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The impact of urban agglomeration on ozone precursor conditions: A systematic investigation across global agglomerations utilizing multi-source geospatial datasets

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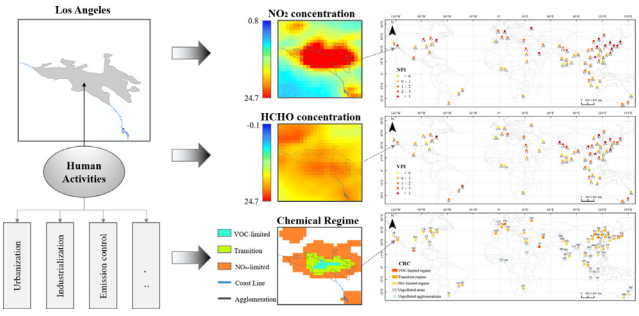
HIGHLIGHTS

- Not all agglomerations have a positive impact on ozone precursor conditions.
- Linkages between natural and human activities on agglomeration influence are explored.
- Agglomeration influence intensifies with urbanization and industrialization.
- Trends of precursors are opposite in developed and developing agglomerations.

GRAPHICAL ABSTRACT

The influence of urban agglomeration on ozone formation (e.g. Los Angles).



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ABSTRACT

Urbanization significantly influences ozone via two conditions of its formation: 1) precursor concentration; and 2) chemical regime. Recently, there has been raised concern about the influence of urban agglomerations on these two conditions. Although valuable efforts have been made, some contrary viewpoints exist. Meanwhile, urban agglomerations in developed and developing regions are experiencing different urbanization processes, so a systematic comparison between these two regions is warranted. In this context, by leveraging multi-source geospatial datasets, this paper systematically gauges the influence of urban agglomerations on ozone precursor conditions and further investigates the spatiotemporal variations. Based on the analysis of 71 global agglomerations during 2005–2016, it is found that: 1) not all urban agglomerations have a positive effect on ozone precursor conditions; 2) the negative effects of urban agglomerations can be attributed to the low latitudes and the ecological areas ($p < 0.05$); 3) the agglomeration influence intensifies with the increase of built-up area, population, and latitude ($p < 0.05$); 4) the anthropogenic nitrogen oxide (NO_x) emission from all sectors can aggravate the magnitude of the urban agglomeration influence ($p < 0.05$), while for volatile organic compounds (VOC_s), only the contribution of industrial emissions is significant ($p < 0.05$); and 5) in view of the temporal dynamics, the influence of urban agglomeration on ozone precursor condition is opposite in developed and developing regions. This study will provide important insights for future urban agglomeration studies and ozone pollution monitoring with geospatial datasets.

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1. Introduction

Over the past few decades, the world has experienced rapid urbanization (United Nations, 2010). On the one hand, the distances between urban areas are narrowing, and urban areas are merging into a new form, i.e., urban agglomerations, for the purposes of economic competitiveness and sustainable development (Taubenböck and Wiesner, 2015). This can profoundly alter the emission and transmission of air pollutants. On the other hand, the increase in urban agglomeration population has led to an increase in energy consumption and the emission of air pollutants (Schneider et al., 2015). Against this backdrop, it is necessary to examine the impact of urban agglomerations on air pollutants.

Ozone is receiving growing attention, not only for its detrimental effect on human health and vegetation, but also for its role as a greenhouse gas. As a secondary air pollutant, ozone is formed through the photochemical reaction of its precursors, including nitrogen oxide (NO_x) and volatile organic compounds (VOC_s) (Martin et al., 2004). As one of the ozone formation conditions, the precursor concentration has been found to be different between the central areas of urban agglomerations and the surrounding backgrounds. In previous studies, it has been proved that this agglomeration-background difference can be utilized to describe the agglomeration influence on ozone precursor (Baklanov et al., 2016; Kar et al., 2010; Safieddine et al., 2013). Given that the intensive human activities within urban agglomerations emit large quantities of ozone precursors, the precursor concentration in urban agglomerations can be expected to be higher than that of the surrounding background (i.e., a positive effect) (Baklanov et al., 2016; Kar et al., 2010; Safieddine et al., 2013). However, the opposite phenomenon (i.e., a negative effect) has also been reported in previous studies (Safieddine et al., 2013). It is difficult to explain this inconsistent result, in view of the limited study regions (i.e., single or several agglomerations), the different definitions of "urban agglomeration" (Jin and Holloway, 2015; Safieddine et al., 2013; Schneider et al., 2015), and the divergence in urbanization levels (Lamsal et al., 2013; Schneider et al., 2015; Zhang et al., 2011), industrial activity (Huang et al., 2013; Zhang et al., 2011), and ecological areas (Zhu et al., 2014, 2017). Therefore, a comprehensive study investigating the spatial variation in the influence of urban agglomerations on ozone precursors and the underlying drivers is warranted.

As the other critical condition of ozone formation, the chemical regimes can be divided into three categories (i.e., NO_x-limited, VOC-limited, and transition regimes) (Duncan et al., 2010; Guo et al., 2017; Martin et al., 2004; Monks et al., 2015). In the different chemical regimes, the relationships between ozone and NO_x are varied: the rise in NO_x concentration leads to an increase of ozone in a NO_x-limited regime, but a decrease in a VOC-limited regime (Duncan et al., 2010; Guo et al., 2017; Martin et al., 2004; Monks et al., 2015). Hence, urban agglomerations with the same NO_x concentrations but different chemical regimes may have different effects on ozone formation. Therefore, it is important to correctly represent the chemical regime of urban agglomerations. In previous local-scale studies, the heterogeneity in the chemical regimes within urban areas has been addressed (Jin and Holloway, 2015; Ou et al., 2016; Witte et al., 2011). For example, Witte et al. (2011) showed that the VOC-limited regime comprised approximately half of the Beijing urban area, while the non-urban area in Beijing is dominated by the transition regime. However, to date, there has been a lack of systematically quantitative research on the heterogeneity of the chemical systems in urban agglomerations. In this paper, to address this problem, an effective metric, i.e., the proportion of each chemical regime, is calculated.

In addition, the temporal trend of the urban agglomeration influence on ozone formation also calls for further study. Firstly, previous studies have only analyzed the trends of precursor concentrations in urban agglomerations (Schneider et al., 2015; Schneider and van der A, 2012), which might introduce interference from the surrounding background. Secondly, most of these studies have utilized the average chemical regime to represent the impact of the urban agglomeration on the chemical regime (Choi and Souri, 2015; Jin and Holloway, 2015). In view of the uneven chemical regimes within urban agglomerations (Jin and Holloway, 2015; Ou et al., 2016; Witte et al., 2011), the average value of the chemical regime can import bias into the trend analysis. Finally, it is necessary to systematically compare the urban agglomeration influence on ozone precursor conditions between developed and developing regions, given their disparities in urbanization processes (Birch and Wachter, 2011) and precursor control measures (Huang et al., 2017b; Simon et al., 2015).

Traditionally, the influence of an anthropogenic source on ozone precursor conditions is quantified by the difference between an air monitoring station located near the source and another in the surrounding natural background (Novotny et al., 2011; Xu et al., 2011). However, the point measurements from these stations cannot represent the photochemical conditions of whole study regions in an unbiased manner. As an alternative, satellite observations can supply spatially and temporally continuous precursor concentration and chemical regime information (Jin and Holloway, 2015; Sagan et al., 2018; Ziemke et al., 2006). Meanwhile, multi-source geospatial data can offer information about ecological and anthropogenic factors (including the urbanization level (Ural et al., 2011), industrial activities (Wong et al., 2013), and ecological areas (Huang and Friedl, 2014)), which provides support to evaluate the relationship between these factors and the urban agglomeration influence on ozone precursor conditions (Lasaponara, 2013).

In this study, by leveraging multi-source geospatial datasets, we aimed to gauge the impacts of urban agglomerations on ozone precursor conditions, investigate the underlying factors of the spatial variation, and systematically compare the trends between developed and developing regions. To eliminate the uncertainties deriving from the definition of "urban agglomeration", 71 global urban agglomerations were extracted utilizing the maps of the Global Human Settlement Layer (GHSL) (Pesaresi et al., 2013). With satellite precursor observations for 2005–2016 provided by the Ozone Monitoring Instrument (OMI) onboard the NASA Aura spacecraft, two indices—the precursor pollution intensity (PPI) and the chemical regime coverage (CRC)—were developed to describe the influence of urban agglomerations on precursor concentration and the chemical regime, respectively. Based on these observations, this study aimed to answer the following questions:

1) Do all urban agglomerations have a positive impact on ozone precursor conditions?
2) If not, what factors contribute to the variation of the urban agglomeration influence?
3) How has the urban agglomeration influence on ozone precursor conditions changed over the last decade?

The findings presented below will be helpful for garnering a comprehensive understanding of the influence of urban agglomerations on ozone.

2. Materials and methods

2.1. Extracting urban agglomerations and backgrounds

The urban agglomeration, as a continuous urban area with dense population as well as various human activities

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(Taubenböck and Wiesner, 2015), is an ideal research unit for monitoring the effect of complex human activities on ozone formation conditions. In this paper, an "urban agglomeration" is defined as a spatial entity that contains at least two spatially connected cities and more than 5 million inhabitants. Following the method of Taubenböck (2015), the spatial extents of the main urban agglomerations were determined (for more details, see the Supporting Information (SI) Text 1), utilizing the built-up map, the population density map, and the settlement map of 2015 GHSL, which is a framework providing accurate global human settlement information (the overall accuracy is as high as 90%) (Pesaresi et al., 2013). Once an agglomeration was delineated, its surrounding background was set as the dry land that covered ±3° in latitude and longitude around the targeted agglomeration and that was isolated from other agglomerations, as suggested by Kar et al. (2010). For the coastal agglomerations, the areas of natural background that overlapped with the sea were excluded to reduce the bias from the marine. In total, 71 agglomerations were extracted in this study, most of which locate in Asia and North America (See Fig. 1, Table S1 in the SI). To include agglomerations from six continents, three Oceania urban agglomerations with populations less than 5 million, i.e., Melbourne, Brisbane, and Sydney, were considered in this study. Based on the United Nations Framework Convention on Climate Change Annex I and II (United Nations, 2010), six districts with different development degrees were selected: three developed regions, i.e., North America, Europe, and East Asia; and three developing regions, i.e., the Middle East, South Asia, and Southeast Asia (Fig. 1).

2.2. Quantifying the influence of urban agglomerations on ozone precursor conditions

To quantify the impact of urban agglomerations on ozone formation conditions, two kinds of metrics were developed from the perspective of precursor concentration and chemical regime. Firstly, the precursor pollution intensity (PPI) was used to indicate the influence of urban agglomerations on precursor concentration, the equations for which are as follows:

$$\mathrm{NPI} = \mathrm{NO_{x,AG}} - -\mathrm{NO_{x,BK}} \quad (1)$$

$$\mathrm{VPI} = \mathrm{VOC_{s,AG}} - -\mathrm{VOC_{s,BK}} \quad (2)$$

where NPI and VPI represent the pollution intensity of NO_x and VOC_s, respectively. $NO_{x,\ AG}$ and $VOC_{s,\ AG}$ represent the average NO_x and VOC_s concentration in the central urban agglomerations, respectively. $NO_{x,\ BK}$ and $VOC_{s,\ BK}$ denote the average concentration in the surrounding backgrounds. As suggested by Kar et al. (2010), the background, which is less affected by human activities, can be viewed as a baseline of the local environment (see Fig. 2a, b). Values higher than the uncertainty of the precursor products indicate that the urban agglomeration has a positive impact on precursor concentration, and vice versa.

To calculate the average precursor concentration, the satellite-provided tropospheric nitrogen dioxide (NO_2) and formaldehyde (HCHO) columns, which are widely used as surrogates for NO_x and VOC_s (Duncan et al., 2010; Martin et al., 2004), were obtained from the OMI (Sagan et al., 2018; Ziemke et al., 2006). The OMI,

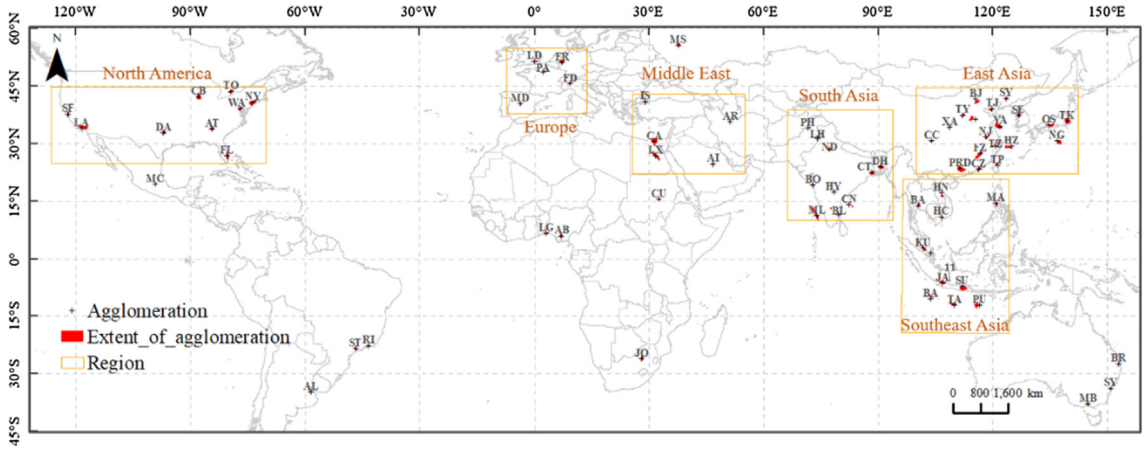

Fig. 1. Locations of the 71 global agglomerations and the six delineated regions. The labels represent the abbreviations for the names of the urban agglomerations. For the full names of the urban agglomerations and the values of the PPI, see the Supporting Information Table S1.

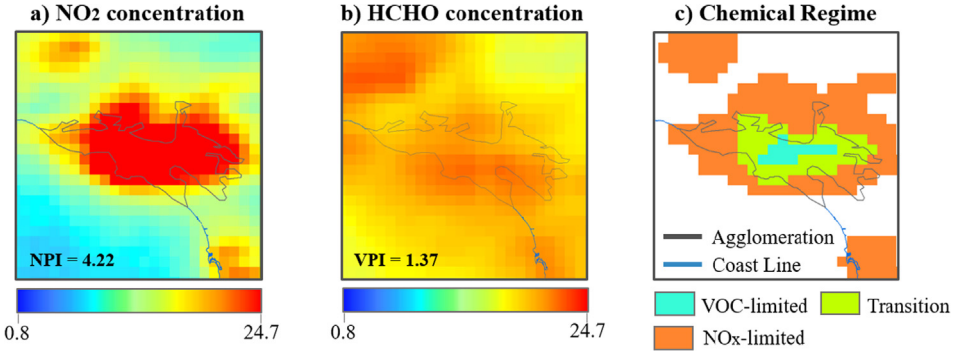

Fig. 2. The spatial patterns of nitrogen dioxide (NO_2) concentration, formaldehyde (HCHO) concentration, and chemical regime at Los Angeles, U.S. and their surrounding backgrounds, during 2005–2016.

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which is an ultraviolet/visible nadir-viewing spectrometer onboard the Aura satellite in a polar sun-synchronous orbit, can provide daily global coverage of ozone and its precursors at 13:30 local time. Considering the high uncertainty of the daily products, monthly mean Dutch OMI NO_2 (DOMINO-V2) with a resolution of 0.125° × 0.125° and an uncertainty of 1×10^{15} mol/cm^2 was utilized in this study (Boersma et al., 2010). The monthly mean Belgian Institute for Space Aeronomy (BIRA, version 14) OMI HCHO with a spatial resolution of 0.25° × 0.25° and an uncertainty of 0.7×10^{16} mol/cm^2 was also used (De Smedt et al., 2015). The feasibility of using these data in analyzing ozone precursor has been proved in previous studies (Baek et al., 2014; Boersma et al., 2011; Duncan et al., 2010; Martin et al., 2004). In order to keep accordance with the resolution of the NO_2 data, the HCHO data were resampled to 0.125°.

According to previous studies (Duncan et al., 2010; Martin et al., 2004), within an urban agglomeration, a polluted area is defined as an area within which the NO_2 concentration is higher than 2.5×10^{15} mol/cm^2; otherwise, the area is assigned as a non-polluted area. The ambient NO_x can arise from both natural sources (e.g., biogenic emissions, soil microbial activity, and lighting) and anthropogenic sources (e.g., fossil fuel combustion, biomass burning). For areas with an NO_2 concentration of less than 2.5×10^{15} mol/cm^2, it is implied that the ozone precursors of these regions are less likely to be emitted by anthropogenic sources (Duncan et al., 2010; Martin et al., 2004). Therefore, to highlight the influence of urban agglomerations, the areas with NO_2 concentrations of less than 2.5×10^{15} mol/cm^2 were classified as non-polluted areas.

However, polluted areas with the same NO_2 concentration but different chemical regimes can have different effects on ozone. With this characteristic, the chemical regime coverage (CRC), which is defined as the proportion of the chemical regimes within an urban agglomeration, can act as a complementary measurement to NO_2 concentration to depict the influence of urban agglomerations on chemical regime. In a natural background, the major contribution to NO_2 concentration is lighting (Duncan et al., 2010). As such, the NO_2 concentration in natural backgrounds is much lower than in polluted regions. As a result, the natural background is generally characterized as a non-polluted area. A larger polluted area indicates a larger extent of the urban agglomeration influence. Moreover, in view of the heterogeneous chemical regimes in polluted areas, the CRC discriminates each chemical regime, which can address the diversity of urban agglomeration pollution.

To calculate the CRC, the urban agglomerations were first divided into pixels with resolutions of 0.125°. This resolution was sufficient to capture most of the characteristics of the chemical regimes within the pixels (Hodnebrog et al., 2011). Polluted areas within the urban agglomerations were then extracted according to the DOMINO-V2 data. Note that the urban agglomerations whose pixel concentrations were all less than 2.5×10^{15} mol/cm^2 were considered as unpolluted agglomerations. Next, each polluted area was assigned into a regime, according to the criteria proposed by Jin et al. (2015), based on the monthly mean global maps of the HCHO/NO_2 ratio of the OMI. The HCHO/NO_2 ratio, which associates closely to the VOC/NO_x ratio, exhibits a good performance in determining chemical regimes (Duncan et al., 2010; Martin et al., 2004), and the reliability of the HCHO/NO_2 ratio in estimating chemical regimes has been confirmed in previous studies (Duncan et al., 2010; Martin et al., 2004). Finally, the influence of an urban agglomeration on the chemical regime was characterized as the coverage of each regime within the agglomeration.

2.3. Spatial variation of the urban agglomeration influences

To investigate the spatial variation in the urban agglomeration influence on ozone formation conditions and the possible driving forces, four factors were considered in this study: 1) urbanization level; 2) anthropogenic emissions; 3) latitude; and 4) ecological areas. A Spearman's rank correlation analysis was conducted to examine the relationship between the urban agglomeration influence and these factors. Based on the PPI and the size of the polluted areas, the 71 agglomerations were divided into several groups (i.e., positive vs negative NPI agglomerations, positive vs negative VPI agglomerations, and polluted vs unpolluted agglomerations), to investigate the differences under distinct agglomeration influences. It is known that ozone formation is active during periods with favorable meteorological conditions, such as high temperature, low humidity and strong solar radiation. Unfortunately, a global synoptic system which classifies these meteorological conditions is still lacking at present. Hence, to focus on the time period of ozone formation, the study period is restricted by the combination of total solar radiation and month (i.e., summer time), as both high temperature and strong solar radiation are conducive to ozone formation (Bell et al., 2007; Li et al., 2019; Wang et al., 2014). In more details, during the summertime, observations with solar radiation lower than 150 W/m^2 are excluded, as ozone formation is relatively rare when the total solar radiation is lower than 150 W/m^2 (Rizk, 1992). Solar radiation flux were acquired from the National Centers for Atmospheric Prediction and the National Center for Atmospheric Research (NCEP/NCAR) reanalysis data set (Kalnay et al., 1996). For the number of valid observations, see the Fig. S1.

Built-up area and population density maps from the 2015 GHSL (Pesaresi et al., 2013) were utilized to estimate the urbanization level of the urban agglomerations. The forest area sizes of the urban agglomerations were utilized as the indicators of ecological areas. The forest cover within the agglomeration background was extracted from the International Geosphere-Biosphere Program (IGBP) global vegetation classification scheme of the MODIS Land Cover Type product (Friedl et al., 2010). Given that the contributions of different emissions sources, including the emissions of power plants, road transportation, industry, and residential, to the urban agglomeration influence can be different, anthropogenic precursor emissions with global distributions were collected from the Emissions Database for Global Atmospheric Research (EDGAR v4.3.1), which is an emissions database developed by the European Commission (Crippa et al., 2016; Huang et al., 2017a). Due to the data availability, annual anthropogenic emissions for 2005–2012 and forest cover for 2005–2013 were utilized.

2.4. Temporal variations of the urban agglomeration influences

To estimate the trends of the PPI, a linear regression model in conjunction with bootstrap resampling and Fourier terms (Gardiner et al., 2008) was applied, which can eliminate the interference of the meteorological conditions (David and Nair, 2013) and sporadic wildfires (Pfister et al., 2008):

$$m(t) = A + Bt + \sum_{n=1}^{4} C_n cos(n2\pi t) + \sum_{n=1}^{4} D_n sin(n2\pi t) + \varepsilon \quad (3)$$

where m(t) is the monthly pollution intensity; t is the month (expressed in fractional year format); A and B represent the mean term in the first year and the annual trend, respectively; and C_n and D_n refer to the Fourier terms. Following Gardiner et al. (2008), a bootstrap method with 2000 iterations was performed to estimate the 95% confidence interval for the derived trend (i.e., B in Eq. (3)) (Gardiner et al., 2008).

To analyze the temporal variations of CRC, the difference between 2006 and 2015 was computed for each urban agglomeration. To reduce the interference of meteorological conditions (Witte et al., 2011) and sporadic wildfires (Jing et al., 2014), the

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average summer regime coverage spanning 2005–2008 and 2013–2016 was utilized to represent the coverage in 2006 and 2015, respectively. Upon deriving the trends of the PPI and CRC, a comprehensive comparison was made between the developed and developing regions.

3. Results and discussion

3.1. The positive and negative influences of urban agglomerations on ozone precursor conditions

If the PPI is higher than the uncertainty of the precursor products or the pollution area of the urban agglomeration is greater than zero, the urban agglomeration is considered to have a positive effect on the ozone precursor conditions, and vice versa. By averaging the PPI and CRC over 2005–2016, the direction (i.e., negative and positive) of the influence for each agglomeration was established (Fig. 3). Overall, although most urban agglomerations exhibit a positive effect on ozone precursor conditions, there are still 16 unpolluted urban agglomerations and 10 (12) urban agglomerations with negative NPI (VPI).

Table 1 lists the pairwise difference in the factor of interest (e.g., built-up areas) between the urban agglomerations. Most of the high-latitude urban agglomerations are polluted areas (Fig. 3, and Student's *t*-test $p < 0.001^{**}$), and are positive in NPI (Fig. 3, and Student's *t*-test $p < 0.001^{**}$) and VPI (Fig. 3, and Student's *t*-test $p < 0.001^{**}$). Meanwhile, with the aid of ecological areas, VOC_s pollution can be lighten (Student's *t*-test $p = 0.031$).

3.2. Possible mechanisms underlying the spatial variability of the PPI and CRC

In more details, the spatial variability can be summarized as follows (Fig. 3). According to the PPI, the urban agglomeration influence can be divided into the following categories: 1) both high NPI and VPI, such as New York (NY), Beijing (BE), the Pearl River Delta (PRD), and Tokyo (TK), indicating the collocated anthropogenic NO_x and VOC_s emissions of these urban agglomerations; 2) high NPI but low VPI, such as Cologne (CL), Riyadh (RI), Handan (HD), Taiyuan (TY), and Mexico City (ME); 3) high VPI but low NPI, such as Peshawar (PH), Hanoi (HN), and Chengdu (CD), where the opposite phenomenon between 2) and 3) can be attributed to the disproportionate emissions of NO_x and VOC_s; and 4) both low NPI and VPI, such as the urban agglomerations in tropical regions (30°S ~ 30°N), suggesting the interference of the high temperature and the forest areas in the tropics.

In terms of CRC, the urban agglomerations are generally covered by more than one regime, indicating the heterogeneity of the chemical regimes within urban agglomerations. When more than 50% of the urban agglomeration area is dominated by a specific chemical regime, it is called a "chemical-dominated regime". According to the daily wall-to-wall satellite observations over 2005–2016, only three urban agglomerations (i.e., Buraydah, Cologne, and Mexico) are VOC-dominated regime, 16 urban agglomerations are transition- dominated regime, and 18 urban agglomerations are NO_x-dominated regime.

Urban agglomerations can exert effects on ozone formation conditions through influencing the emission, transmission, and deposition process of the ozone precursors. To investigate the possible mechanisms underlying the spatial variability of the PPI and CRC, the geographical and anthropogenic factors are summarized below (Table 2).

3.2.1. Latitude

Both the NPI and VPI show a close linkage with latitude (Table 2), which is due to the decomposition- and transmission-related geographical factors and the regional ozone precursor emissions. In the tropics, the high temperature promotes the photolysis of NO_x, and hence the NPI is weaker in the tropics (30°S ~ 30°N) than in mid-latitudes (3°N ~ 6°N, 30°S ~ 60°S, see Fig. 3) (David and Nair, 2013). In terms of the VPI, high temperatures promote the emission of biogenic VOC_s in background areas, which attenuates the VPI of tropical agglomerations (30°S ~ 30°N) (Fu et al., 2007). Meanwhile, as the lifetime of NO_x in mid-latitudes is longer, which enables the transmission of precursors to more distant areas, the polluted areas are larger in mid-latitude regions than in the tropics (Table 2).

3.2.2. Urbanization level

As effective indicators of urbanization level, both built-up areas and population size affect the ozone formation conditions through influencing the emission and transmission process of ozone precursors. Significant correlations between built-up areas, population, and the PPI and CRC are apparent (see the last three columns in Table 2). The conversion of natural land use to built-up areas and the growth of population can lead to the rise of anthropogenic emissions (Schneider et al., 2015). Meanwhile, the presence of high-density population and building areas is usually accompanied by skyscrapers, which may impede the dispersion of ozone precursors (Shi et al., 2016). Together, these factors lead to the accumulation of ozone precursors over urban agglomerations, which indirectly increases the intensity and the extent of the urban agglomeration influence.

3.2.3. Anthropogenic emissions

The contribution of each anthropogenic emission source with specific characteristics is further explored. As shown in Table 2, the NPI is significantly correlated with all the emissions sectors, indicating that all the anthropogenic emissions can aggravate the magnitude of the urban agglomeration influence, while the moderate natural NO_x emissions from lightning, soil, and forest fires hardly affect the NPI (Vestreng et al., 2007). Meanwhile, the increase in each anthropogenic NO_x emission source also leads to the expansion of the polluted areas, as shown by the significant positive correlation apparent in Table 2. NO_x concentration decays with the distance to the emission source, which is a closely related factor to emission concentration and lifetime (Su et al., 2009). Therefore, a rise in anthropogenic NO_x emissions indicates an increase in the decay distance, leading to the expansion of the polluted areas.

Although a previous study argued that biogenic VOC_s emissions may attenuate the agglomeration influence (Boeke et al., 2011), the VPI is still correlated with the emissions of industrial sectors, suggesting that the contribution of the industrial emissions to the urban agglomeration influence is the most significant. The industrial emissions also significantly correlate with the coverage of the VOC-limited regime, since industry, a point source with heavy anthropogenic emissions, is generally surrounded by a VOC-limited regime. In terms of residential emissions, there is a significant correlation with the NO_x-limited regime, as residential regions, an area source with relatively light anthropogenic emissions, are generally surrounded by a NO_x-limited regime. Meanwhile, the transportation emissions correlate positively with the coverage of both the NO_x-limited regime and transition regime, as transportation, a line source with heavy anthropogenic emissions, is generally surrounded by a transition-limited regime.

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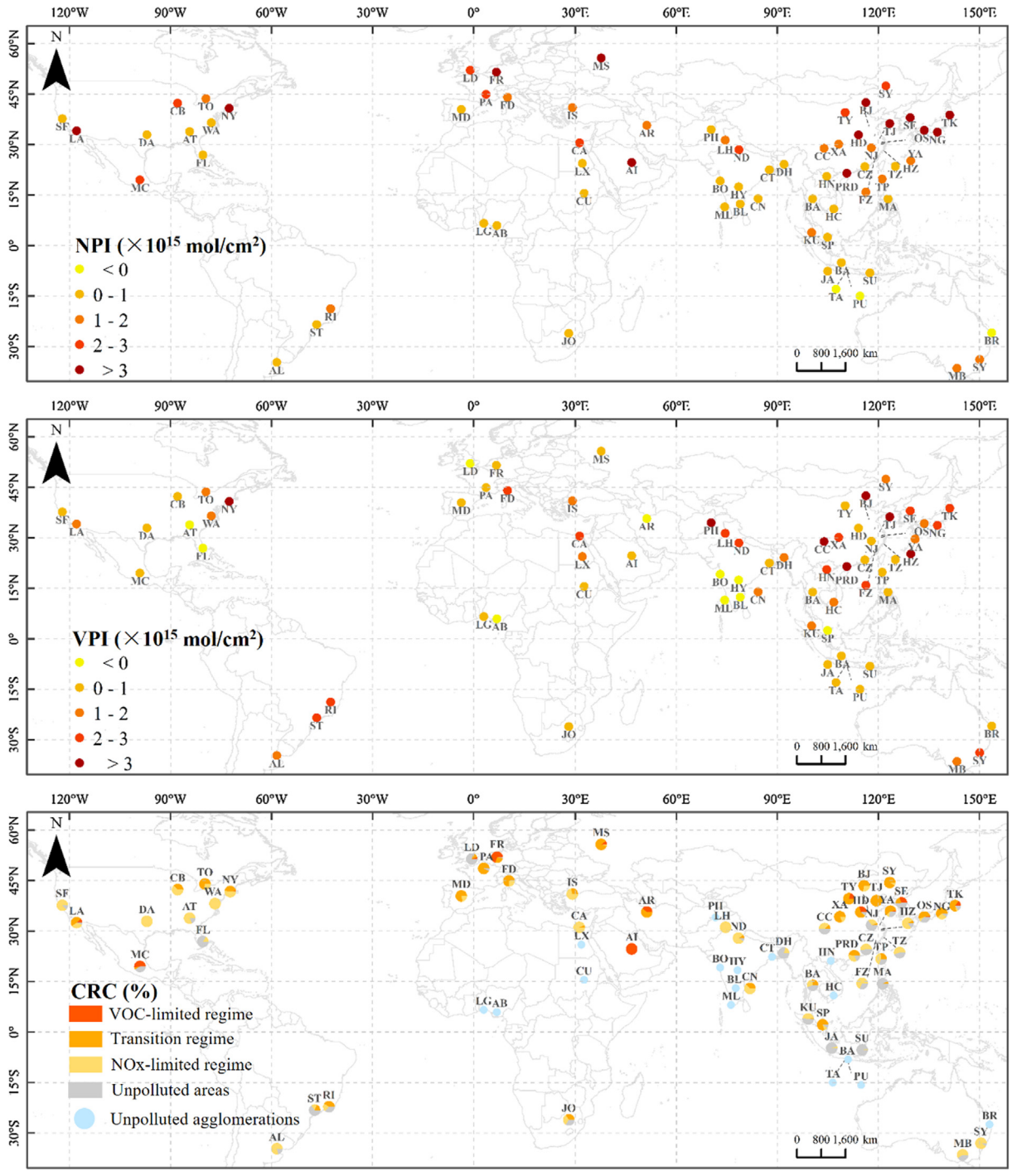

Fig. 3. Spatial distribution of NPI, VPI, and CRC across the 71 global agglomerations in 2005–2016. The labels represent the abbreviations for the names of the urban agglomerations. For the full names of the urban agglomerations and the values of the PPI, see the Supporting Information Table S1.

Table 1
Pairwise difference (Student's *t*-test) in the factor of interest between urban agglomerations.

Urban agglomerations	Built-up areas	Population	Latitude	Anthropogenic emissions				Ecological areas
				Power plants	Industry	Residential	Transportation	
Positive vs negative NPI	0.052	0.557	<0.001**	0.122	0.097	0.191	0.093	0.126
Positive vs negative VPI	0.061	0.125	<0.001**	0.099	0.080	0.067	0.134	0.031*
Polluted vs unpolluted	0.090	0.108	<0.001**	0.117	0.108	0.088	0.071	0.007**

* and ** indicate that the difference between groups is significant, i.e., $p < 0.05$ and $p < 0.01$, respectively.

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Table 2
Spearman's rank correlation coefficients between the PPI, CRC and possible driving factors, including population, built-up areas, latitude, and anthropogenic emission from different sectors (i.e., power plants, industry, residential, transportation sectors).

Factor of interest		PPI		Polluted areas	CRC		
		NPI	VPI		VOC-limited	Transition	NO_x-limited
Geographic	Latitude	0.420**	0.234**	0.624**	0.393**	0.560**	0.109**
Urbanization level	Population	0.374**	0.505**	0.440**	0.397**	0.310**	0.690**
	Built-up areas	0.717**	0.412**	0.724**	0.350**	0.687**	0.710**
NO_x emissions	Power plants	0.138*	0.079	0.103*	0.068	0.083	0.034
	Industry	0.352**	0.133	0.345**	0.280*	0.013	0.191
	Residential	0.285*	0.086	0.318**	0.177	0.275*	0.158
	Transportation	0.239*	0.128	0.336**	−0.046	0.168*	0.298*
VOC_s emissions	Power plants	0.102	0.039	0.158	0.031	−0.094	−0.003
	Industry	0.262	0.199*	0.215	0.192	−0.154	−0.161
	Residential	−0.119	0.111	0.239*	0.043	−0.062	−0.117
	Transportation	−0.006	0.050	0.021	0.056	−0.099	0.156

* and ** indicate that the difference between groups is significant, i.e., $p < 0.05$ and $p < 0.01$, respectively.

3.3. Opposite trends in agglomeration influence between developed and developing regions

3.3.1. The trends of agglomeration influence intensity

In view of the NPI, its trends indicate that the intensity of the urban agglomeration influence is weakened in developed regions, while it is strengthened in developing regions (Fig. 4). A statistically significant ($p < 0.05$) increasing trend in NPI from 2005 to 2016 is seen over 15 urban agglomerations, 14 of which are located in developing regions. The increasing trend is most pronounced in Dhaka, where the NPI is increasing by more than 0.19×10^{15} mol/cm^2 yr^{-1}. In contrast, statistically significant ($p < 0.05$) decreasing trends in NPI are found over 27 urban agglomerations, 22 of which are situated in developed regions. The opposite changes in NPI between the developed and developing regions could be due to their different environmental management policies during 2005–2016, one stringent and the other less so. The control policies toward ozone precursors, which have been put in place by several

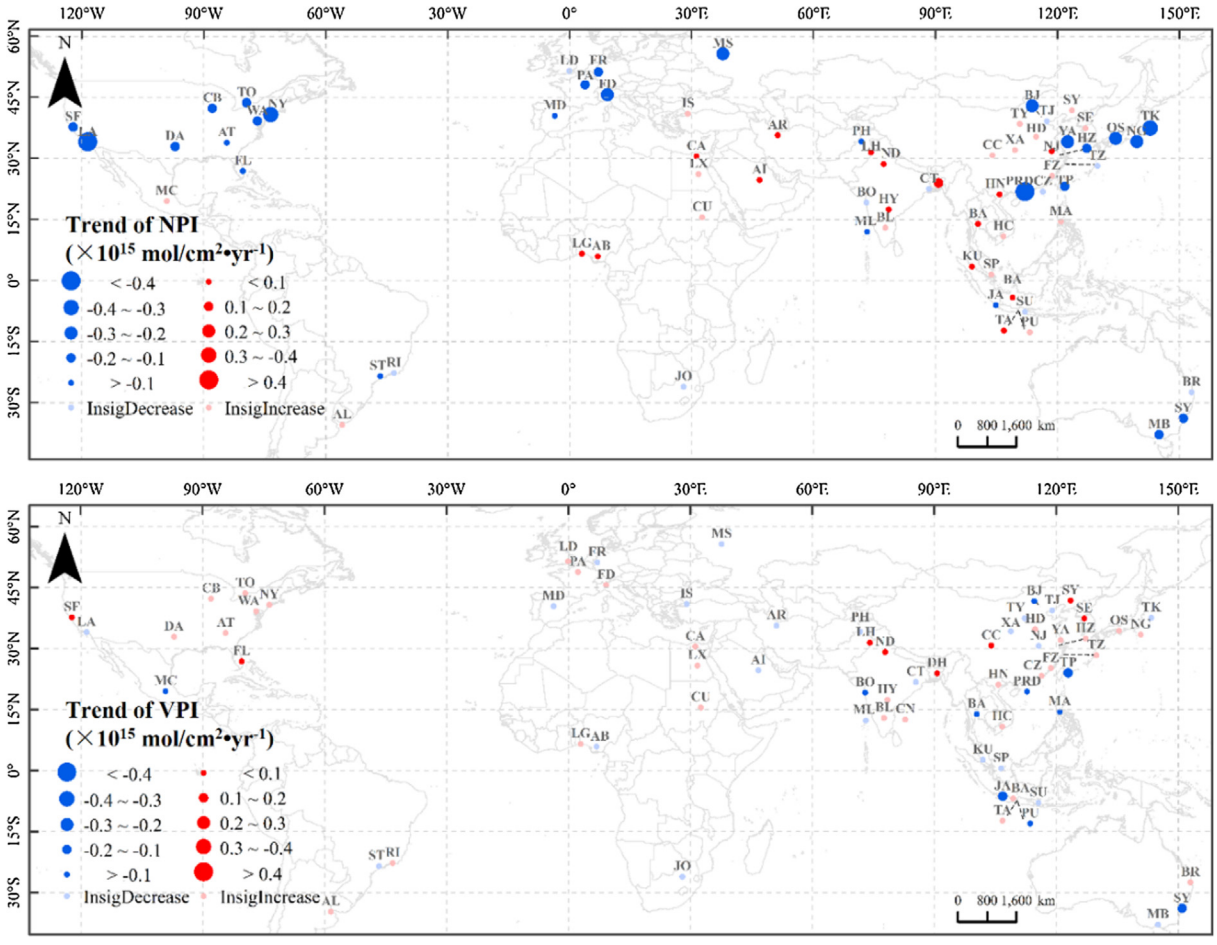

Fig. 4. Trends of NPI and VPI during 2005–2016. Both InsigDecrease and InsigIncrease suggest that the trend is insignificant ($p > 0.05$). The labels represent the abbreviations for the names of the urban agglomerations. For the full names of the urban agglomerations and the values of the PPI, see the Supporting Information Table S1.

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developed urban agglomerations, have effectively lowered the anthropogenic NO_x emissions (Castellanos and Boersma, 2012). In comparison, the anthropogenic NO_x emissions in developing urban agglomerations are generally unrestricted, leading to the increasing anthropogenic NO_x emissions and intensifying NPI (Taubenböck et al., 2009).

With regard to the VPI, the trends are weak and insignificant in most of the urban agglomerations (see Fig. 4). The insignificant trends can be attributed to the following factors. Firstly, given the variety of VOC_s species (Monks et al., 2015), there are more barriers to regulate VOC_s emissions than controlling NO_x emissions. Secondly, the biogenic volatile organic compounds ($BVOC_s$) emitted by terrestrial vegetation, which account for more than 90% of the total VOC_s emissions globally, are essentially uncontrollable (Guenther et al., 2006). Thirdly, the increasing trends in anthropogenic non-methane volatile organic compounds ($NMVOC_s$) emissions can be offset by the decline in $BVOC_s$ induced by the climate and land-use change, leading to the insignificant trends ($p > 0.05$) in VOC_s (Jin and Holloway, 2015).

3.3.2. The trends in chemical regime coverage

With respect to the changes in the coverage of chemical regimes, the disparities between developed and developing regions are also notable (Fig. 5). As a result of the significantly decreasing

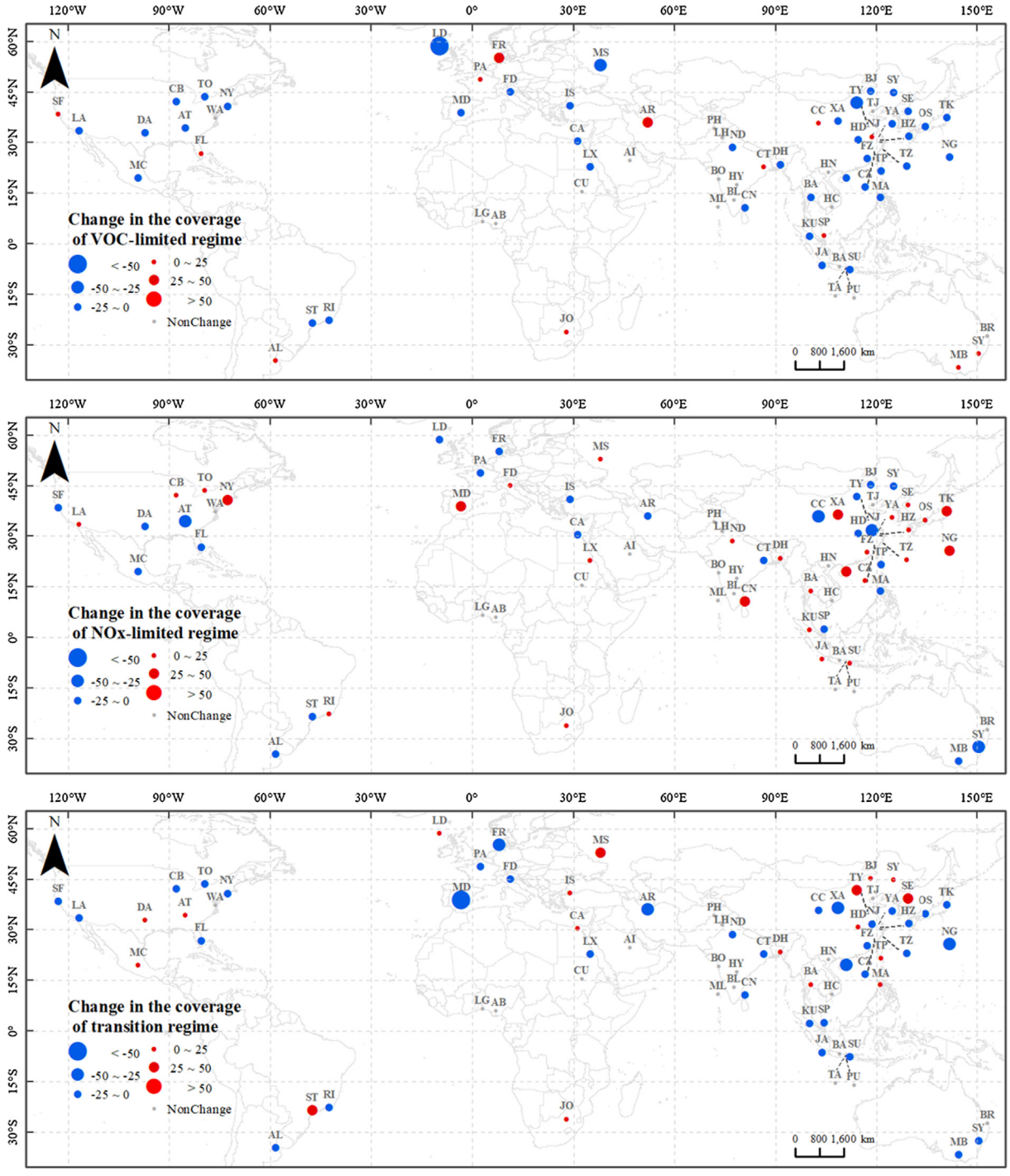

Fig. 5. The changes in CRC between 2005 and 2016. NonChange suggests that there is no change in the CRC during 2005–2016. The labels represent the abbreviations for the names of the urban agglomerations. For the full names of the urban agglomerations and the values of the PPI, see the Supporting Information Table S1.

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NPI but insignificantly increasing VPI, the chemical regimes in developed urban agglomerations have shifted from VOC-limited regime to NO_x-limited regime. Thirteen urban agglomerations experienced the transition from VOC-limited regime to NO_x-limited regime over the study period, most of which are located in developed regions. The most significant change is observed in London, where 80.9% of the VOC-limited regime switched to a NO_x-limited regime. The control measures toward ozone precursors can lead to the shift of chemical regime from VOC-limited regime to NO_x-limited regime, as also locally monitored in different ways, such as model simulation and real-world experiments (Ou et al., 2016; Witte et al., 2011). In contrast, the change toward VOC-limited regime witnessed in developing urban agglomerations is due to the rapid increase in the NPI and the weak trends of the VPI. The shift toward VOC-limited regimes is apparent in 16 agglomerations, 11 of which are in developing regions.

3.4. The usage of CRC, PPI to represent the agglomeration influence

Firstly, this study underscores the advantages of representing the urban agglomeration influence on the chemical regime with the CRC. Due to the heterogeneous chemical regimes within urban agglomerations, previous studies that utilized the average values to evaluate the urban agglomeration influence on the chemical regime might be biased (Choi and Souri, 2015; Jin and Holloway, 2015; Ou et al., 2016; Witte et al., 2011). The average of the chemical regime only reflects the dominant regime within the urban agglomeration, leading to the biased conclusion that most urban agglomerations are NO_x-limited or transition regimes (see Fig. S2). According to the wall-to-wall observations and CRC calculation, the urban agglomerations are generally dominated by the NO_x-limited and transition regimes. In addition, the temporal variation in chemical regime will likely be overlooked if the agglomeration influence is represented as the average of the chemical regime. For example, the VOC-limited regime in Tehran is extending steadily (see Fig. S3 in the SI), while the average shows that the chemical regime of Tehran is a VOC-limited regime throughout 2005–2016 (Choi and Souri, 2015).

Secondly, it is important to distinguish the contribution of the urban agglomerations from their surrounding backgrounds in a temporal analysis. The transmission of air pollution from the background to the urban agglomerations has been shown in previous studies from different perspectives (e.g., air mass analysis, back trajectory analysis, model analysis) (David and Nair, 2013; Varotsos et al., 2014; Wang et al., 2009; Xu et al., 2011). The average precursor concentration of an urban agglomeration consists of the contribution from both the agglomeration and the background (Baklanov et al., 2016). For example, the decreasing trends of the precursors can be offset by the increasing trends in the background (Baklanov et al., 2016; Wang et al., 2009).

Thirdly, these results have significant implications for achieving ozone abatement in urban agglomerations. Although previous studies have addressed ozone abatement measures for a specific chemical regime (Monks et al., 2015), ozone pollution is still challenging, mainly due to the lack of knowledge of the evolution of chemical regimes worldwide. From the results of this study, it has been shown that most urban agglomerations are experiencing the change from VOC-limited regime to NO_x-limited regime, to different degrees. As reported by Ou et al., (2016), since 2015, for the PRD urban agglomeration, a further reduction in VOC_s would not lead to the abatement of ozone, and priority should be given to the controlling of NO_x emissions. It follows that, to take effective measures for ozone pollution control, a timely and reliable knowledge of the chemical regime is of great importance. Furthermore, it may be biased to adopt the same control measures throughout the urban agglomerations, given the heterogeneous regimes within the urban agglomerations. According to the results of this study, it is recommended to take customized measures in each area under a specific chemical regime.

Finally, the comparison between our results and previous studies based on air monitoring stations indicates the advantage of satellite measurements in evaluating the urban agglomeration influence on ozone precursor conditions. Satellite measurements can provide wall-to-wall daily observations of ozone precursors (David and Nair, 2013; Jin and Holloway, 2015; Kar et al., 2010; Mahajan et al., 2015; Safieddine et al., 2013), while the measurements of air monitoring stations are spatially sparse and limited in coverage (Paoletti et al., 2014; Xu et al., 2011). Therefore, the measurements of air monitoring stations cannot represent the photochemical conditions of whole study regions, which may lead to the different conclusions. For example, Jing et al. (2014) demonstrated that the chemical regime of Great Chicago has shifted from NO_x-limited to VOC-limited over 2005–2013, based on the observations of 23 air monitoring stations. However, the emissions inventories for the same period report decreasing NO_x emissions and increasing VOC_s emissions (Jing et al., 2014; Xing et al., 2013), indicating that the regime has moved toward a NO_x-limited regime. Consistent with the results of the emissions inventories (Xing et al., 2013), our results show that the NO_x-limited regime in Chicago is indeed enlarging (see Fig. S4 in the SI).

4. Conclusions

Urbanization has significantly influenced ozone precursor conditions through changing the precursor concentrations and chemical regimes. Recently, attention has been focused toward the influence of urban agglomerations, a newly emerging urban form, on ozone precursor conditions. Satellite observations can provide spatially continuous information about precursor concentrations and chemical regimes over the long term, enabling the comprehensive investigation of the urban agglomeration influence. While previous studies have validated the feasibility of using satellite observations to evaluate urban agglomeration influence on ozone precursor conditions, there have been some inconsistent results. Furthermore, the temporal analyses of urban agglomeration influence are mainly based on the annual average values, which can introduce bias into the conclusions.

Hence, this study aimed to address the aforementioned problems through the systematic investigation of urban agglomerations. Based on a 12-year spatiotemporal analysis of 71 global urban agglomerations, our results showed that: 1) not all urban agglomerations have a positive effect on ozone precursor conditions, and the urban agglomerations that have a negative impact are mainly distributed in the tropics and in areas with an ecological background ($p < 0.05$); 2) the magnitude and the extent of the urban agglomeration influence are aggravated by urbanization and the industrialization process ($p < 0.05$); 3) during 2005–2016, opposite changes in PPI and chemical regime can be observed between developed and developing regions, which is consistent with their disparities in environmental management policies during the study period. In order to effectively abate ozone pollution, a timely knowledge of the CRC and PPI is needed. Overall, this study will provide important insights for future urban agglomeration developments and ozone pollution monitoring with satellite instruments.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.135458.

References

Baek, K.H., Kim, J.H., Park, R.J., Chance, K., Kurosu, T.P., 2014. Validation of OMI HCHO data and its analysis over Asia. Sci. Total Environ. 490, 93–105.
Baklanov, A., Molina, L.T., Gauss, M., 2016. Megacities, air quality and climate. Atmos. Environ. 126, 235–249.
Birch, E.L., Wachter, S.M., 2011. Global urbanization. University of Pennsylvania Press.
Boeke, N.L., Marshall, J.D., Alvarez, S., Chance, K.V., Fried, A., Kurosu, T.P., et al., 2011. Formaldehyde columns from the Ozone Monitoring Instrument: Urban versus background levels and evaluation using aircraft data and a global model. J. Geophys. Res., 116
Boersma, K.F., Eskes, H.J., Dirksen, R.J., van der, A.R.J., Veefkind, J.P., Stammes, P., et al., 2011. An improved tropospheric NO2 column retrieval algorithm for the Ozone Monitoring Instrument. Atmos. Meas. Technol. 4, 1905–1928.
Boersma, K.F.F., Dirksen, R.J., Brunner, D., Zhou, Y., Huijnen, V.V., Eskes, H., et al., 2010. An improved retrieval of tropospheric NO_2 columns from the Ozone Monitoring Instrument. Adv. Mater.
Bell, M.L., Goldberg, R., Hogrefe, C., Kinney, P.L., Knowlton, K., Lynn, B., et al., 2007. Climate change, ambient ozone, and health in 50 US cities. Clim. Change 82, 61–76.
Castellanos, P., Boersma, K.F., 2012. Reductions in nitrogen oxides over Europe driven by environmental policy and economic recession. Sci. Rep. 2, 265.
Choi, Y., Souri, A.H., 2015. Seasonal behavior and long-term trends of tropospheric ozone, its precursors and chemical conditions over Iran: A view from space. Atmos. Environ. 106, 232–240.
Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., et al., 2016. Forty years of improvements in European air quality: regional policy-industry interactions with global impacts. Atmos. Chem. Phys. 16, 3825–3841.
David, L.M., Nair, P.R., 2013. Tropospheric column O3 and NO2 over the Indian region observed by Ozone Monitoring Instrument (OMI): Seasonal changes and long-term trends. Atmos. Environ. 65, 25–39.
De Smedt, I., Stavrakou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., et al., 2015. Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from combined OMI and GOME-2 observations. Atmos. Chem. Phys. 15, 12519–12545.
Duncan, B.N., Yoshida, Y., Olson, J.R., Sillman, S., Martin, R.V., Lamsal, L., et al., 2010. Application of OMI observations to a space-based indicator of NOx and VOC controls on surface ozone formation. Atmos. Environ. 44, 2213–2223.
Friedl, M.A., Sulla-Menashe, D., Tan, B., Schneider, A., Ramankutty, N., Sibley, A., et al., 2010. MODIS Collection 5 global land cover: Algorithm refinements and characterization of new datasets. Remote Sens. Environ. 114, 168–182.
Fu, T.-M., Jacob, D.J., Palmer, P.I., Chance, K., Wang, Y.X., Barletta, B., et al., 2007. Space-based formaldehyde measurements as constraints on volatile organic compound emissions in east and south Asia and implications for ozone. J. Geophys. Res. [Atmos.], 112.
Gardiner, T., Forbes, A., de Mazière, M., Vigouroux, C., Mahieu, E., Demoulin, P., et al., 2008. Trend analysis of greenhouse gases over Europe measured by a network of ground-based remote FTIR instruments. Atmos. Chem. Phys. 8, 6719–6727.
United Nations, 2010. United Nations Framework Convention on Climate Change. Review of European Community & International Environmental Law 1, 270–277.
Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P.I., Geron, C., 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). Atmos. Chem. Phys. 6, 3181–3210.
Guo, H., Ling, Z.H., Cheng, H.R., Simpson, I.J., Lyu, X.P., Wang, X.M., et al., 2017. Tropospheric volatile organic compounds in China. Sci. Total Environ. 574, 1021–1043.
Hodnebrog, Ø., Stordal, F., Berntsen, T.K., 2011. Does the resolution of megacity emissions impact large scale ozone?. Atmos. Environ. 45, 6852–6862.
Huang, G., Brook, R., Crippa, M., Janssens-Maenhout, G., Schieberle, C., Dore, C., et al., 2017a. Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970–2012. Atmos. Chem. Phys. 17, 7683–7701.
Huang, J., Zhou, C., Lee, X., Bao, Y., Zhao, X., Fung, J., et al., 2013. The effects of rapid urbanization on the levels in tropospheric nitrogen dioxide and ozone over East China. Atmos. Environ. 77, 558–567.
Huang, T., Zhu, X., Zhong, Q., Yun, X., Meng, W., Li, B., et al., 2017b. Spatial and Temporal Trends in Global Emissions of Nitrogen Oxides from 1960 to 2014. Environ. Sci. Technol. 51, 7992–8000.
Huang, X., Friedl, M.A., 2014. Distance metric-based forest cover change detection using MODIS time series. Int. J. Appl Earth Obs. 29, 78–92.
Jin, X., Holloway, T., 2015. Spatial and temporal variability of ozone sensitivity over China observed from the Ozone Monitoring. J. Geophys. Res. [Atmos.] 120, 7229–7246.
Jing, P., Lu, Z., Xing, J., Streets, D.G., Tan, Q., O'Brien, T., et al., 2014. Response of the summertime ground-level ozone trend in the Chicago area to emission controls and temperature changes, 2005–2013. Atmos. Environ. 99, 630–640.
Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., et al., 1996. The NCEP/NCAR 40-year reanalysis project. Bull. Am. Meteorol. Soc. 77, 437–472.
Kar, J., Fishman, J., Creilson, J.K., Richter, A., Ziemke, J., Chandra, S., 2010. Are there urban signatures in the tropospheric ozone column products derived from satellite measurements?. Atmos. Chem. Phys. 10, 5213–5222.
Lamsal, L.N., Martin, R.V., Parrish, D.D., Krotkov, N.A., 2013. Scaling relationship for NO2 pollution and urban population size: a satellite perspective. Environ. Sci. Technol. 47, 7855–7861.
Lasaponara, R., 2013. Geospatial analysis from space: Advanced approaches for data processing, information extraction and interpretation. Int. J. Appl Earth Obs 20, 1–3.
Li, K., Jacob, D.J., Liao, H., Shen, L., Zhang, Q., Bates, K.H., 2019. Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China. Proc. Natl. Acad. Sci. 116, 422–427.
Mahajan, A.S., De Smedt, I., Biswas, M.S., Ghude, S., Fadnavis, S., Roy, C., et al., 2015. Inter-annual variations in satellite observations of nitrogen dioxide and formaldehyde over India. Atmos. Environ. 116, 194–201.
Martin, R.V., Fiore, A.M., Van Donkelaar, A., 2004. Space-based diagnosis of surface ozone sensitivity to anthropogenic emissions. Geophys. Res. Lett, 31.
Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., et al., 2015. Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. Atmos. Chem. Phys. 15, 8889–8973.
Novotny, E.V., Bechle, M.J., Millet, D.B., Marshall, J.D., 2011. National Satellite-Based Land-Use Regression: NO2 in the United States. Environ. Sci. Technol. 45, 4407–4414.
Ou, J., Yuan, Z., Zheng, J., Huang, Z., Shao, M., Li, Z., et al., 2016. Ambient Ozone Control in a Photochemically Active Region: Short-Term Despiking or Long-Term Attainment?. Environ. Sci. Technol. 50, 5720–5728.
Paoletti, E., De Marco, A., Beddows, D.C.S., Harrison, R.M., Manning, W.J., 2014. Ozone levels in European and USA cities are increasing more than at rural sites, while peak values are decreasing. Environ. Pollut. 192, 295–299.
Pesaresi, M., Huadong, G., Blaes, X., Ehrlich, D., Ferri, S., Gueguen, L., et al., 2013. A Global Human Settlement Layer From Optical HR/VHR RS Data: Concept and First Results. IEEE. J. STARS 6, 2102–2131.
Pfister, G.G., Wiedinmyer, C., Emmons, L.K., 2008. Impacts of the fall 2007 California wildfires on surface ozone: Integrating local observations with global model simulations. Geophys. Res. Lett, 35.
Rizk HF, 1992. Effect of solar radiation on surface ozone in Cairo.
Safieddine, S., Clerbaux, C., George, M., Hadji-Lazaro, J., Hurtmans, D., Coheur, P.F., et al., 2013. Tropospheric ozone and nitrogen dioxide measurements in urban and rural regions as seen by IASI and GOME-2. J. Geophys. Res. [Atmos.] 118, 10555–10566.
Sagan, V., Pasken, R., Zarauz, J., Krotkov, N., 2018. SO2 trajectories in a complex terrain environment using CALPUFF dispersion model, OMI and MODIS data. Int. J. Appl Earth Obs 69, 99–109.
Schneider, P., Lahoz, W.A., van der, A.R., 2015. Recent satellite-based trends of tropospheric nitrogen dioxide over large urban agglomerations worldwide. Atmos. Chem. Phys. 15, 1205–1220.
Schneider, P., van der, A.R.J., 2012. A global single-sensor analysis of 2002–2011 tropospheric nitrogen dioxide trends observed from space. J. Geophys. Res. Atmos. 117.
Shi, Y., Lau, K.K.-L., Ng, E., 2016. Developing street-level PM2.5 and PM10 land use regression models in high-density hong kong with urban morphological factors. Environ. Sci. Technol. 50, 8178–8187.
Simon, H., Reff, A., Wells, B., Xing, J., Frank, N., 2015. Ozone trends across the United States over a period of decreasing NOx and VOC emissions. Environ. Sci. Technol. 49, 186–195.
Su, J., Jerrett, M., Beckerman, B., 2009. A distance-decay variable selection strategy for land use regression modeling of ambient air pollution exposures. Sci. Total Environ. 407, 3890–3898.
Taubenböck, H., Wegmann, M., Roth, A., Mehl, H., Dech, S., 2009. Urbanization in India – Spatiotemporal analysis using remote sensing data. Comput. Environ. Urban Syst. 33, 179–188.
Taubenböck, H., Wiesner, M., 2015. The spatial network of megaregions - Types of connectivity between cities based on settlement patterns derived from EO-data. Comput. Environ. Urban Syst. 54, 165–180.
Ural, S., Hussain, E., Shan, J., 2011. Building population mapping with aerial imagery and GIS data. Int. J. Appl Earth Obs 13, 841–852.
Varotsos, C., Christodoulakis, J., Tzanis, C., Cracknell, A.P., 2014. Signature of tropospheric ozone and nitrogen dioxide from space: A case study for Athens. Greece. Atmos. Environ. 89, 721–730.

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Vestreng V, Mareckova K, Kakareka S, Malchykhina A, Kukharchyk T, 2007. Inventory review 2007. Emission Data reported to LRTAP Convention and NEC Directive, MSC-W Technical Report. 1.

Wang, Y., Ying, Q., Hu, J., Zhang, H., 2014. Spatial and temporal variations of six criteria air pollutants in 31 provincial capital cities in China during 2013–2014. Environ. Int. 73, 413–422.

Wang, H., Jacob, D.J., Le Sager, P., Streets, D.G., Park, R.J., Gilliland, A.B., et al., 2009. Surface ozone background in the United States: Canadian and Mexican pollution influences. Atmos. Environ. 43, 1310–1319.

Witte, J.C., Duncan, B.N., Douglass, A.R., Kurosu, T.P., Chance, K., Retscher, C., 2011. The unique OMI HCHO/NO2 feature during the 2008 Beijing Olympics: Implications for ozone production sensitivity. Atmos. Environ. 45, 3103–3111.

Wong, M.S., Sarker, M.L.R., Nichol, J., Lee, S-c, Chen, H., Wan, Y., et al., 2013. Modeling BVOC isoprene emissions based on a GIS and remote sensing database. Int. J. Appl Earth Obs 21, 66–77.

Xing, J., Pleim, J., Mathur, R., Pouliot, G., Hogrefe, C., Gan, C.-M., et al., 2013. Historical gaseous and primary aerosol emissions in the United States from 1990 to 2010. Atmos. Chem. Phys. 13, 7531–7549.

Xu, J., Ma, J.Z., Zhang, X.L., Xu, X.B., Xu, X.F., Lin, W.L., et al., 2011. Measurements of ozone and its precursors in Beijing during summertime: impact of urban plumes on ozone pollution in downwind rural areas. Atmos. Chem. Phys. 11, 12241–12252.

Zhang, Y.N., Xiang, Y.R., Chan, L.Y., Chan, C.Y., Sang, X.F., Wang, R., et al., 2011. Procuring the regional urbanization and industrialization effect on ozone pollution in Pearl River Delta of Guangdong, China. Atmos. Environ. 45, 4898–4906.

Zhu, L., Jacob, D.J., Mickley, L.J., Marais, E.A., Cohan, D.S., Yoshida, Y., et al., 2014. Anthropogenic emissions of highly reactive volatile organic compounds in eastern Texas inferred from oversampling of satellite (OMI) measurements of HCHO columns. Environ. Res. Lett. 9, 114004.

Zhu, L., Mickley, L.J., Jacob, D.J., Marais, E.A., Sheng, J., Hu, L., et al., 2017. Long-term (2005–2014) trends in formaldehyde (HCHO) columns across North America as seen by the OMI satellite instrument: Evidence of changing emissions of volatile organic compounds. Geophys. Res. Lett. 44, 7079–7086.

Ziemke, J.R., Chandra, S., Duncan, B.N., Froidevaux, L., Bhartia, P.K., Levelt, P.F., et al., 2006. Tropospheric ozone determined from Aura OMI and MLS: Evaluation of measurements and comparison with the Global Modeling Initiative's Chemical Transport Model. J. Geophys. Res., 111

Please cite this article as: J. Li, Y. Gao and X. Huang, The impact of urban agglomeration on ozone precursor conditions: A systematic investigation across global agglomerations utilizing multi-source geospatial datasets, Science of the Total Environment, https://doi.org/10.1016/j.scitotenv.2019.135458