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Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects

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Abstract

High concentrations of ozone in urban and industrial regions worldwide have long been a major air quality issue. With the rapid increase in fossil fuel consumption in China over the past three decades, the emission of chemical precursors to ozone—nitrogen oxides and volatile organic compounds—has increased sharply, surpassing that of North America and Europe and raising concerns about worsening ozone pollution in China. Historically, research and control have prioritized acid rain, particulate matter, and more recently fine particulate matter ($PM_{2.5}$). In contrast, less is known about ozone pollution, partly due to a lack of monitoring of atmospheric ozone and its precursors until recently. This review summarizes the main findings from published papers on the characteristics and sources and processes of ozone and ozone precursors in the boundary layer of urban and rural areas of China, including concentration levels, seasonal variation, meteorology conducive to photochemistry and pollution transport, key production and loss processes, ozone dependence on nitrogen oxides and volatile organic compounds, and the effects of ozone on crops and human health. Ozone concentrations exceeding the ambient air quality standard by 100-200% have been observed in China's major

urban centers such as Jing-Jin-Ji, the Yangtze River delta, and the Pearl River delta, and limited studies suggest harmful effect of ozone on human health and agricultural corps; key chemical precursors and meteorological conditions conductive to ozone pollution have been investigated, and inter-city/region transport of ozone is significant. Several recommendations are given for future research and policy development on ground-level ozone.

1. Introduction

Ozone (O_3) in the troposphere plays a central role in the oxidation of chemically and climatically relevant trace gases, thereby regulating their lifetime in the atmosphere. As a strong oxidant, O_3 at ground level is detrimental to human health and vegetation. Tropospheric O_3 is also the third most important greenhouse gas. Because of its importance to air quality and climate change, O_3 has received continuous attention in the past three decades from both the scientific and regulatory communities (e.g., Monks et al., 2015; NARSTO, 2000; NRC, 1991).

Photochemical smog—characterized by elevated concentrations of O_3, other gases, and particulates—results from chemical reactions between nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight (NRC, 1991). This type of air pollution, first discovered in the 1950s in Los Angeles, has been found in major urban and industrial regions throughout the world. Extensive research, mostly in North America, has investigated the chemical and meteorological processes responsible for ozone formation and transport, and these findings have been comprehensively reviewed (Hidy, 2000; Jenkin and Clemitshaw, 2000; Kleinman, 2000; NARSTO, 2000; NRC, 1991; Solomon et al., 2000). Although research on urban ozone pollution began in the early 1980s in a western city, little systematic research and coordinated ozone monitoring was performed in China until the mid-2000s, partly because research and control efforts during that period were focused on sulfur (acid rain) and particulate matter. Nonetheless, intensive field measurements have revealed very high concentrations of ozone in or near some large Chinese cities. For instance, an hourly mixing ratio of up to 286 ppbv was observed in summer 2005 at a rural mountain site north of Beijing (Wang et al., 2006a), and summer peak ozone concentrations increased from 1980 to 2003 at a sub-urban (now urban) site in Beijing (Shao et al., 2006). In the past decade, particularly the past 5 years,

extensive ozone monitoring has been carried out in urban and rural locations by national and local environmental and meteorological agencies. The available data reveal that ambient ozone concentrations in major urban areas have continued to increase despite recent reductions in the emissions of SO_2 (since 2006) and NO_x (since 2011) (http://www.mep.gov.cn). According to monitoring results from 74 Chinese cities, the mean daily 8-hr maximum concentrations increased from approximately 69.5 ppbv in 2013 to approximately 75.0 ppbv in 2015 while the percentage of non-compliant cities increased from 23% to 38%, whereas the metrics on other pollutants improved from 2013 to 2015 (China Environment Report 2014 and 2015, available at http://www.mep.gov.cn, in Chinese). It has been suggested that elevated ozone levels in China adversely affect agricultural crops (Chameides et al., 1999; Feng et al., 2015 and references therein) and human health (Brauer et al., 2016; Li et al., 2015b). Current projections indicate that ozone pollution is likely to worsen in future (Wang et al., 2013). A review of known factors that determine ozone formation and distribution in China is therefore needed to aid in formulating a mitigation policy and to guide future research.

This review focuses on ground-level ozone in urban and polluted rural areas of mainland China and Hong Kong, and is structured as the follows. Section 2 gives a brief review of chemical mechanisms for ozone formation; Section 3 reviews the field measurements of ozone and ozone precursors; Section 4 summarizes the typical meteorological conditions associated with high ozone events; Section 5 examines the use of observations to identify ozone formation regimes, including several indicators and observation-constrained model studies; Section 6 reviews the emission-based model results on the sources of ozone; Section 7 covers the effects of ozone on agricultural corps and human health; and Section 8 provides a summary and some recommendations for future research and control. The review focuses on findings which have been published in English-language literature.

2. Brief review of ozone formation mechanism

This section gives a condense review of the chemical mechanisms of ozone production and loss and non-linear chemistry to lay down the foundation for discussion of the studies of the complex relationship between ozone and its precursors. Ozone in the Earth's atmosphere is

ultimately formed from the combination reaction of atomic oxygen (O^3P) and molecular oxygen (O_2) (*R1*). In the stratosphere, photolysis of O_2 by the short-wavelength ultraviolet (UV) radiation ($\lambda \leq 240$ nm) supplies atomic oxygen and facilitates the formation of the O_3 layer (Chapman, 1930). In the troposphere with little UV radiation, photolysis of NO_2 at wavelengths ≤ 424 nm (*R2*) becomes the primary source of O^3P atoms and prompts O_3 formation. Once formed, O_3 readily reacts with NO to regenerate NO_2 (*R3*). The *R1*-*R3* reactions result in a null cycle when no other chemical species are involved.

$$O(^3P) + O_2 + M \rightarrow O_3 + M \qquad (R1)$$

$$NO_2 + h\nu \rightarrow NO + O(^3P) \qquad (R2)$$

$$O_3 + NO \rightarrow NO_2 + O_2 \qquad (R3)$$

However, in reality, the troposphere contains alternative oxidants (i.e., HO_2 and RO_2) that efficiently convert NO to NO_2 (*R4* and *R5*), resulting in the accumulation of O_3. The *R4*, *R5*, and *R2* reactions establish an efficient "NO_X cycle" that produces O_3 without consumption of NO_X (see Figure 1). The other important chemistry cycle that affects O_3 formation is the so-called "RO_X (RO_X = OH + HO_2 + RO_2) radical cycle" that continuously supplies HO_2 and RO_2 to oxidize NO to NO_2. It generally starts from the OH-initiated degradation of VOCs that produce RO_2 radicals (*R7*), followed by *R5* converting RO_2 to RO, *R7* converting RO to HO_2, and finally *R4* to regenerate OH from HO_2. Each RO_X cycle oxidizes two molecules of NO to NO_2, which then produces two molecules of O_3 through the "NO_X cycle" and recycles NO. The coupling of both chemistry cycles and the photochemical formation of O_3 are illustrated in Figure 1.

$$HO_2 + NO \rightarrow OH + NO_2 \qquad (R4)$$

$$RO_2 + NO \rightarrow RO + NO_2 \qquad (R5)$$

$$OH + RH + O_2 \rightarrow RO_2 + H_2O \qquad (R6)$$

$$RO + O_2 \rightarrow HO_2 + \text{carbonyls} \qquad (R7)$$

The RO_X and NO_X cycles are terminated by the cross reactions of RO_X and/or NO_X. At high NO_X conditions, the termination process is dominated by the reactions of NO_2 with OH (*R8*) and RO_2 (*R9*), which form nitric acid and organic nitrates (the so-called NO_Z species). At low NO_X conditions, the dominant termination processes are self-reactions of HO_2 (*R10*) and cross-reactions of HO_2 and RO_2 (*R11*), producing hydrogen peroxides (H_2O_2) and organic peroxides. As such, the relative abundances of NO_Z and peroxides (e.g., H_2O_2/HNO_3) can reflect the ambient atmospheric conditions, i.e., low-NO_X or high-NO_X, and are usually adopted as indicators to infer the O_3 formation regimes (*e.g.*, NO_X-limited or VOC-limited; see Section 5.1).

$$OH + NO_2 \rightarrow HNO_3 \qquad (R8)$$

$$RO_2 + NO_2 \leftrightarrow RO_2NO_2 \qquad (R9)$$

$$HO_2 + HO_2 \rightarrow H_2O_2 + O_2 \qquad (R10)$$

$$HO_2 + RO_2 \rightarrow RO_2H + O_2 \qquad (R11)$$

Another key process of O_3 formation is the primary production of RO_X radicals from the closed-shell molecules. It initiates the aforementioned RO_X cycle and hence plays a central role in ozone production. In the polluted troposphere, the RO_X radicals arise mainly from the photolysis of O_3, HONO, and carbonyls, but also from the ozonolysis reactions of unsaturated VOCs, and the relative contribution of each source may vary from one place to another (Xue et al., 2016). New sources of atmospheric radicals (and radical precursors) have been revealed, including unknown daytime source(s) of HONO (e.g., Kleffmann, 2007) and the nocturnal formation of nitryl chloride ($ClNO_2$) with subsequent release of chlorine atoms the next day. Reactions between Cl and VOCs enhance the photochemical formation of ozone via a gas-phase mechanism similar to that of OH (e.g., Riedel et al., 2014).

$$RH + Cl + O_2 \rightarrow RO_2 + HCl \qquad (R12)$$

A common feature of O_3 formation is the non-linear dependence of O_3 production on its precursors, i.e., NO_X and VOCs. With low NO_X/VOCs, the intensity of the "NO_X cycle" is

weaker than that of the "RO_X cycle", and hence it becomes the limiting factor in O_3 production. Such a scenario is commonly known as the NO_X-limited O_3 formation regime. With high NO_X/VOCs, in contrast, O_3 production is mainly limited by the intensity of the "RO_X cycle", and is known as VOC-limited. If NO levels are much higher, the usual case in polluted urban zones, O_3 production is suppressed by the *R3* reaction and falls into a "NO_X titrated regime." Therefore, identifying the O_3 formation regime is a fundamental step in the science-based regulation of O_3 air pollution and has been a major area of O_3 pollution research in China (e.g., Wang et al., 2010; Xue et al., 2014b; Zhang et al., 2007; Zhang et al., 2008c).

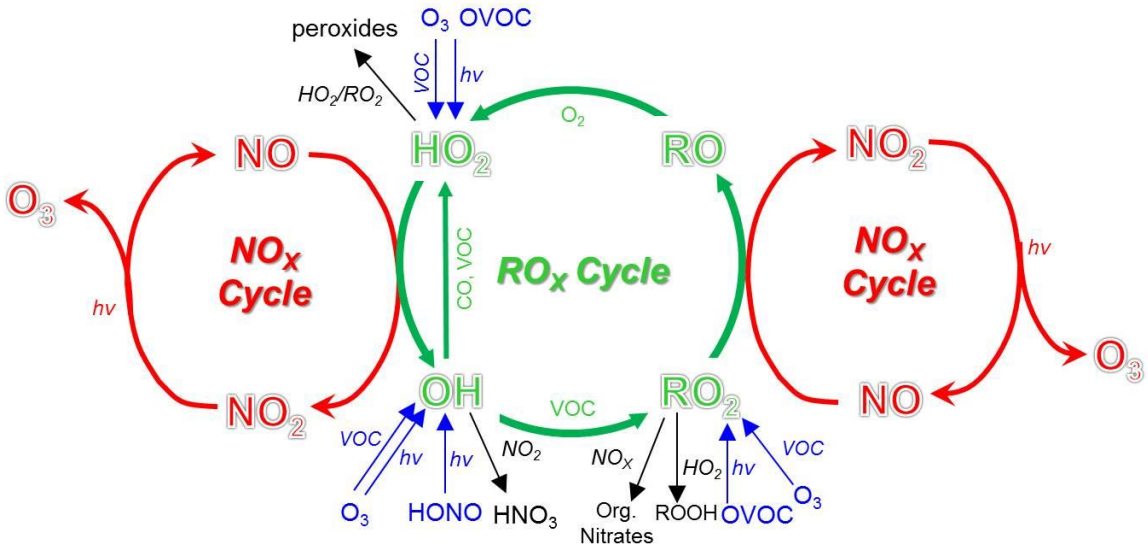

Figure 1. Sketch of photochemical ozone formation mechanism and coupling of "RO_X cycle" and "NO_X cycle." Reaction pathways shown in red, green, blue, and black refer to "NO_X cycle," "RO_X cycle," radical initiation, and termination processes, respectively.

3. Field measurements of ozone and ozone precursors in China

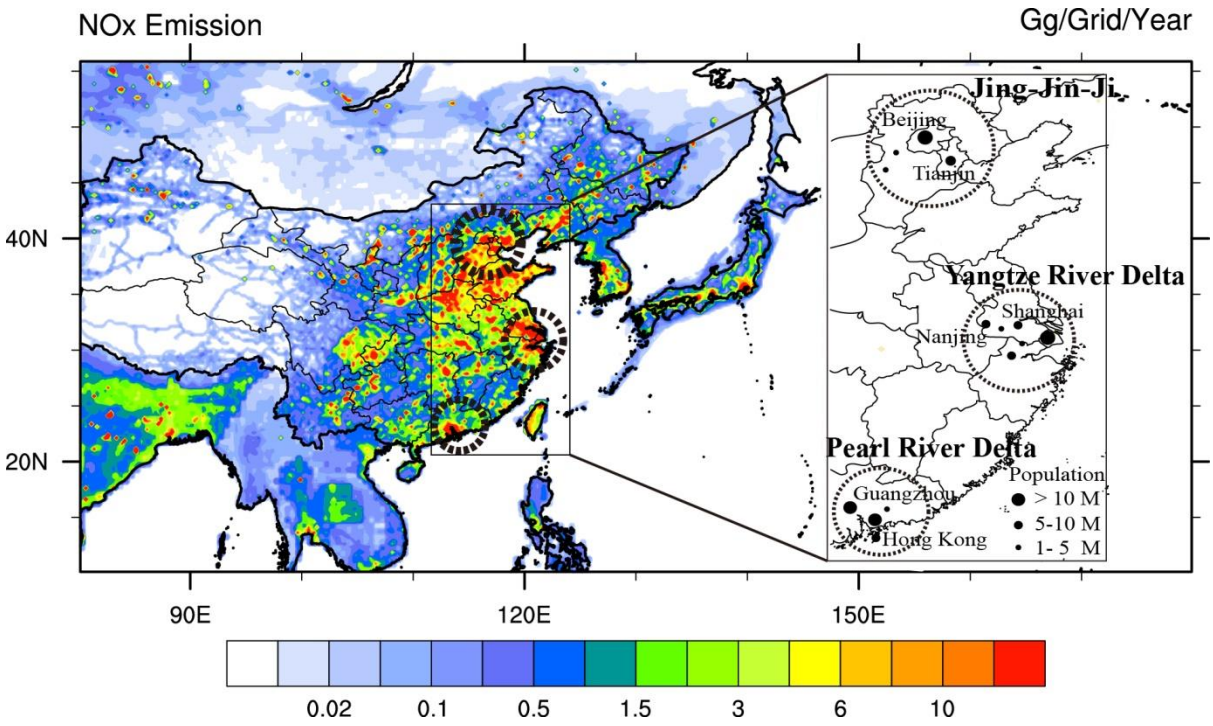

Figure 2. Map of China showing the three most developed regions—Jing-Jin-Ji (Beijing-Tianjin-Hebei), Yangtze River Delta (including Shanghai), and Pearl River Delta (including Guangzhou and Hong Kong–Shenzhen)—and NOx emission intensity for 2013 with resolution of 0.25˚ × 0.25˚. Emission data is from Tsinghua University available at http://meicmodel.org/. Also shown are major cities in three regions.

The majority of the Chinese population lives in the eastern part of China, especially in the three most developed regions, "Jing-Jin-Ji" (Beijing-Tianjin-Hebei), the Yangtze River Delta (YRD; including Shanghai-Jiangsu-Zhejiang-Anhui), and the Pearl River Delta (PRD; including Guangzhou, Shenzhen, and Hong Kong). These regions consistently have the highest emissions of anthropogenic precursors (Figure 1), which have led to severe region-wide air pollution. As indicated earlier, ozone measurements before year 2000 were scarce in mainland China. Although ozone was included in China's first ambient air quality standards in 1996 (which adopted an hourly concentration of 240 μg/m^3 [~112 ppbv at 273 K, 101.3 kPa] for urban and industrial areas), it was not regularly monitored or reported by environmental agencies until 2012, after China revised its O_3 standards to hourly and 8-hourly maximum values of 200 μg/m^3 and 160 μg/m^3, respectively (http://www.mep.gov.cn). The China Meteorological Administration (CMA) began to continuously monitor surface ozone in 2005 at its three regional background air monitoring stations (i.e., Longfengshan in Heilongjiang province in

the northeast, Shangdianzi to the north of Beijing, and Lin'an to the southwest of Shanghai). Historical ozone data from the two government networks currently are not openly accessible, although partially quality controlled hourly data are being reported in real-time online by the China Environmental Monitoring Center (http://www.cnemc.cn).

In Hong Kong, the Hong Kong Environmental Protection Department (HKEPD) began to measure ozone at several air quality monitoring stations in early 1990s, but only in densely populated urban areas. In 1993, the Hong Kong Polytechnic University established the first regional station (Hok Tsui) in South China, which has been operational ever since (Wang et al., 2009a). In 1997 and 1999, two non-urban stations were set up by the HKEPD, one to the northeast and the other to the southwest of the city center. Hourly data from this network are available at http://epic.epd.gov.hk/EPICDI/air/station/. The Hong Kong Observatory has been launching ozonesonde since 1993 initially on a monthly basis (with enhanced weekly releases in 1993-1994 and 2000-2001), and the frequency was increased to weekly release since April 2003 (http://www.weather.gov.hk/publica/reprint/r1173.pdf). The data are publically assessable at the World Ozone and Ultraviolet radiation Center (http://woudc.org/data/stations/?id=344).

Because of the lack of nationwide ozone monitoring data in earlier years, the ozone pollution situation can only be discerned from limited campaign-type measurements. Table 1 a-d summarizes the ozone field studies in the three most developed regions and other areas of mainland China and Hong Kong. It should be noted that although the references listed (a total of 91) may not be exhaustive and do not cover Chinese-language literature, we believe that they should represent comprehensive field studies in mainland China. The first study of photochemical pollution in China was conducted in the early 1980s in Lanzhou, a city situated in a valley in western China with a large petrochemical facility (Tang et al., 1989). Ozone was measured sporadically in Beijing during the summers of 1982 through 2003 (Shao et al., 2006) and in Guangzhou in southern China (Zhang et al., 1998). In Lin'an, ozone (and ozone precursors) was measured for a full year in 1994 (Yan et al., 1997) and again from 1999 to 2010, as part of the China Map project (Guo et al., 2004b; Wang et al., 2002; Wang et al.,

193 2001a). In Hong Kong, aircraft measurements of ozone and several pollutants were taken in
194 1994 along the Hong Kong border (Kok et al., 1997); surface measurements of ozone, ozone
195 precursors and other pollutants were taken at a pollution-receptor site in western Hong Kong
196 in 2001 and 2002 (Guo et al., 2006; Wang et al., 2003; Zhang et al., 2007). In the summers of
197 2004, 2005, and 2006, ozone and ozone precursors were measured downwind of four major
198 cities (Beijing, Shanghai, Guangzhou, and Lanzhou) (Wang et al., 2006a; Xue et al., 2014a;
199 Zhang et al., 2009). A major international field program was also conducted during the fall of
200 2004 in the PRD region (Zhang et al., 2008b and references therein).

201 Since 2005, the number of photochemical studies has increased drastically in mainland China,
202 especially in the PRD region in the south, the Jing-Jin-Ji region in the north, and the YRD
203 region in the east. Many of these studies were international joint efforts, some of which were
204 conducted to improve air quality for the Beijing 2008 Summer Olympics. The findings of these
205 studies are given in the sections below.

Table 1a. Summary of field measurements of ozone and ozone precursors in the Jing-Jin-Ji region.

Site	Measurement period	Type	Ozone precursors	Maximum O_3 (ppbv)	Reference
Zhongguancun, Beijing	1982-2003 (summer, sporadic)	Suburban	/	>200	Shao et al. (2006)
Shangdianzi, Beijing	Sep 2003-Dec 2006	Rural	NOx (NO+NO_2), CO	~175	Meng et al. (2009); Lin et al. (2008)
	2004-2015				Ma et al. (2016)
Changping, Beijing	Jun-Jul 2005 (downwind)	Rural	NO, CO, NMHCs	286	Wang et al. (2006a); Wang et al. (2010)
	Jul-Aug 2008 (downwind)		NOx, CO, NMHCs		Wang et al. (2010)
Miyun, Beijing	Jun-Aug 2006, 2007, 2008	Rural	NO, CO	~170	Wang et al. (2009c)
PKU[a], Beijing	Aug-Sep 2006	Urban	NO, NO_2, VOCs	123	Chou et al. (2009) Shao et al. (2009a)
	Aug-Sep 2008		NO, NO_2, CO, NMHCs		Chou et al. (2011)
Tianjin	Sep-Oct 2006	Urban	NO, NO_2	117	Han (2011)
Gucheng, Hebei	Jul 2006-Sep 2007	Rural	NO, NO_2, CO	162	Lin et al. (2009)
Aircraft measurement, NCP[a]	2007-2010	Regional	NO, NO_2,	60-120	Chen et al. (2013)

Multiple, Beijing	Jun-Sep 2007	Urban-Rural, 4 sites	NO, NO_2, CO, NMHCs	171-275	Xu et al. (2011)
CMA[a], Beijing	Nov 2007- Mar 2008	Urban	NO, NO_2, CO	70	Lin et al. (2011)
Multiple, Beijing	Jun-Aug 2008	Urban-Rural, 2 sites	NO, NO_2, CO	~150-180	Ge et al. (2012)
Aoyuncun, Beijing	Jul-Aug 2008	Urban	NO, NO_2	180	Gao and Zhang (2012)
Multiple, Beijing	Jul-Aug 2008	Urban-Rural, 3 sites	CO, NOx, NMHCs	190	Wang et al. (2010)
IAP, CAS[a], Beijing	Jul-Sep 2008	Urban	NO, NO_2	128	Sun et al. (2011)
Wuqing, Tianjin	Jul-Aug 2009	Urban	NO, NO_2, VOCs	~200	Ran et al. (2012)
Multiple, Beijing	Jul 2010-Aug 2011	Urban-Suburban, 4 sites	NO, NO_2, CO, VOCs	175	Wei et al. (2015)

[a] PKU: Peking University; NCP: Northern China Plain; CMA: China Meteorological Administration; IAP, CAS: Institute of Atmospheric Physics, Chinese Academy of Sciences;

Table 1b. Summary of field measurements of ozone and ozone precursors in the YRD region.

Site	Measurement period	Type	Ozone precursors	Maximum O_3 (ppbv)	Reference
Shanghai	1990-1994	Urban	NOx	~100	Xu et al. (1997)
Lin'an	1991-2006 (non- continuous)	Rural	NOx	156	Xu et al. (2008)
	Aug 1994-Aug 1995		NO, NO_2,VOCs	112	Luo et al. (2000)
	Jun1999-Apr 2001		NOx, CO, NMHCs	145	Cheung and Wang (2001); Wang et al. (2001a); Wang et al. (2002); Wang et al. (2004); Guo et al. (2004b)
Multiple, YRD	May 1999-Jun 2000	Rural, 6 sites	NO, NO_2, CO	116	Wang et al. (2005)
Nanjing	Jan 2000-Feb 2003	Urban	NO, NO_2, CO, NMHCs	125	Tu et al. (2007)
	Aug 2011-Jul 2012	Suburban	NO, CO	~177	Ding et al. (2013)
	Jun 2013-Aug 2013	Urban, 4 sites	NO, NO_2, CO, VOCs	135	An et al. (2015)
SEMC [a], Shanghai	Jan 2001-Dec 2004	Urban	NO_2	~126 (8-hr)	Zhang et al. (2006)
	Mar 2004-Dec 2005				Huang et al. (2009)
Taicang, Jiangsu	May-Jun 2005	Rural	NO, NO_2, CO, NMHCs	127	Xue et al. (2014b)

Downtown, Shanghai	Jun 2006-Jun 2007	Urban	NOx, CO, NMVOCs	128	Ran et al. (2009)
Aircraft measurement, YRD	30 Sep-11 Oct 2007	Regional	NOx, CO, VOCs	60	Geng et al. (2009)
Fudan University, Shanghai	Jan-Dec 2013	Urban	NO_2, CO	179	Shi et al. (2015)

[a] SEMC: Shanghai Environmental Monitoring Center.

Table 1c. Summary of field measurements of ozone and ozone precursors in the PRD region.

Site	Measurement period	Type	Observed ozone precursors	Maximum O_3 (ppbv)	Reference
Hok Tsui, Hong Kong	1994-2007	Rural	NOx ,CO, VOCs[a]	~200	Wang et al. (1998); Wang et al. (2003); Wang et al. (2009a)
Multiple, Hong Kong	1994-1999	Urban-Rural, 10 sites	NO_2	167	Lee et al. (2002)
Aircraft measurement, Hong Kong	Oct-Nov 1994	Regional	NO, CO	115	Kok et al. (1997)
Tung Chung, Hong Kong [a]	2001- 2013	Suburban	NO, NO_2, CO, NMHCs	~200	Zhang et al. (2007); Guo et al. (2009); Cheng et al. (2010); Xue et al. (2014a);); Xue et al. (2016); Zhang et al. (2016); Guo et al. (2014)
Tai O, Hong Kong	Aug 2001-Dec 2002	Rural	CO, NO, NMHCs	203	Wang et al. (2003); Wang and Kwok (2003); Zhang et al. (2007); Guo et al. (2006)
Aircraft measurement, PRD	Oct 2004	Regional	NOx	~100	Wang et al. (2008)
Xinken, Guangzhou	Oct-Nov 2004	Rural	NO, NO_2, CO, NMHCs	163	Zhang et al. (2008b)
Downtown, Guangzhou	Oct-Nov 2004	Urban	NO, NO_2, CO, NMHCs	~210	Zhang et al. (2008b)
	Jul 2006				Lu et al. (2010b)
Wanqingsha, Guangzhou	Apr-May 2004	Suburban	NOx, NMHCs	212	Xue et al. (2014b)
	Oct 2004		NO, NO_2, CO, NMHCs,		Zhang et al. (2008a)
	Oct-Dec 2007		NO, CO, NMHCs		Guo et al. (2009); Cheng et al. (2010)
	Nov-Dec 2007, Nov-Dec 2008		NO, NO_2, CO, NMHCs,		Zhang et al. (2012)

Multiple, PRD	2006-2007	Urban–Rural, 16 sites	NO, CO	31 (monthly)	Zheng et al. (2010)
Backgarden, Qiangyuan	Jul 2006	Suburban	NO, NO_2, CO, NMHCs	~200	Lu et al. (2010b); Lu et al. (2012)
Tai Mo Shan, Hong Kong	Oct.-Nov. 2010	Suburban mountain top	NO, NO_2, CO, NMHCs	~118	Guo et al. (2012)
	Nov- Dec. 2013				Wang et al. (2016); Brown et al. (2016)

[a] Regular monitoring station with intensive studies.

Table 1d. Summary of field measurements of ozone and ozone precursors in other regions of China.

Site	Measurement period	Type	Observed ozone precursors	Maximum O_3 (ppbv)	Reference
Multiple, Chongqin	1993-1996	Urban-Rural, 20 sites		93	Zheng et al. (1998)
Longfengshan, Heilongjiang	Aug 1994-Jul 1995	Rural	NOx, CO, NMHCs	~85	Li et al. (1999)
Waliguan, Qinghai (Global baseline station with intensive studies)	Aug 1994-Dec 1995	Remote continental	NOx, CO, NMHCs	130	Li et al. (1999)
	Aug 1994- Dec 2013		/	~65 (monthly)	Xu et al. (2016)
	Apr-May 2003, Jul-Aug 2003		NO, CO, VOCs	~80	Wang et al. (2006b); Xue et al. (2014b)
	Jun-Jul 2006		CO	91	Zhang et al. (2009)
	Jul-Aug 2006		NO, NO_2, CO	~62	Xue et al. (2011)
Mt. Tai, Shandong	Jul-Nov 2003	Rural mountain top	CO	160	Gao et al. (2005)
	Mar 2004-May 2007		NO, NO_2, CO		Kanaya et al. (2013)
	May 2004		/		Wang et al. (2006c)

	Jun. 2006		NO, NOx, CO, NMVOCs		Kanaya et al. (2009); Kanaya et al. (2013); Li et al. (2007)
	Jun 2006-Jun 2009 Jun-Aug 2014 Jun-Aug 2015		NO, NO_2, CO		Sun et al. (2016)
Ji'nan，Shandong	Jun 2003-Oct 2004	Urban	/	144	Shan et al. (2009a); Shan et al. (2009b)
Mt. Huang, Anhui	May 2004	Rural mountain top	/	114	Wang et al. (2006c)
Lanzhou, Gansu	Jun-Jul 2006	Suburban	NOx, CO, NMHCs	161	Zhang et al. (2009); Xue et al. (2014b)
	Jun-Jul 2013	Urban-Suburban, 2 sites	NOx, CO NMHCs	186	Jia et al. (2016)
Aircraft measurement, Jilin	Jun-Jul 2007	Regional	CO, VOCs	~140	Ding et al. (2009)
Xi'an, Shan'xi	Mar 2008-Jan 2009	Urban	/	140	Wang et al. (2012a)
Wuhan, Hubei	Feb 2013-Oct 2014	Urban	NOx, CO, VOCs	~85 (daytime average)	Lyu et al. (2016)

According to the published data, the Beijing area has the highest peak ozone concentrations of the three most developed regions. Wang et al. (2006a) conducted field measurements for 6 weeks at a rural site 50 km north of Beijing city center in June and July 2005 and frequently observed hourly O_3 close to 200 ppbv; the highest value was 286 ppbv, a level unsurpassed in the available literature. Even during the first 2 weeks of emission control for the Beijing Olympic Games, hourly ozone mixing ratios in the range of 160 to 180 ppbv were observed in urban Beijing (Wang et al., 2010). In comparison, the highest hourly O_3 level reported in the YRD region was 140 to 167 ppbv, and hourly O_3 mixing ratios of up to 200 to 220 ppbv were reported in the PRD region, which includes Hong Kong (Zhang et al. (2007); Guangdong EPD reports, available at: http://www.gdep.gov.cn/hjjce/kqjc/). Data from the national networks, once available, would give a full picture of the severity of ozone pollution across the country.

Seasonal variations in ozone pollution in the three regions are clearer. In the north and in the YRD, the highest seasonal mean ozone occurs from late May to July (Ding et al., 2013; Ding et al., 2008; Li et al., 2007; Wang et al., 2001a; Xu et al., 2008), like many other mid-latitude locations in the Northern Hemisphere. In contrast, the PRD region peaks in the fall (October) (Wang et al., 2009a; Zheng et al., 2010). This feature can be explained by the interplay of chemistry and meteorology that contributes to a different seasonal maximum of ozone in southern China than in the north. The different seasonal peaks of surface ozone have implications for assessment of its effects on crops.

Long-term (>10 years) changes of ozone pollution have been reported based on limited data. In Hong Kong, where a continuous record is available, both background and urban ozone (O_x = O_3 + NO_2 in urban areas) have increased since the early 1990s (at a rate of 0.27 to 0.58 ppbv/year) (Wang et al., 2009a; Xue et al., 2014a). In the PRD, O_3 was found to increase at a rate of 0.86 ppbv/year from 2006 to 2011 (Li et al., 2014). In the YRD region, the mean monthly highest 5% ozone at Lin'an increased at a rate of 1.8 ppbv/year during 1991-2006 (Xu et al., 2008). In the North China plain (NCP), aircraft data from the MOZAIC program showed a large increase (2%/year) in the summertime boundary-layer ozone during 1995-2005; the surface daily 1-hr maximum ozone in urban Beijing also increased at rate of 1.3%/year during

2001-2006 (Tang et al., 2009) and the daily 8-hr maximum O_3 at rural Shangdianzi rose at a rate of 1.1 ppbv/year during 2003-2015 (Ma et al., 2016). Non-continuous measurements at Mt. Tai, the highest peak in the NCP, showed an increase of 1.7-2.1%/year during the summer months from 2003 to 2015 (Sun et al., 2016). All of these results point to worsening photochemical pollution in China's major developed regions. According to available emission and satellite data (Duncan et al., 2016; http://data.stats.gov.cn/; Krotkov et al., 2016; Kurokawa et al., 2013; Mijling et al., 2013; Ohara et al., 2007; Richter et al., 2005; Streets et al., 2001; Zhao et al., 2013), NOx emission increased since 1980s, peaked in 2011 and decreased since in the three major regions due to implementation of NOx control in China's 12th Year Plan (2011-15) and a slowdown in manufacturing activities. Emission of VOCs have also increased in mainland China since the 1980s (Lu et al., 2013; Ohara et al., 2007), with no turning point up to 2015 according to satellite data on formaldehyde vertical column (De Smedt et al., 2015). For Hong Kong which is located in the PRD region, NOx and VOC emissions have been reduced by 28% and 65%, respectively, during 1997-2014 (http://www.epd.gov.hk/). The paradox increase in ambient ozone levels in Hong Kong have been attributed largely to the increasing concentrations in air transported into the territory (Xue et al., 2014a). It will be of great interest to see whether the continuing NOx emission reduction and long-awaited VOC control in China will affect the ambient ozone levels in the near future.

The ozone precursors—NOx (NO + NO_2) and VOCs, including CO, non-methane hydrocarbons (NMHC), and oxygenated organics—were measured initially in ad hoc field studies. With large increases in research funding and increasing awareness of the importance of VOCs, speciated VOCs have been measured in many recent field studies and also by governments' agencies. The ozone precursor data and other concurrently measured pollutants have been used to characterize site environments, identify sources, elucidate ozone-forming potentials and regimes, apportion sources of VOCs, and validate model simulations. For instance, field measurements at Lin'an revealed that biomass burning (biofuel and crop residues) was an important source of ozone precursors in rural areas in the YRD region (Cheung and Wang, 2001; Guo et al., 2004b). Reactive aromatics were found to dominate ozone production in Hong Kong and in the PRD region (So and Wang, 2004; Xue et al., 2014a;

Xue et al., 2014b; Zhang et al., 2007; Zhang et al., 2008c), and source apportionment studies show that solvent use and vehicle exhausts are the main source of these compounds (Guo et al., 2006; Guo et al., 2004a). Detailed discussions on the use of these data are given in later sections of this review.

4. Meteorological influence on ozone episodes

Meteorological conditions conducive to photochemical episodes in China have been extensively studied. Tropical cyclones and continental anticyclones are the main meteorological systems related to ozone episodes. Anticyclones (i.e., high pressure systems) create favorable conditions at the center, e.g., sunny weather and low wind velocity, for pollution accumulation and O_3 production (Ding et al., 2013; Gao et al., 2005). Ozone episodes in the PRD region (including Hong Kong) are often influenced by tropical cyclones in the western Pacific. Due to large-scale subsidence at the outskirts of the low-pressure systems, the PRD region often has fine weather with intense sunlight, high temperatures, and light winds (Ding et al., 2004; So and Wang, 2003).

A number of studies have examined the local meteorological parameters during ozone episodes, including solar radiation, temperature, relative humidity, wind speed and direction, and cloud cover. Similar to findings in other parts of the world, elevated ozone concentrations generally occurs on days with strong sunlight and low winds, which favor the photochemical production of ozone and the accumulation of ozone and its precursors. Wind direction is also important because it affects pollution transport, giving rise to high ozone in downwind locations. For instance, in Beijing, northwesterly airflows bring relatively clean air masses from the region of the Inner Mongolia region, but southerly winds can carry ozone and ozone precursors from Hebei and Shandong Provinces to Beijing (Duan et al., 2008; Han, 2011; Ma et al., 2011; Wang et al., 2010). In the coastal cities like Hong Kong, ozone episodes are often associated with weak northwesterly winds which transport pollution from the inner PRD cities to the coastal areas (e.g., Wang et al., 2001b; Wang et al., 2001c).

Mountain-valley winds have been shown to be an important dynamic feature in mountainous areas. Upslope winds bring pollutants including ozone from low-lying areas to the peak of Mt.

Tai (1534 m a.s.l) in Shandong (Gao et al., 2005) and to the top of Mt. Tai Mo Shan (957 m a.s.l) in Hong Kong (Guo et al., 2013), contributing to the elevated daytime concentrations of ozone and other pollutants observed at these sites. Beijing is surrounded by mountains from the northeast to the west. In summer, upslope winds transport O_3 and other pollutants to the northern mountainous areas in the afternoon and valley winds return them to the flat southern areas at night (Gao and Zhang, 2012; Wang et al., 2006a; Wang et al., 2009c).

Sea-land breezes are another important meteorological phenomena that distribute ozone and its precursors in coastal cities such as those in the YRD and PRD. Ding et al. (2004) simulated the sea-land breezes during a multiday episode in the PRD region. On episode days, from midnight to noon of the following day, the land breezes and offshore synoptic winds brought the ozone precursors from inland and coastal cities to areas over the ocean, where air pollutants accumulated due to a low mixing height and clam wind. In the afternoon, the ozone-laden air masses were recycled to the coast by onshore sea breezes, with most sites receiving the highest levels of ozone between 13:00 and 14:00 local time. Shan et al. (2010) observed a similar pattern in a coastal site in Jinshan District, Shanghai. The ozone levels were much higher during the sea breeze than the land breeze. Tie et al. (2009) found that the sea breeze is noticeable in the city of Shanghai under calm conditions. The sea breeze changes the southerly wind to an easterly direction, resulting in a cycling wind pattern in which the weak surface wind and the sea breeze trap O_3 in the city, giving rise to high ozone concentrations in the afternoon.

5. Observation-based analysis of ozone-precursors relationship

Several observation-based approaches have been developed to diagnose the O_3-precursor relationships from the field measurement data. Overall, these methods can be divided into two categories—indicator species correlation and observation-based kinetic calculations (e.g., the chemical box model and steady state calculation). In this section, we describe several widely used methods and their applications in studies of ozone pollution in China.

5.1. Observed indicators of O_3 formation regimes

5.1.1. Ozone production efficiency

Ozone production efficiency (OPE = $\Delta O_3/\Delta NO_Z$) is one of the most widely used indicators to infer the O_3 formation regimes, partly due to the simple measurements of O_3 and NO_Z (NO_Z = NOy - NO_X). It is defined as the number of O_3 molecules produced per oxidation of a NO_2 molecule to a NO_Z species (Trainer et al., 1993) and can be realized as the number of "NO_X (or RO_X) cycles" that occur before termination. Lower OPE values (e.g., ≤4) indicate inefficient radical recycling (see Figure 1), in which the supply of VOCs is the limiting factor, and thus points to the VOC-controlled O_3 formation regime. In contrast, higher OPE values (e.g., ≥7) suggest that radical cycling is efficient and that O_3 formation is mainly controlled by the availability of NO_X (i.e., NO_X-limited). OPE values in the medium range (e.g., 4 < OPE < 7) indicate that O_3 production is controlled by both NO_X and VOCs in a so-called mixed (or transition) regime.

Technically, the OPE can be derived from the regression slope of the scatter plots of O_3 versus NO_Z (e.g., Xue et al., 2011). In addition to the formal definition of $\Delta O_3/\Delta NO_Z$, several derivatives are used to determine the OPE according to the specific conditions. In urban atmospheres with high NO_X levels, for instance, total oxidants ($O_X = O_3 + NO_2$) are usually adopted instead of O_3 to take into account the O_3 titration by NO, and OPE can then be estimated as the $\Delta O_X/\Delta NO_Z$ ratio (e.g., Wang et al., 2010). In rural areas where the air masses are relatively aged, i.e., with lower NO_X/NOy ratios, NOy (or NO_X* that includes NO_X and some NO_Z species) has been used to deduce the OPE values (as $\Delta O_3/\Delta$NOy or $\Delta O_3/\Delta NO_X$*), in the absence of NO_Z measurements (e.g., Wang et al., 2006a). All of these indicator species pairs can be selected to estimate the OPE according to the specific atmospheric and experimental conditions, although the derived exact values may differ slightly.

Table 2 summarizes some OPE values reported over China in the past decade. A glance at this table reveals several noticeable features. First, most OPE investigations has been conducted around Beijing and to a lesser extent in the PRD region in south China, with sparse efforts in other areas. Second, the observationally derived OPE values vary widely (from 1.1 to 20.2) from place to place, and even case by case in the same locale, suggesting spatial and temporal heterogeneity in O_3 formation regimes. Third, a nonlinear relationship between O_3 and O_3

precursors is clearly illustrated in the O_3 (or O_X) versus NO_Z scatter plot. For example, Wang et al. (2010) examined OPE at both suburban and rural sites in Beijing and found larger OPE values of 7.7 and 6.5, corresponding to a NO_X-limited or transition regime during low NO_X conditions (i.e., NO_Z < 10 ppbv) compared to 2.7 and 4.0 (VOC-limited) at high NO_X conditions (NO_Z > 10 ppbv). Analysis of OPE is a relatively easy way to identify the ozone formation regime from field observations.

An appropriate data set forms the basis for high-quality OPE analysis. The data usually chosen for OPE calculation include either afternoon data while maximum photochemistry is occurring (e.g., Wang et al., 2010) or a specific pollution plume with strong ozone production (e.g., Wang et al., 2006a). The former provides an overall average estimation of the OPE for a longer period, although the mixing of various air plumes may raise some uncertainty for analysis. The latter case ideally derives the OPE for a polluted plume and is hence capable of providing the most accurate estimation, but it is solely relevant to a specific pollution event. In addition, the location of study sites plays an important role in determining the regional-scale O_3 formation regime by the OPE method. As mentioned above, the observed OPE values show large spatial variations within a given area. Taking Beijing as an example, the OPE-derived O_3 formation regimes at different sites vary significantly from highly VOC-limited (OPE = 1.1) to highly NO_X-limited (OPE = 20.2; Table 2). Obviously, it is impossible to formulate a universal mitigation strategy for regional O_3 pollution based on results from a single locale. To obtain a holistic understanding of the O_3 formation regime at a regional scale, representative regional monitoring networks should be developed to determine OPE at high spatial (and temporal) resolutions. Built on this, a strict statistical analysis can be enforced to achieve more robust results.

5.1.2. H_2O_2/NO_Z ratio

Another indicator of the chemistry regime of O_3 formation is the H_2O_2/NO_Z (or H_2O_2/HNO_3) ratio. For VOCs-limited regimes (i.e., high NO_X/VOCs conditions), $RO_X + NO_X$ reactions forming NO_Z species dominate the termination process and one would expect lower H_2O_2/NO_Z ratios. In comparison, higher H_2O_2/NO_Z ratios indicate the dominance of radical cross-

reactions (e.g., HO_2 + HO_2) in the radical termination processes, corresponding to the high VOCs/NO_X conditions and a NO_X-limited regime. Transition values for the H_2O_2/NO_Z ratio also occur, implying a transition in O_3 production from a VOC-limited to a NO_X-limited regime (Hammer et al., 2002; Millard and Toupance, 2002).

Compared to the O_3/NO_Z indicator, the H_2O_2/NO_Z ratio has rarely been used in previous studies in China, partly owing to a lack of concurrent measurements of H_2O_2 and NOz. Figure 3 presents both O_3-NO_Z and H_2O_2-NO_Z scatter plots during two groups of O_3 pollution episodes observed at a polluted suburban site in Hong Kong (Tung Chung) in fall 2011. We can clearly see the consistency between both indicators in determining the O_3 formation chemistry. Given the wider measurements of O_3 compared to H_2O_2, OPE should remain the preferred indicator of the relationship between O_3 and precursors, with the H_2O_2/NO_Z ratio as an important supplement.

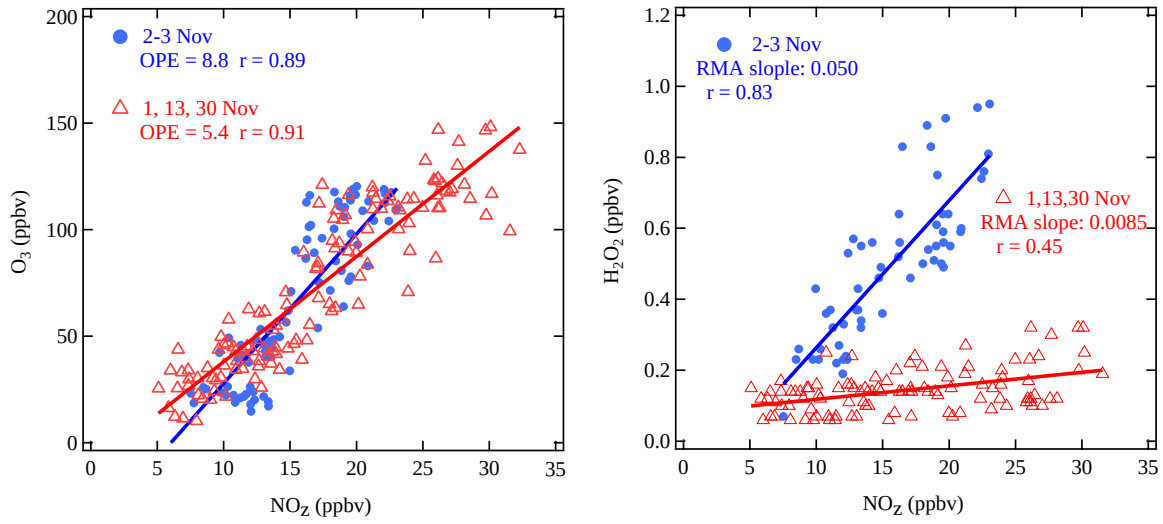

Figure 3. Scatter plots of (a) O_3 vs. NO_Z and (b) H_2O_2 vs. NO_Z during two sets of photochemical episodes at Tung Chung, Hong Kong, in fall 2011 (unpublished data). The slopes were calculated based on the Reduced Major Axis method. (RMA) (e.g., Wang et al. 2010).

5.2. Measurement-constrained model analysis of O_3 sensitivity to various precursors

The observation-based model (OBM) combines in-situ field observations and chemical box modeling. It is built on widely-used chemistry mechanisms (e.g., MCM, Carbon Bond, RACM

or SAPRC), and applied to the observed atmospheric conditions to simulate various atmospheric chemical processes, including the in-situ O_3 (usually considered as O_X) production rate. As mentioned in Section 2, O_X production is primarily facilitated by the oxidation of NO to NO_2 by HO_2 and RO_2 radicals (*R4* and *R5*), and the O_X production rate ($P(O_X)$) can be computed with Equation *E1*. The chemical loss of O_X results mainly from the photolysis of O_3 (*R13* and *R14*); reactions of O_3 with OH (*R15*), HO_2 (*R16*) and unsaturated VOCs (*R17*); reactions of NO_2 with OH (*R8*) and RO_2 (*R9*), reactions of NO_3 with VOCs (*R18*) and the heterogeneous loss of N_2O_5 on particles (*R19*) (Xue et al., 2014). Thus the loss rate of O_X can be described as Equation *E2*. The difference between *E1* and *E2* then gives the net O_X production rate.

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \quad (R13)$$

$$O(^1D) + H_2O \rightarrow OH + OH \quad (R14)$$

$$O_3 + OH \rightarrow HO_2 + O_2 \quad (R15)$$

$$O_3 + HO_2 \rightarrow OH + 2O_2 \quad (R16)$$

$$O_3 + \text{VOCs} \rightarrow \text{carbonyls} + \text{Criegee Biradicals} \quad (R17)$$

$$NO_3 + \text{VOCs} \rightarrow \text{products} \quad (R18)$$

$$N_2O_5 + \text{paricle} \rightarrow \text{products} \quad (R19)$$

$$P(O_X) = k4\,[HO_2][NO] + \sum(k5[RO_2][NO]) \quad (E1)$$

$$L(O_X) = k14[O(^1D)][H_2O] + k15[O_3][OH] + k16\,[O_3][HO_2] + \sum(k17[O_3][\text{VOC}]) + k8[OH][NO_2] + k9[RO_2][NO_2] + 2\sum(k17[NO_3][\text{VOC}]) + 3k19[N_2O_5] \quad (E2)$$

The sensitivity of ozone production to various O_3 precursors, including NO_X, VOCs, and even individual VOC species, can be quantified by conducting sensitivity modeling analyses with an assumed reduction in the concentration of target precursors. A key parameter that infers the O_3 formation regime is the relative incremental reactivity (RIR), which is defined as the ratio of the decrease in O_X production rate to a given reduction in the precursor concentration (Cardelino and Chameides, 1995). Higher positive RIR values indicate the compounds to which O_3 production is more sensitive, and the species with negative RIR play a negative role

418 (i.e., O_3 titration) in ozone formation (Xue et al., 2014a). Therefore, RIR has important
419 implications for developing a science-based control strategy for O_3 pollution.

420 In the past decade, OBM has been widely applied to assess the O_3-precursors relationship in
421 China. The available results from previous studies are summarized in Table 3. Most work has
422 been conducted in the PRD, the YRD, and the NCP, with only few studies in other areas. In
423 terms of the O_3 formation regime, most of the study areas (e.g., the PRD, YRD and the urban
424 areas in the NCP) are found in VOCs-limited zones, with anthropogenic VOCs (i.e., reactive
425 aromatics and alkenes) playing a dominant role. Nonetheless, some variations were found in
426 the chemistry regime of O_3 formation in different regions in China. Xue et al. (2014b) examined
427 the O_3-precursors relationship and VOC reactivity in four major Chinese cities (i.e., Beijing,
428 Shanghai, Guangzhou and Lanzhou) and found that O_3 formation was governed mainly by NO_X
429 at a rural site in Beijing, by aromatics in Guangzhou, by aromatics and alkenes in Shanghai,
430 and by NO_X and alkenes in Lanzhou. More observation-based modeling studies are still needed,
431 especially in the less-tapped regions and rural areas, to achieve a thorough understanding of
432 ozone formation chemistry across China. It should be noted that the OBM analysis requires
433 measuring NO at sub-ppb levels and > 50 number of VOCs; care must be taken to ensure
434 accurate determination of these critical chemicals, especially VOCs which are difficult to
435 measure.

436

Table 2. Summary of the OPE evaluation studies over China.

Region	Site [a]	Period	Site type	Indicator species	OPE Values	Reference
Beijing	Changping	Jun-Jul 2005 (episodes)	Rural downwind	O_3/NOy	3-6	Wang et al. (2006a)
		Jul-Aug. 2008		Ox/NOz	6.5±0.54 (NOz<10) 4.0±0.80 (NOz>10)	Wang et al. (2010)
	PKU[a]	Aug-Sep 2006 (episodes)	Urban	Ox/NOz	3.9-9.7	Chou et al. (2009)
	CMA[a]	Nov 2007- Mar 2008	Urban	Ox/NOz	1.1±1.6	Lin et al. (2011)
	CRAES[a]	Jul-Aug 2008	Suburban	Ox/NOz	7.7±0.78 (NOz<10) 2.7±0.49 (NOz>10)	Wang et al. (2010)
	Shangdianzi	Jul-Aug 2008 (episodes) Jul-Aug 2008 (background)	Rural background	Ox/NOz	4.0±0.32 5.3±0.55	Ge et al. (2012)
	IAP, CAS[a]	Aug 2 2008 (episode) Aug 24 2008 (episode)	Urban	Ox/NOz	6.9 20.2	Sun et al. (2011)
Pearl River Delta	Guangzhou	Summer 2006	Urban	Ox/NOz	2.1	Lu et al. (2010b)
	Back garden	Summer 2006	Suburban		7.8	
	Dinghushan	Oct 29-30 2008 (episodes)	Rural	Ox/NOz	>10	Sun et al. (2010)
Yangtze River Delta	Lin'an	Summer 2001	Rural	O_3/NOx*	~8	Wang et al. (2001a)
North China Plain	Mt. Tai	Jun 2006	Rural	O_3/NOz	5.8±0.5	Kanaya et al. (2013)
		Jun 2006-2015 Jul-Aug 2006-2015		O_3/NO_2*	3.9-14.9 3.6-15.0	Sun et al. (2016)
Qinghai-Tibetan Plateau	Waliguan	Jul-Aug 2006 (episodes)	Remote	O_3/NOz	7.7-11.3	Xue et al. (2011)

[a] PKU: Peking University; IAP, CAS: Institute of Atmospheric Physics, Chinese Academy of Sciences; CMA: China Meteorological Administration; CRAES: Chinese Research Academy of Environmental Science.

Table 3. Summary of the OBM studies over China.

Region	Site	Sampling period	Site type	Chemical Mechanism	O_3 Formation Regime	Dominant VOCs [a]	References
Pearl River Delta (including Hong Kong)	PRD (8 sites)	summer, autumn 2000	Urban/suburban	–	VOCs-limited (Guangzhou)	AHC	Shao et al. (2009b)
	Hong Kong (5 sites)	Oct 1-Dec 31, 2002	Urban/suburban	CB–IV	VOCs-limited	R–AROM	Zhang et al. (2007)
	Tung Chung	autumn 2002-2013	Suburban	MCM3.2/CB–IV	VOCs-limited	R–AROM	Cheng et al. (2010); Xue et al. (2014a); Xue et al. (2014b)
	Guangzhou	2004 Nov/2006 Jul	Urban	CB–IV	VOC-limited	AHC	Lu et al. (2010b); Zhang et al. (2008c)
	Xinken	2004 Nov	Rural	GIT–OBM	VOC-limited	AHC	Zhang et al. (2008c)
	Back garden	2006 Jul	Suburban	CB–IV	transition	–	Lu et al. (2010b)
	Wan Qing Sha	Oct 23-Dec 1 2007	Suburban	CB–IV	VOCs-limited	R–AROM	Cheng et al. (2010); Ling et al. (2011)
	Tsuen Wan	autumn 2010	Urban	MCM3.2	VOCs-limited	R–AROM	Guo et al. (2013); Ling et al. (2014)
	Tai Mo Shan	Sep-Nov 2010	Semirural site	MCM3.2	transition	–	Guo et al. (2013)
Yangtze River Delta	Taicang	Apr 4-Jun 1 2005	Urban	MCM3.2	VOCs-limited	R–AROM	Xue et al. (2014b)
	Shanghai	Nov 15-Nov 26 2005	Urban	NACR–MM	VOCs-limited	R-AROM	Geng et al. (2007)
	Shanghai (5 sites)	Jan 2006-May 2007	Urban/suburban/rural	NACR–MM	VOCs-limited (urban)	R–AROM	Geng et al. (2008)
	Jiangsu	May 15-Jun 24 2010	Rural	RACM	VOC-limited (morning) to NO_X-limited (afternoon)	–	Pan et al. (2015)
	Nangjing (4 sites)	Jun-Aug 31, 2013	Urban/suburban	CB–IV	VOCs-limited	Alkenes	An et al. (2015)
North China Plain	Mount Tai	Jun 2006	Mountain	RACM	NO_X-limited (mainly)	–	Kanaya et al. (2009)
	Peking University	Aug-Sep 2006/Aug 2007	Urban	CBM4/SAPRC–07	transition /VOCs-limited	–	Liu et al. (2012); Lu et al. (2010a)
	Yufa	Aug-Sep 2006	suburban	CBM4	Mixed-limited	–	Lu et al. (2010a)

	Wuqing	summer 2009	Suburban	NACR–MM	NO_X-limited	–	Ran et al. (2011)
	Yanshan	Jul-Aug of 2010 and 2011	Industrial	NACR–MM	NO_X-limited	–	Wei et al. (2015)
	Tieta	Jun-Oct, 2010/Jul-Aug 2010 and 2011	Urban	NACR–MM	transition /VOCs-limited	–	Ran et al. (2012); Wei et al. (2015)
Western and Central China	Lanzhou	Jun 19-Jul 16 2006	Urban	MCM3.2	NO_X-limited	–	Xue et al. (2014b)
	Wuhan	Feb 2013-Oct 2014	Urban	MCM3.2	VOCs-limited	AHC	Lyu et al. (2016)

[a] AHC represents anthropogenic hydrocarbons; R-AROM represents reactive-aromatics; AHC represents anthropogenic hydrocarbons

6. Processes analysis of ozone and source attribution

6.1. Ozone source apportionment

Severe O_3 air pollution events over highly urbanized areas have prompted the development of emission-driven photochemical grid modeling to better understand the pollution sources/sinks, formation mechanisms, and regional source contribution, and to help develop effective control strategies. Techniques applied in these models include the factor separation technique, ozone source apportionment technology, integrated process analysis, and response surface modeling (Li et al., 2012a; Liu et al., 2010; Qu et al., 2014; Xing et al., 2011).

6.1.1. The Jing-Jin-Ji region

Regional air quality models such as CAMx, CMAQ, and WRF-Chem have been applied to analyze the source contribution of ozone. Wang et al. (2009b) investigated high ozone events in Jing-Jin-Ji using Ozone Source Apportionment Technology (OSAT) with Geographic Ozone Assessment Technology (GOAT). They found that the main contribution to high afternoon ozone levels in June 2000 was from the urban and southern Beijing areas, which accounted for 31.6% and 12.6%, respectively. Furthermore, they found a significant contribution of precursor emissions from southern Hebei Province and Tianjin, which were estimated at 16.9% and 12.6%, respectively. This result is consistent with an early study by Streets et al. (2007) using CMAQ, which found that ozone air pollution in Beijing was derived partially from regional emissions outside of Beijing (e.g., Hebei), reportedly 20% to 30% in July 2001 and 35% to 65% during the high ozone episodes. Mijling et al. (2013) indicated that Hebei province is one of the nation's top three NO_x emissions. The high local emissions in Jing-Jin-Ji have rendered the contribution of long-range transport from other continents (e.g., Europe) insignificant (Fu et al., 2012). In general, urban Jing-Jin-Ji is found to be VOC-limited, due to the abundance of NOx emissions from local industrial and traffic sources, whereas the rural area of Jing-Jin-Ji tends to be NOx limited (Chou et al., 2009; Wang et al., 2010). This finding is further supported by a model study by Liu et al. (2010) that found $P(H_2O_2)/P(HNO_3)$ ratios below 0.2 in urban Beijing and Tianjin during summer with CMAQ (Tonnesen and Dennis, 2000). Additional analysis by Wang et al. (2009b) using OSAT source category analysis suggested that the relative ozone contributions from various sources in Beijing are 31.6% (mobile), 20% (industrial), 13% (point) and 12% (biogenic).

In terms of regional emission control, Qu et al. (2014) applied multi-"brute force" CMAQ model simulations (August 2007) with a factor separation technique to identify the effects of

individual emission source types. They found that power plant emissions play an important role of in peak ozone levels in southwestern Jing-Jin-Ji. Moreover, a 30% across-the-board reduction in industrial and transportation emissions would result in a maximum reduction of 20 ppbv in the Jing-Jin-Ji area. In another study, Xing et al. (2011) examined multiple emission reduction options using the CMAQ response surface model and found that solely reducing local NOx or VOC emissions in the Beijing area (even by 90%) would be insufficient to meet the ozone Class 2 Ambient Air Quality Standard of China. It is recommended that synchronous regional reductions in VOC and NOx by 60% to 80% are needed across the entire Jing-Jin-Ji region.

6.1.2. The PRD

In the PRD region, with its different emission characteristics and meteorological conditions, the regional and source category contributions to high O_3 differ from those in Beijing. Using the CAMx model implemented with OSTA, Li et al. (2012c) reported that between 7 and 10 November 2006, the top sources of average daytime ozone in the PRD were mobile (28%), area (14%), point (5%), marine (2%), and biogenic sources (5%), respectively, and in summer (24 and 25 July 2006), they were 30% (mobile), 12% (area), 16% (point), 3% (marine), and 8% (biogenic), respectively. Li et al. (2013) further extended the ozone contribution estimates for Hong Kong and the PRD to all four seasons and established a full precursor contribution matrix for ozone concentration in all PRD cities. They suggested that during ozone episodes, the top contributors to the ozone in PRD cities were mobile, point, and area sources, and that local and PRD regional sources accounted for 68% to 80% of ozone concentrations in summer, 35% to 55% in fall, and 19% to 32% in spring and winter (Li et al., 2013). These values could be even higher with the recent findings of an ozone production pathway from HONO chemistry, which further enhanced the local ozone production by 6% to 12% in Hong Kong and major PRD cities (Zhang et al., 2016).

In terms of regional emission control, it is suggested that the PRD government should focus on the local emissions reduction, particularly in the mobile source category, to maximize the effectiveness of control policies for long-term ozone reduction. Ou et al. (2016) suggested that 43% of NOx and 40% of anthropogenic VOCs reduction in the mobile sources can be achieved by applying Stage V of the Vehicle Emission Standards in the PRD areas. They also suggested a VOC-focused control with a reduction ratio of 1:2 (anthropogenic VOC to NO_x) to effectively reduce the peak O_3 levels in urban and industrial areas.

6.1.3. The YRD

Compared with the two regions mentioned above, source apportionment studies for the YRD are relatively limited. Using CAMx with OSAT, Li et al. (2015a) and Wang et al. (2014) reported in August 2010 and July 2013 that the top sources of the 8-hr maximum ozone were mobile (19% to 23%), point (5.7% to 9.8%), industrial (38.3% to 53.0%), and biogenic (6.6% to 17%). Li et al. (2015a) found that the regional contributions of non-YRD emissions to the averaged ozone in the Shanghai, Jiangsu and Zhejiang areas were 43%, 49%, and 60%, respectively, indicating a large influence of emissions from outside the YRD. On high ozone days, the contributions from local and northern Zhejiang to the average ozone in Shanghai were suggested to be 29% and 20%, respectively, which are consistent with the results (13% to 21%) of CMAQ process analysis by Li et al. (2012a). Using the WRF-Chem model coupled with an ozone tagging method, Gao et al. (2016) studied the source regions for ozone pollution in the YRD and showed that the region was affected by surrounding upwind provinces such as Anhui, Shandong, and Henan-Hebei, with average contributions of 16.2%, 13.6%, and 9.0%, respectively, during the study period.

6.2. Impact of HONO and $ClNO_2$ on ozone formation in China

Photolysis of emitted and secondarily formed HONO during the daytime could significantly increase the levels of OH radicals that accelerate ozone formation (Aumont et al., 2003). However, early modeling studies of ozone mostly underestimated daytime HONO sources and thus their contributions to ozone production (Zhang et al., 2016). Elevated levels of HONO (0.15 to 3.24 ppbv) have been observed across the Jing-Jin-Ji (mostly Beijing), YRD, and PRD regions (Hendrick et al., 2014; Hou et al., 2016; Li et al., 2012b; Su et al., 2008; Xu et al., 2015) and were suggested to considerably enhance the ROx concentrations and accelerate the ROx cycles in these regions (Liu et al., 2012; Tang et al., 2015). Regional model simulations have suggested that elevated HONO in China could lead to an increase of 3 to 10 ppbv in the 8-h maximum surface O_3 concentrations in most areas of the Jing-Jin-Ji region (An et al., 2013; Li et al., 2011), an increase of 60% to 250% in OH, HO_2, and RO_2 levels in China's coastal regions, including the YRD (Tang et al., 2015), and an increase of 6% to 12% enhancement in the daily ground-level ozone concentration over the urban areas of the PRD region (Zhang et al., 2016).

$ClNO_2$ is formed at night via heterogeneous reactions of N_2O_5 on aerosols that contain chloride. During the daytime, $ClNO_2$ is photolyzed to release chlorine atoms (Cl) and NO_2, both of which affect ozone production (Osthoff et al., 2008; Thornton et al., 2010). Recent field

519 measurements in the Jing-Jin-Ji and PRD (Hong Kong) regions found high levels of $ClNO_2$
520 (0.35-4.7 ppbv) that contribute 10% to 30% of primary ROx production in the morning hours
521 and enhance the integrated daytime net ozone production by up to 13% at a rural site in the
522 NCP and up to 41% at a mountain site in Hong Kong (Tham et al., 2016; Tham et al., 2014;
523 Wang et al., 2016; Xue et al., 2015). Regional model simulations by Sarwar et al. (2014)
524 suggest that $ClNO_2$ in China could reach up to 0.8 ppbv and lead to an increase of 7 ppbv in
525 the 8-hr maximum ozone concentration. Li et al. (2016) suggested that across the PRD region
526 the $ClNO_2$, which was mostly concentrated within the residual layer (~300 m above the surface),
527 increased O_3 by up to 16% within the PBL. These findings suggest a potentially important
528 contribution of 'new' radical or radical sources to O_3 formation in polluted atmospheres such
529 as in the eastern part of China. Photochemical air quality models should consider these sources
530 and processes.

531 **7. Effect of ozone on crops and human health**

532 **7.1. Effect on crops**

533 Ozone is widely distributed in the troposphere, which means that its presence is found well
534 beyond the more polluted cores of large cities. Thus it has the potential to damage crops and
535 vegetation or affect human health in populations dispersed over continental areas. Ozone
536 affects vegetation through a range of mechanisms (Bhatia et al., 2012), and its effects on crops
537 in China have recently been reviewed by Feng et al. (2015). The interaction between vegetation
538 and air pollution begins with the transfer to vegetation via turbulent diffusion. High ozone
539 concentrations are found in spring, and typically in the afternoon when the wind speed is often
540 low; i.e., conditions that limit the transfer of ozone. The phytotoxicity of ozone is largely due
541 to its oxidative capacity and the generation of oxidizing entities such as OH, O_2- and H_2O_2.
542 These attack the composition, structure, and function of the plasma membrane, and peroxide
543 can be transported through the membrane and generate species that affect the signal chains via
544 messenger molecules. Common symptoms of foliar injury are: changes in pigmentation,
545 chlorosis and premature senescence; the effects on water use make crops appear to wilt (Feng
546 et al., 2014). These processes affect plant growth, and McKee and Long (2001) suggested that
547 the changes to allocation and development ozone causes are more important than its effects on
548 photosynthesis and biomass accumulation.

549 Chameides et al. (1999) suggested that ozone concentrations across China were sufficiently
550 enough to affect winter wheat production. More recently, Wang et al. (2012b) and Feng et al.

(2015) argued that rising ozone concentrations pose problems for China's food security. These concerns are heightened by the possibility that changes in climate and precursor emissions seem likely to continue to increase and have led to a number of experimental studies, especially on the effect of ozone on wheat and rice (Feng et al., 2003; Wang et al., 2012b). Exposure to 62 ppb of ozone gives a yield reduction in China's field-grown rice of 14% to 20%, although wheat is more sensitive than rice (Wang et al., 2012b). Although a range of methods are available to estimate the yield loss, they can give very different values. In the case of spring wheat across China (which is more affected than winter wheat), the predicted losses in yield for 2020 vary between 2% and 29%. In the case of rice, the losses may be between 3.7% and 10%. Losses of summer corn may be as much as 64% and soybeans 45% by 2020, although these estimates are also highly variable (Feng et al., 2015).

Clearly more work is needed given the large uncertainties in the likely impact of ozone on food crops, but such studies may contribute to the development of air quality standards for crops. Although it is desirable to reduce ambient ozone concentrations in agricultural areas, a more immediate approach may be to choose more resistant crop varieties (e.g., Feng et al., 2015).

7.2. Effects on human health

Human health is also affected by exposure to ozone and epidemiological studies have demonstrated an association between ambient ozone levels and premature mortality. Ozone is a pulmonary irritant that affects respiratory mucous membranes and other lung tissues. A reduction in lung function means that exposure to elevated concentrations can lead to increased hospital admissions for pneumonia, chronic obstructive pulmonary disease, asthma, allergic rhinitis, and other respiratory diseases. There is likely a causal relationship between cardiovascular outcomes and short-term exposure (<30 days), with indications of biological disease mechanisms below the air quality standard of 75 ppbv (Goodman et al., 2015). It is now recognized that short-term exposures to ozone is also linked to childhood asthma, but the causality has yet to be established (Sousa et al., 2013). It has often been suspected that ozone has a synergistic relationship with other pollutants, and in line with this there are recent observations of this in relationship to asthma (Alexis and Carlsten, 2014).

The ozone standards set by the WHO (2005) are based on the small but convincing, associations between daily mortality rates and ozone concentrations, which suggested the need for a downward revision of the earlier value of 120 μg m^{-3} to 100 μg m^{-3} for the 8-hr maximum concentration. New regulations in China reflect these changes, suggesting that ozone air quality

583 standards (GB 3095-2012) for class 1 (remote) areas mandate daily 8-hr and 1-hr maxima of
584 100 and 160 μg m^{-3}, respectively, whereas for class 2 (urban/industrial and surrounding rural)
585 areas, these values are 160 and 200 μg m^{-3}, respectively. There is little evidence of a threshold,
586 and recent work supports sensitivity even to low concentrations. The absence of a threshold
587 argues for more stringent regulations (Goodman et al., 2015; WHO, 2005), and this must be
588 especially true for class 2 urban areas.

589 However, appropriate metrics (Li et al., 2015b) and the difference between indoor and outdoor
590 concentrations (ozone is readily deposited on indoor surfaces) can make exposure estimates
591 problematic and likely health impacts uncertain (e.g. Chen et al., 2012). Although numerous
592 studies have been done on the epidemiology of ozone (e.g. Bell et al., 2007; Bell et al., 2004),
593 they are less common in China (e.g. Yang et al., 2012). Some have examined the acute effects
594 of ambient ozone and ozone metrics in Guangzhou (Li et al., 2015b) and Suzhou (Yang et al.,
595 2012). In cities in the RPD region, increases of 10 μg m^{-3} in ozone concentrations over the prior
596 2 days were associated with a 0.81% increase in the mortality rate (Tao et al., 2012). In Hong
597 Kong, Wong et al. (2006) suggested that the greatest respiratory effects of air pollution are on
598 relatively chronic obstructive pulmonary diseases, with ozone having the greatest effect at
599 1.034 for a 10 μg m^{-3} increase in concentration. More recently, Liu et al. (2013) observed a
600 seasonal sensitivity to ozone exposure in Guangdong, with the effects of ozone lasting longer
601 on cold days with a 3.34% increase for 10 μg m^{-3} accumulating over a 6 day lag period.
602 Shanghai showed a shorter 2-day lag during the cold season, with a 0.45% increase (Zhang et
603 al., 2006).

604 Studies of the regional distribution of the global disease burden (e.g. Brauer et al., 2016). have
605 indicated significant risks from exposure to outdoor air pollutants in China, drawing particular
606 attention to particulate matter and ozone. Throughout China, the financial burden of air
607 pollution in terms of medical expenses and wage and leisure loss is seven times higher for
608 particulate matter than for ozone (Matus et al., 2012). However, any future increases in ozone
609 will tend to elevate the importance of ozone, because as a secondary pollutant it is generally
610 more difficult to regulate. A key obstacle to understanding the health effects of ozone is the
611 paucity of studies in China that explore the effects across the breadth and geographical diversity
612 of the country. In addition, the strong gradient between indoor and outdoor ozone
613 concentrations influences human exposure.

614 **8. Summary and recommendations**

615 This review focuses on published research findings in English-language literature on surface
616 ozone processes in China, including ozone abundance and its relationship to atmospheric
617 dynamics and chemical processes. We offer the following conclusions and recommendations:

618 • The available data have clearly shown serious ozone pollution in China's major cities,
619 especially in the three most developed regions, namely "Jing-Jin-Ji," the YRD, and the
620 PRD. Data from other populated and fast-developing regions are needed to get the full
621 picture of ozone pollution in China. In this regard, it is strongly suggested that the ozone
622 data from nation-wide air monitoring networks be made available for such assessments.
623 • Measurements and modeling studies have revealed that in most urban and industrial
624 regions in the eastern half of China, ozone production is limited by VOCs. However,
625 more studies in forested areas downwind of urban/industrial centers are needed to
626 determine the transition to NOx dominated regimes and to develop ozone precursor
627 control strategies. In view of successful NOx control in coal fired power plants since
628 2010 but delayed actions in reducing VOCs emissions, it is likely that some regions
629 will change to NOx-limited regimes. It is recommended that implementation of
630 measures to control VOCs along with NOx should be speeded up.
631 • Reactive aromatics are found to be the predominant contributor to ozone formation in
632 many urban areas in China. Control measures targeting solvent use along with vehicular
633 emissions are advised.
634 • Cross-boundary transport has been shown to be an important cause of high ozone events
635 in Beijing, Shanghai, and Hong Kong, highlighting the need to control precursors not
636 only in the jurisdiction concerned but also over larger regions.
637 • Despite a good understanding of the overall ozone formation mechanism, recent
638 research has revealed some new sources of radicals (e.g., additional sources of HONO
639 and chlorine from the photolysis of $ClNO_2$) that require better understanding to improve
640 the predictive capability of current photochemical models. Some processes involve
641 heterogeneous reactions (such as the uptake of N_2O_5 on aerosol surfaces), which could
642 be particularly important in China in view of the high aerosol loading in China's
643 atmosphere.
644 • In recent years, numerous VOCs measurement programs have been undertaken by both
645 the research community and government agencies. Inter-comparison of the
646 measurement techniques of different groups is needed to ensure data comparability in

view of the technical difficulties in sampling, identifying, and quantifying hundreds of VOCs.

- Most previous field studies were conducted at surface sites, and more vertical measurements of chemical and meteorological parameters are needed to understand the processes in the whole boundary layer and the exchanges with the free troposphere.
- Research on the effects of ozone on human health and crops is limited relative to that on atmospheric processes. It is recommended that more research be conducted on the effects of ozone and consideration be given to further lowering the guideline values for Class 2 areas.

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