arellano et al., 2011; Janssen et al., 2012; van Stratum et al., 2012). The model has been validated under various dynamical conditions (Barboro et al., 2014; Janssen et al., 2012; van Heerwaarden et al., 2010).

3 Results

3.1 O\textsubscript{3} formation sensitivity regime classification

In Fig. 1a, the CLASS model is applied to generate O\textsubscript{3} isopleths, which illustrate O\textsubscript{3} as a function of NO\textsubscript{2} and HCHO values. The isopleths show that O\textsubscript{3} formation is a highly non-linear process in relation to NO\textsubscript{2} and HCHO. When NO\textsubscript{2} is low, the O\textsubscript{3} increases with increasing NO\textsubscript{2}. As NO\textsubscript{2} increases, the O\textsubscript{3} eventually reaches a local maximum. At
higher NO\textsubscript{2} concentrations, the O\textsubscript{3} would decrease with increasing NO\textsubscript{2}.

We first evaluate if satellite-based HCHO and NO\textsubscript{2} columns can capture the non-linear O\textsubscript{3}–NO\textsubscript{2}–HCHO chemistry shown by the CLASS model. In order to obtain a representative observation sample, we create monthly mean ground-based O\textsubscript{3} and NO\textsubscript{2} observations of 360 cities across China from the Chinese national air quality monitoring network from 2016 to 2019 and the background station observations from Naha, Japan, for comparison. Temperature is also a major factor in O\textsubscript{3} chemistry. O\textsubscript{3} pollution is rare when the ambient temperature is below 20 °C (Sillman, 2003). The seasonality of ground-level O\textsubscript{3} concentrations also exhibited monthly variability peaking in summer and reaching the lowest levels in winter over China (W. N. Wang et al., 2017). In addition, long NO\textsubscript{3}\textsuperscript{-} lifetime and low concentrations of OH and RO\textsubscript{2} radicals would lead most regions of China to a VOC-limited regime in winter (Shah et al., 2020). Therefore, we focus in this study on May–October as the summer period when meteorology is favourable for O\textsubscript{3} formation (Jin et al., 2017).

By directly connecting HCHO columns from OMI observations with ground-based measurements of NO\textsubscript{2} and O\textsubscript{3} from 360 cities across China during May–October from 2016 to 2019 in Fig. 1b, we find that the satellite-based HCHO columns and ground-based NO\textsubscript{2} concentrations can capture non-linear O\textsubscript{3} chemistry consistent with the CLASS model results. It indicates that tropospheric HCHO columns from OMI can represent the near-surface HCHO environment as revealed by previous studies (Martin et al., 2004; Duncan et al., 2010; Jin et al., 2017). The overall O\textsubscript{3}–NO\textsubscript{2}–HCHO chemistry is also captured by satellite-based HCHO and NO\textsubscript{2} columns in Fig. 1c, where we construct the O\textsubscript{3} isopleth using only observations.

| Item                        | Status or value |
|-----------------------------|-----------------|
| Total simulation time       | 12 h            |
| Time step                   | 60 s            |
| Initial ABL height          | 200 m           |
| Mixed layer                 | Off             |
| Initial mixed-layer potential temperature | 288 K |
| Initial temperature jump at height | 1 K   |
| Wind                        | Off             |
| Surface scheme (sea or land) | Off             |
| Chemistry                   | On              |

Figure S2a in the Supplement shows monthly O\textsubscript{3} concentration in winter (December–January–February), which rarely exceed 160 µg/m\textsuperscript{3}, including the FNR thresholds derived using summertime data. Based on Fig. S2b, we assume that our FNR thresholds [2.3, 4.2] derived using summertime data will be valid for all seasons. Three regimes can be roughly identified from the FNR thresholds we adopted: a VOC-limited regime should occur when the FNR < 2.3, and a NO\textsubscript{2}-limited regime should occur when the FNR > 4.2.
Figure 1. (a) The simulated O$_3$ isopleths versus NO$_2$ and HCHO using the CLASS model. (b) The 360 cities’ monthly mean in situ O$_3$ concentrations versus in situ NO$_2$ concentrations and HCHO columns from OMI observations in the summer during 2016–2019. Note that daily ground-based O$_3$ and NO$_2$ observations are calculated from hourly observations at OMI overpass time (averaged at 13:00 and 14:00LT). The O$_3$ numeric value of the grid cells is average of all points falling in each bin. (c) Same as (b) but with NO$_2$ columns from OMI observations. (d) The top 10% monthly O$_3$ values and corresponding FNRs of each city. FNR thresholds are defined as the ±30% range from the median of monthly O$_3$ exceeding 160 µg/m$^3$ in the top 10% dataset.

The FNR between 2.3 and 4.2 reflects the transition between the two regimes.

### 3.2 Variations in O$_3$ formation sensitivity in China

Figure 2a and b show the photochemical regime classification over China in summer of 2016 and 2019 using our FNR thresholds. Combined with the China provincial administrative division in Fig. S3 in the Supplement, we see the VOC-limited regimes mainly appear in the North China Plain (NCP), the Yangtze River Delta (YRD), and the Pearl River Delta (PRD), and the NO$_x$-limited regimes dominate the remaining areas, which are consistent with results from N. Wang et al. (2019) and Jin and Holloway (2015). In the NCP, the VOC-limited regimes are found in Beijing and some big cities in Hebei province, central regions in Shandong province, and Henan province. Transitional regimes control the remaining regions of Shandong province and Henan province and most regions of Hefei province. In the YRD, the VOC-limited regimes are found in Shanghai and southern Jiangsu province. In the PRD, the VOC-limited regimes are found in Guangzhou. Outside the NCP, YRD and PRD, the VOC-limited regimes concentrate in city centres of Shenyang, Chengdu, Chongqing, Xi’an, and Wuhan, which are surrounded by transitional regimes in the suburban areas. It has been acknowledged that the urban O$_3$ formations are generally VOC-limited due to the large amount of NO$_x$ emissions from diverse sectors, like transportation, industry, residential sector, and power plants (Shao et al., 2009; Wang et al., 2009; Sun et al., 2011). The NO$_x$-limited or transitional regimes dominated O$_3$ formation in the suburban and rural areas of eastern China (Xing et al., 2011; Jin et al., 2017).

Comparison of O$_3$ sensitivities between 2016 and 2019 shows noticeable changes from VOC-limited regime to transitional regime in the NCP, YRD, and PRD. In the NCP, the continuous area of VOC-limited regimes that occurred in 2016 change to transitional regimes in 2019. The VOC-limited regimes remain in central Beijing, Tianjin, Shijiazhuang, Jinan, and Zhengzhou. In the YRD, Shanghai and Nanjing remain in the VOC-limited regime, and other cities mostly change to the transitional regime. In the PRD, the VOC-limited regime still controls Guangzhou, while the transitional regimes control its surrounding cities.
Figure 2. (a) Photochemical regime classification over China in the summer of 2016. (b) Same as (a) but for 2019. Note that no data grids in (a) and (b) corresponds to monthly HCHO columns below the detection limit ($2 \times 10^{15}$ molecule/cm$^2$) or NO$_2$ columns lower than $1.5 \times 10^{15}$ molecule/cm$^2$. (c) Mean HCHO columns from OMI over China in the summer of 2016. (d) Same as (c) but for 2019. (e) Mean NO$_2$ columns from OMI over China in the summer of 2016. (f) Same as (e) but for 2019.

Figure 2c and d show mean HCHO columns over China in the summer of 2016 and 2019. The columns exceed $15 \times 10^{15}$ molecule/cm$^2$ in megacity clusters, such as in the NCP, YRD, and PRD, as well as the Sichuan Basin. Shen et al. (2019) found large increases of HCHO columns during May–September over 2005–2016 in the NCP and the YRD, consistent with the trend of anthropogenic VOC emissions. Our results show that the satellite HCHO columns increase in the NCP and the YRD and decrease in the PRD and in the Sichuan Basin during May–October of the 2016–2019 period. Figure 2e shows mean NO$_2$ columns over China in the summer of 2016. The NCP, YRD, PRD, Sichuan Basin, and Urumqi have high levels ($80 \times 10^{15}$ molecule/cm$^2$) of NO$_2$ columns. Figure 2f shows the satellite NO$_2$ columns have a strong decline in the NCP, the PRD, Hunan, Hubei, and Jiangxi provinces in summer from 2016 to 2019. However, the YRD shows increasing NO$_2$ columns in 2019.
We select typical cities (Beijing, Shanghai, Guangzhou, Neijiang, Lhasa, and Naha) to analyse in more detail the O₃ formation sensitivity in the summers of 2016 to 2019 in Fig. 3. These cities are selected based on their different chemical regimes in 2016. The locations of the six cities are shown in Fig. S4 in the Supplement. Economically developed megacities or provincial capital cities such as Beijing, Shanghai, and Guangzhou, with high levels of tropospheric NO₂ and HCHO, remain in the VOC-limited regime over 2016–2019. The reduction of tropospheric NO₂ results in a shift in the O₃ formation sensitivity in cities such as Neijiang over 2016–2019. Lhasa as a city with low NO₂ and the background station in Naha with even lower HCHO and NO₂ columns remain in the NOₓ-limited regime over 2016–2019.

As we know, O₃ increases with increasing NOₓ in the NOₓ-limited regime and decreases with increasing NOₓ in the VOC-limited regime. The contrast between NOₓ-limited and VOC-limited regimes illustrates the difficulties involved in developing policies to reduce O₃ in NOₓ polluted regions. Reductions in VOCs will only be effective in reducing O₃ if VOC-limited chemistry predominates. Reductions in NOₓ will be effective only if NOₓ-limited chemistry predominates and may actually increase O₃ in VOC-sensitive regions. If cities belonging to the VOC-limited regime like Beijing only focus on the reduction of NOₓ while ignore the control of VOC emissions, they will experience a process of rising O₃ concentrations, the more NOₓ decrease, the greater the increase in O₃ will be.

3.3 Observed response of ground-level O₃ to chemical formation sensitivity

To validate the regimes derived from satellite observations, we also analyse the surface NO₂ observations from ground-based measurements. Figure 4a and b show the mean ground-based NO₂ concentrations in summer of 2016 and 2019. According to the NOₓ surface emission estimates derived with DECSO from OMI observations, the NOₓ emissions in eastern China (18° N, 104° E, 41.5° N, 124° E) decrease from 5.93 Tg/yr in 2016 to 4.21 Tg/yr in 2019. Such a strong decline in NOₓ emissions led to decreasing ambient NO₂ concentrations at NCP (Beijing, Shijiazhuang, Zhengzhou, Jinan) and YRD (Hefei and other cities in Anhui province). In Fig. 4c, the national average NO₂ concentration decrease by 14.4 % in summer from 2016 to 2019.

Figure 4d and e show the mean ground-based O₃ concentration of about 360 cities across China in summer of 2016 and 2019. Generally, the O₃ levels in western China are lower than in eastern China. In 2016, few cities have an average O₃ concentration above 140 µg/m³. In 2019, cities with a mean O₃ concentration exceeding 140 µg/m³ occurred at the NCP (Tianjin, Shijiazhuang, some cities in Shandong and Henan province), the YRD (Nanjing), and the PRD (Guangzhou). In Fig. 4f, we see the number of cities with average O₃ values above 140 µg/m³ increases rapidly from 2.20 % in 2016 to 31.37 % in 2019. The cities with an average O₃ value below 80 µg/m³ decreased from 11.02 % in 2016 to 2.24 % in 2019. In addition, the nationwide O₃ average in summer increased year by year from 2016 (104.86 µg/m³) to 2019 (125.14 µg/m³). K. Li et al. (2019) reported the increasing O₃ trends in summer in megacity clusters of eastern China and the highest O₃ concentrations are in the NCP, which are consistent with our results.

A complex coupling of primary emissions, chemical transformation, and dynamic transport at different scales determine the O₃ pollution (Jacob, 1999). NOₓ and VOCs play important roles in O₃ formation. Emissions of NOₓ and VOCs to the environment are the starting point of O₃ pollution problems. During the past decade in China, ambitious steps have been taken to control NOₓ emissions. In 2013, the Chinese State Council issued the APPAP. Stringent control measures were carried out since then, including phasing out highly emitting industries, closing outdated factories, tightening industrial emission standard, improving fuel quality (N. Wang et al., 2019). However, to the other important O₃ precursors, VOCs, less attention has been given in emission control strategy. M. Li et al. (2019) concluded that anthropogenic NMVOC emissions in China during 1990–2017 have been increasing continuously due to the dramatic growth in activity rates and absence of effective control measures. Following China’s past control strategy on VOCs, we can regard VOC emissions as rising or in steady state.

The reduction of the NOₓ emissions for cities in the VOC-limited regime is one of the main reasons for the increasing of O₃. Figure 5a shows the difference of total NOₓ emissions derived from OMI observations in summer in east China between 2019 and 2016. A decline in NOₓ emissions centres at the NCP, YRD and PRD, where most areas belong to the VOC-limited regime. In order to provide further insight into the impact of NOₓ emission variations on O₃ concentrations, five selected typical cities (Beijing, Shanghai, Guangzhou,
Figure 4. (a) Mean ground-based NO$_2$ concentration at each city in the summer of 2016. (b) Same as (a) but for 2019. (c) The bars indicate the number of cities (left axis) in a certain NO$_2$ range in summer from 2016 to 2019. The black line indicates the average NO$_2$ concentration (right axis) of all cities. (d) Mean ground-based O$_3$ concentration at each city in summer of 2016. (e) Same as (d) but for 2019. (f) Same as (c) but for O$_3$. Note that daily in situ NO$_2$ and O$_3$ data are the average of 13:00–14:00 LT of the sites in each city.

Neijiang and Naha) are shown in more detail (see Fig. 5b and c). For cities under the control of VOC-limited chemistry (Beijing, Shanghai and Guangzhou), accompanied with decreasing NO$_x$ emissions, O$_3$ concentrations generally show an opposite behaviour to NO$_x$ emissions. The O$_3$ formation sensitivity in Neijiang shows a shift from the transitional to the NO$_x$-limited regime over 2016–2019. The reduction of NO$_x$ emissions in the transitional regime is accompanied by decreasing O$_3$ in Neijiang. Although the O$_3$ data in Naha for 2016–2018 are unavailable, we see that O$_3$ concentrations in Naha are low in 2019, and NO$_x$ emissions are stable during 2016–2019. Note that we find a qualitative relationship between NO$_x$ emission and the O$_3$ response patterns, confirming the non-linear O$_3$–NO$_2$–VOC chemistry but not in a quantitative sense. For example, the changes of NO$_x$ emissions in Beijing (−2.17 Gg N/cell), Shanghai (−1.18 Gg N/cell), Guangzhou (−0.28 Gg N/cell), and Neijiang (−0.15 Gg N/cell) during 2016–2019 lead to different levels of O$_3$ changes in Beijing (10.43 µg/m$^3$), Shanghai (7.81 µg/m$^3$), Guangzhou (25.54 µg/m$^3$), and Neijiang (−22.66 µg/m$^3$). Because of the VOC-limited chemistry conditions, O$_3$ increases with decreasing NO$_x$ emissions in Bei-

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Figure 5. (a) Differences in total NO\textsubscript{x} emissions derived from OMI observations in summer in east China between 2019 and 2016. (b) Variations in total NO\textsubscript{x} emissions in five cities (Beijing, Shanghai, Guangzhou, Neijiang, and Naha) in summer from 2016 to 2019. (c) Variations in mean ground-based O\textsubscript{3} concentrations in five cities in summer from 2016 to 2019.

The measures in response to the outbreak of the COVID-19 lead to sudden changes of NO\textsubscript{x} emissions and anthropogenic HCHO emissions in China in the beginning of 2020 (Wang et al., 2020; Hui et al., 2020). We analyse the change of O\textsubscript{3} concentrations during the lockdown period to validate our method. To look into COVID-19 lockdown impacts on short-term O\textsubscript{3} level, we choose two time periods covering 357 cities across China: period I (3–23 January 2020) and period II (9–29 February 2020), to avoid the coincidence of Chinese New Year holidays (24 January to 8 February 2020).

Figure 6a shows enhanced O\textsubscript{3} levels in most cities of eastern China during the COVID-19 lockdown, except for some cities in PRD and Fujian province. The cities with O\textsubscript{3} concentration increases of more than 40 µg/m\textsuperscript{3} are located in the NCP and the YRD, i.e. the populous regions of China, indicating a potential negative health effect from O\textsubscript{3} exposure in these regions. Figure 6b shows strong reductions in NO\textsubscript{x} emissions in eastern China, especially in Henan, Hubei, and Jiangsu provinces, where as a consequence of the lockdown, transportation, construction, and light industry activities have been dramatically decreased.

Assuming that our observation-based FNR thresholds derived using summertime data also apply during winter, we see that most regions of eastern China belong to the VOC-limited regime during periods I and II in Fig. 6c and d. Previous studies also reported that the O\textsubscript{3} chemistry in the urban areas in China in wintertime is in a VOC-limited regime due to the relative lack of HO\textsubscript{x} radicals (Seinfeld and Pandis, 2016). During winter (VOC-limited conditions), when the concentration of NO\textsubscript{x} is high and the level of UV radiation is low, the O\textsubscript{3} production varies inversely with the NO\textsubscript{x} concentration (Sillman et al., 1990). During the lockdown period, both the anthropogenic emissions of NO\textsubscript{x} and VOCs were reduced. The NO\textsubscript{x} reduction during the lockdown is higher than the VOC reduction according to Sicard et al. (2020). The reductions of VOC emissions are generally effective in reducing O\textsubscript{3} concentrations. However, such air quality improvements are largely offset by reductions in NO\textsubscript{x} emissions leading to increases in O\textsubscript{3} concentrations due to the strongly VOC-limited conditions in the NCP in winter (Xing et al., 2020). The NO\textsubscript{x} reduction during the lockdown is higher than the VOC reduction (Sicard et al., 2020). Thus,
a reduction in NO\textsubscript{x} leads to an increase in the O\textsubscript{3} concentrations in most regions of eastern China during period II. Besides, reduction of freshly emitted NO in particular from road traffic alleviates O\textsubscript{3} titration locally (Seinfeld and Pandis, 2016; Levy et al., 2014). The O\textsubscript{3} titration occurs particularly in winter (less photolysis reactions of NO\textsubscript{2}) under high NO\textsubscript{x} levels (Sillman, 1999). However, the lockdown measures result primarily in a lower titration of O\textsubscript{3} by NO due to the reduction in local NO\textsubscript{x} emissions by road transport, which also enhances O\textsubscript{3} levels in urban areas. On the other hand, some cities, mainly located in southeastern China, showed decreasing O\textsubscript{3} levels. Zhao et al. (2020) concluded that the cause of O\textsubscript{3} decline in these cities is the emission changes of NO\textsubscript{x} and VOC. In Fig. 6c we see that some cities in Fujian and Guangdong provinces belong to the transitional regime. Theoretically, the transitional regime should correspond to the conditions at which O\textsubscript{3} formation is most efficient, indicating that reductions or increases in NO\textsubscript{x} and VOCs will reduce the O\textsubscript{3} concentration.

4 Conclusion
Satellite-based HCHO/NO\textsubscript{2} ratios and ground-based O\textsubscript{3} measurements were directly connected to capture the non-
We presented the level of O₃ formed from photo-oxidation of total measured HCHO only not differentiating the contributions from different sources (directly emitted or photochemically formed). Due to the higher temperature and stronger solar radiation in summer, the higher concentration level of HCHO mainly results from the intense photo-oxidation of VOCs. Emission sources of HCHO, as a tracer of VOCs, can be anthropogenic and biogenic. Shen et al. (2019) found that the OMI HCHO distribution follows their anthropogenic inventory in megacity clusters over China, while it does not follow the biogenic emissions inventory. Despite the fact that local sources of anthropogenic VOCs are difficult to identify, our FNR thresholds derived from satellite-based information have the potential to provide important information to air quality planners. Compared with stringent control measures for NOₓ emissions, VOC emissions got less attention as the other O₃ precursor in China. The case study of O₃ level changes during the COVID-19 lockdown in China demonstrated that the strong reductions in anthropogenic NOₓ emissions resulted in significant O₃ enhancement due to the VOC-limited regime in winter. It indicates that a protocol with strict measures to control NOₓ emissions, without simultaneous VOC emissions controls for power plants and heavy industry, such as petrochemical facilities, achieves only limited effects on O₃ pollution.

Data availability. Satellite data used in this research can be obtained from public sources. The OMI tropospheric NO₂ product from the QA4ECV project can be obtained from https://doi.org/10.21944/qa4ecv-no2-omi-v1.1 (Boersma et al., 2017), and the HCHO product can be obtained from https://doi.org/10.18758/71021031 (De Smedt et al., 2017b).

The hourly O₃ and NO₂ observations of Chinese ground stations can be accessed from third parties (http://www.temis.nl/emissions/region_asia/datapage.php (Ding et al., 2018).

The hourly O₃ and NO₂ observations of Chinese ground stations can be accessed from third parties (http://www.temis.nl/emissions/region_asia/datapage.php (Ding et al., 2018).

The hourly O₃ and NO₂ observations of Naha station are provided by the Japanese Atmospheric Environmental Regional Observation System (AEROS; http://soramame.taiki.go.jp/Download.php, Japanese Ministry of the Environment, 2021).

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