

# Increasing External Effects Negate Local Efforts to Control Ozone Air Pollution: A Case Study of Hong Kong and Implications for Other Chinese Cities

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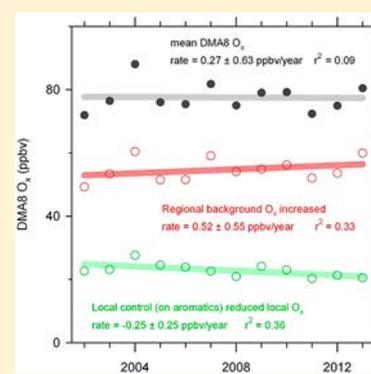
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## Supporting Information

**ABSTRACT:** It is challenging to reduce ground-level ozone ( $O_3$ ) pollution at a given locale, due in part to the contributions of both local and distant sources. We present direct evidence that the increasing regional effects have negated local control efforts for  $O_3$  pollution in Hong Kong over the past decade, by analyzing the daily maximum 8 h average  $O_3$  and  $O_x$  ( $=O_3+NO_2$ ) concentrations observed during the high  $O_3$  season (September–November) at Air Quality Monitoring Stations. The locally produced  $O_x$  showed a statistically significant decreasing trend over 2002–2013 in Hong Kong. Analysis by an observation-based model confirms this decline in in situ  $O_x$  production, which is attributable to a reduction in aromatic hydrocarbons. However, the regional background  $O_x$  transported into Hong Kong has increased more significantly during the same period, reflecting contributions from southern/eastern China. The combined result is a rise in  $O_3$  and a nondecrease in  $O_x$ . This study highlights the urgent need for close cross-boundary cooperation to mitigate the  $O_3$  problem in Hong Kong. China's air pollution control policy applies primarily to its large cities, with little attention to developing areas elsewhere. The experience of Hong Kong suggests that this control policy does not effectively address secondary pollution, and that a coordinated multiregional program is required.



## 1. INTRODUCTION

Ozone ( $O_3$ ) is a principal component of photochemical smog, a form of air pollution that is rife within urban-industrialized regions. High concentrations of  $O_3$  can adversely affect the health of humans, crops, and vegetation, and may alter the climate.<sup>1–3</sup> The ambient  $O_3$  at a given location is produced by a variety of precursors, namely nitrogen oxides ( $NO_x=NO+NO_2$ ), carbon monoxide (CO) and volatile organic compounds (VOCs), which may be emitted either locally or from distances even hundreds of kilometres away.<sup>4,5</sup> Therefore, a major challenge of  $O_3$  pollution regulation is to deal with processes and contributions on different scales.

Ozone pollution has posed an ongoing challenge to researchers and policy makers since it was first discovered in Los Angeles in the 1950s.<sup>6</sup> Recent compilations of long-term monitoring data indicate success in the implementation of pollution-control measures in some metropolitan areas in North America, for example, Los Angeles (California's South Coast Air Basin),<sup>7</sup> New York (the Northeastern U.S. urban corridor)<sup>8</sup> and Houston.<sup>8,9</sup>  $NO_x$  and VOCs have been significantly reduced in both these cities and the surrounding areas.<sup>8,10,11</sup> In contrast, the air quality in China has deteriorated over the last three decades, due to the nation's rapid urban-industrialization. This deterioration includes emergence of widespread ground-level  $O_3$  pollution.<sup>12–14</sup> Hong Kong is

situated on the South China coast, adjacent to the rapidly developing Pearl River Delta (PRD) region (Supporting Information (SI) Figure S1). As Hong Kong has far outstripped mainland China in terms of economic development and environmental regulation (note that Hong Kong and mainland China constitute one country with two administrative systems) and is downwind of mainland China in autumn/winter monsoon seasons, it provides a unique opportunity to evaluate the effectiveness of local efforts to control  $O_3$  pollution against the backdrop of an ongoing increase in  $O_3$ -precursor emissions in the surrounding regions.

Like virtually all other metropolitan areas, Hong Kong has for many years suffered from serious air-pollution problems, including early reports of photochemical smog in the mid-1990s.<sup>15</sup> To improve the territory's air quality, the Hong Kong government has since implemented long-term and step-by-step measures to reduce the emissions of  $NO_x$ , CO and VOCs from vehicles, power plants, industrial, commercial and residential activities (see [http://www.epd.gov.hk/epd/english/environmentinhk/air/prob\\_solutions/strategies\\_apc.html](http://www.epd.gov.hk/epd/english/environmentinhk/air/prob_solutions/strategies_apc.html) for

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details of the air pollution control program). As a result, the anthropogenic emissions of  $O_3$  precursors have significantly decreased since the 1990s (see SI Figure S2 for the estimated emission inventory data). However, this reduction in local emissions does not appear to have alleviated the  $O_3$  pollution problem in Hong Kong. Previous studies have shown that the  $O_3$  concentration in both the background and urban atmospheres increased at an average rate of 0.58 ppbv/year during 1994–2007,<sup>16</sup> and suggested considerable contributions from regional transport in the PRD and super-regional transport from eastern China.<sup>16–18</sup> Thus, it is crucial to quantify the contributions of local and external sources to  $O_3$  trends in Hong Kong and to assess the effectiveness of local pollution-reduction measures.

In this study, we used data obtained from the Air Quality Monitoring Network by the Hong Kong Environmental Protection Department (HKEPD) to analyze the  $O_3$  trends over 2002–2013 as well as the local and regional contributions. An observation-based box model (OBM) was then used with the most comprehensive data at a polluted site (Tung Chung) to verify trends in local  $O_3$  production and to evaluate the effects of reducing individual  $O_3$  precursors. The results of the analysis provide solid evidence that the government's control efforts have reduced local ozone production in Hong Kong. However, due to increasing contributions from southern/eastern China, the overall  $O_3$  concentration has risen. We discuss the implications of Hong Kong's experience for other Chinese cities.

## 2. MATERIALS AND METHODS

**2.1. Data Source.** We analyzed the air-quality data (i.e.,  $O_3$ , CO and  $NO_x$ ) collected at 11 Air Quality Monitoring Stations (AQMS; see SI Figure S1) in Hong Kong during the autumn season (September–November) of 2002–2013. The autumn data was chosen because it is the period when Hong Kong suffers from severe photochemical  $O_3$  pollution and is generally located downwind of mainland China under Asian monsoons.<sup>16,19</sup> The instrumentation, quality assurance and control protocols implemented by the Hong Kong AQMS are similar to those used in the US's air-quality monitoring program. Continuous measurements were available at most AQMS over 2002–2013, with occasional data gaps at 1 or 2 sites in several years (see Table S1 for an overview of the data availability at each AQMS). The daily maximum 8-h average (DMA8)  $O_3$  concentrations, upon which the ambient air quality standards in Hong Kong (75 ppbv) are based, were subject to the analysis. To take into consideration the titration of  $O_3$  by fresh NO emissions,  $O_x$  was used instead of  $O_3$  to determine the regional and local contributions.

The data obtained at Tung Chung (see also SI Figure S1) were subject to a more-detailed OBM analysis, for the following reasons: (1) this site is located in the area of Hong Kong that experiences the most serious  $O_3$  pollution,<sup>20</sup> and (2) the measurements taken at this site comprised the most comprehensive source of data, including long-term observations of hydrocarbons and short-term intensive measurements of oxygenated VOCs (OVOCs) and nitrous acid (HONO). We have two sets of hydrocarbon measurements. One was made by using canister sampling coupled with laboratory detection by gas chromatography (GC) with flame-ionization detection (FID), electron-capture detection and mass-spectrometry detection, which is an internationally recognized system for VOC measurements and has been utilized in dozens of

previous studies (the chemical analysis of samples was carried out at the University of California at Irvine).<sup>21,22</sup> The other set comprised the real-time measurements obtained by a combination of GC separation, photoionization detection and FID detection (Syntech Spectras, model GC955 Series 600/800 POCP; it consists of two systems for detection of  $C_2$ – $C_5$  and  $C_6$ – $C_{10}$  hydrocarbons respectively). The canister data covered a wide range of nonmethane hydrocarbon (NMHC) species ( $C_2$ – $C_{10}$ ; >50 species) at a relatively low time resolution (normally one 24 h sample per week). In comparison, the real-time data had a high time resolution (30 min) but only covered ~30 major  $C_2$ – $C_9$  NMHC species. Canister measurements were available for 2002, 2006, 2007, and 2011, and the validated real-time data covered the period of 2008–2012. Overall, both measurements showed good agreement for alkanes and aromatics but large discrepancy for alkenes (see SI Figure S3 for an intercomparison). It is still not clear about the reason for the discrepancy in ambient alkenes measurements (note that our periodical intercomparison by sampling the same standards showed good agreement between both methods). Considering that the real-time instrument sometimes may not work well for alkenes (especially the low-carbon species) and that the canister measurements are usually believed to be more reliable, we corrected the real-time data according to the canister data in the base analysis. Besides, we conducted another set of analysis with canister data corrected by real-time data, and found that our major conclusions remained unchanged (see SI for a detailed evaluation).

**2.2. Determination of Regional Background and Locally Produced Ozone.** We utilized the method proposed by the Texas Commission on Environmental Quality (TCEQ)<sup>9,23,24</sup> to determine the “regional background  $O_x$ ” transported into the Hong Kong area. Ten AQMS (except for Sham Shui Po site; see SI Figure S1) were selected based on their locations near the urban perimeter and distances from local emission sources. These sites are believed to be capable of measuring regional background  $O_x$  when any of them is upwind of the Hong Kong urban area. The regional background  $O_x$  can be estimated as the lowest DMA8  $O_x$  concentration recorded by these sites. We also computed the area-wide peak  $O_x$  as the highest DMA8  $O_x$  value observed at all of the AQMS (including Sham Shui Po). The locally produced  $O_x$  can be determined as the difference between the peak  $O_x$  and regional background  $O_x$ . The regional background and locally produced  $O_x$  were estimated every day for the autumn seasons of 2002–2013.

Berlin et al. recently evaluated the uncertainties of the TCEQ determination of regional background ozone,<sup>9</sup> which may be overestimated with recirculation of aged local plumes with  $O_3$  produced in the preceding days, and be underestimated if there was a large spatial gradient in background  $O_x$ . Such uncertainties are inevitable for all the observation-based studies. Here it should be recognized that our derived “local  $O_x$ ” refers to the  $O_x$  produced photochemically in Hong Kong within 1 day. Another uncertainty relates to the weak sea breeze penetrating partially and affecting only a few coastal sites, which may also lead to underestimation of background  $O_x$ . This problem should be small in the present study given the dominance of continental outflow from mainland China in autumn. A somewhat subjective but stringent method was adopted here to reduce this problem. We examined the time series obtained at all AQMS, and chose the days on which the DMA8  $O_x$  at the coastal site (i.e., Tap Mun) was distinctly

lower than those at all the other sites and DMA8 O<sub>3</sub> at Hok Tsui. For such days, the lowest DMA8 O<sub>x</sub> at the noncoastal sites was regarded as the regional background O<sub>x</sub>. Only 23 days (2.1%) out of 1092 were revised, and the regional background O<sub>x</sub> was increased on average by 17.8 ppbv on these days. Despite the high data capture rates (normally >97%) at AQMS, some data gaps remain (e.g., at KT in 2002; see SI Table S1), which may also raise uncertainty in the regional background O<sub>x</sub> determination. It is difficult to accurately quantify such uncertainty, but it should be small in view of the facts that the TCEQ method uses the data at ten AQMS (using only the maximum and minimum O<sub>x</sub>) and that the KT data were generally moderate among all AQMS.

We compared the regional background O<sub>x</sub> determined by the TCEQ method with the DMA8 O<sub>3</sub> observed at Hok Tsui, a remote background station of Hong Kong (see the results in SI Figure S4).<sup>16</sup> NO<sub>2</sub> was not routinely measured at Hok Tsui, but its concentrations are generally very low at midday compared to O<sub>3</sub> according to our previous observations.<sup>25</sup> Overall, the TCEQ background O<sub>x</sub> exhibited a good agreement with the O<sub>3</sub> at Hok Tsui, with a slope of  $1.01 \pm 0.03$  (95% confidence intervals) and  $r^2$  of 0.74. This suggests that the TCEQ method provides a reasonable measure of the regional background O<sub>x</sub> entered Hong Kong.

**2.3. Observation-Based Model.** An observation-based chemical box model was used to estimate in situ O<sub>3</sub> production and its sensitivity to changes in O<sub>3</sub> precursors. In brief, the model was based on the Master Chemical Mechanism (MCM v3.2), an explicit mechanism describing the degradation of 143 primary VOCs together with the latest IUPAC inorganic nomenclature.<sup>26–28</sup> In addition to chemistry, we considered physical processes such as dry deposition and atmospheric dilution with evolution of the planetary boundary layer. The photolysis frequencies appropriate for Hong Kong were parametrized using a two-stream isotropic-scattering model under clear-sky conditions (we did not consider the impact on O<sub>3</sub> production of the variation in solar radiations, and the rationale of this study is just to evaluate the effect of O<sub>3</sub> precursor changes), and were calculated within the model as a function of the solar zenith angle.<sup>28,29</sup> A detailed description of this model, which we have used in previous studies,<sup>12,30</sup> is given in the SI.

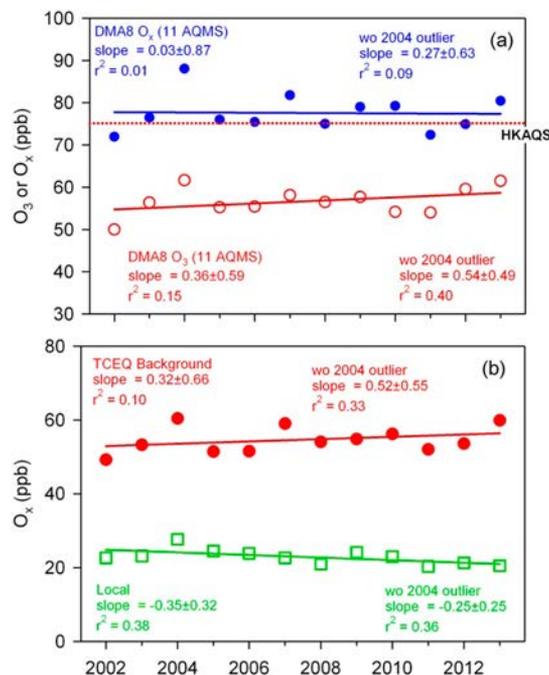
We performed model calculations for the years of 2002 and 2006–2012, during which the VOC measurements were available. The model was constrained by the autumn-average diurnal profiles of O<sub>3</sub>, CO, SO<sub>2</sub>, NO, NO<sub>2</sub>, CH<sub>4</sub>, C<sub>2</sub>–C<sub>10</sub> NMHCs, H<sub>2</sub>O, temperature and pressure measured at Tung Chung. To facilitate a more reasonable modeling analysis, we also prescribed the model with the measurements of OVOCs (17 species; see SI), HONO and aerosol surface density collected at Tung Chung during an intensive campaign in the autumn of 2011 (assuming constant levels for all years). We also conducted sensitivity analysis without constraints of the measured HONO. The results indicated that our major conclusions were unchanged (see SI for a full discussion).

The model read the measurement data every hour to calculate in situ O<sub>x</sub> production, destruction and net rates. The model was run with 00:00 local time (LT) as the initial time. For each simulation, the model preran for 9 days to ensure that it approached a steady state necessary to stabilize the concentrations of unconstrained species. The output of the 10th day was then subject to further analyses. We also

estimated the sensitivity of O<sub>3</sub> production to changes in various O<sub>3</sub> precursors by conducting sensitivity model runs.

### 3. RESULTS

**3.1. O<sub>3</sub>/O<sub>x</sub> Trends.** Figure 1a shows the trends of the autumn-average DMA8 O<sub>3</sub> and O<sub>x</sub> observed in Hong Kong



**Figure 1.** (a) Trends in autumn mean (September–November) DMA8 O<sub>3</sub> and O<sub>x</sub> (O<sub>3</sub>+NO<sub>2</sub>) recorded by 11 AQMS stations. The HKAQS O<sub>3</sub> concentration (75 ppbv) is indicated by the red dotted line. (b) Trends in regional background and local O<sub>x</sub> estimated by the TCEQ method. Linear regressions were made for all years (solid lines) and without the exceptional year 2004 (not shown), with slopes ( $\pm$ 95% confidence intervals) and R squares annotated.

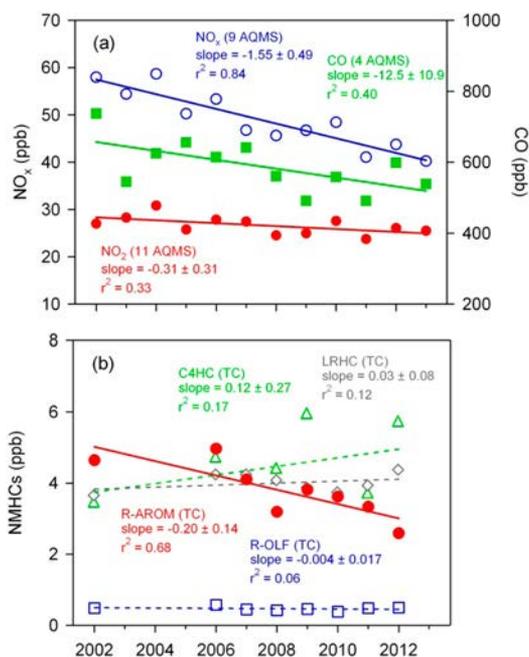
between 2002 and 2013. A slightly increasing trend was found for O<sub>3</sub> with a change rate of  $0.36 \pm 0.59$  ( $\pm$ 95% confidence intervals) ppbv/year, but this increase was of little statistical significance ( $p = 0.21$ ). The poor statistics were mainly driven by the outlying measurements in 2004. If the 2004 data was excluded from the regression, a clear and statistically significant upward trend ( $p = 0.037$ ) with a larger amplitude ( $0.54 \pm 0.49$  ppbv/year) would be obtained. 2004 was an unusual year during which Hong Kong and the PRD region suffered from an extremely unfavorable weather condition.<sup>31</sup> More intense sunlight and the dry weather facilitated stronger production of O<sub>3</sub> in the region.<sup>31</sup> Hence in the subsequent discussion the O<sub>3</sub>/O<sub>x</sub> trends only refer to those derived from analysis of data without the 2004 outlier. In comparison with O<sub>3</sub>, O<sub>x</sub> showed a smaller rise ( $0.27 \pm 0.63$  ppbv/year) but the trend was not statistically significant. Overall, these long-term observations indicate a rise in ambient O<sub>3</sub> and a nondecrease in O<sub>x</sub> in Hong Kong over the past decade.

Figure 1b documents the average regional background O<sub>x</sub> and locally produced O<sub>x</sub> determined by the TCEQ method in the autumn of 2002–2013. The regional background O<sub>x</sub> transported into Hong Kong composed on average approximately 70% of the DMA8 O<sub>x</sub> concentrations, and explained 75% of the measured variability in DMA8 O<sub>x</sub> (see SI Figure S5). In general, the locally produced O<sub>x</sub> only contributed about

30% to the observed DMA8  $O_x$ . To make these results in perspective, the average concentrations of both regional and local  $O_x$  on all days were estimated at 54 and 23 ppbv respectively, contributing to a total mean DMA8  $O_x$  of 77 ppbv. For the  $O_3$  nonattainment days (i.e., days with DMA8  $O_3$  exceeding 75 ppbv), in comparison, the mean regional and local  $O_x$  increased to 77 and 38 ppbv, sustaining a total average DMA8  $O_x$  of 115 ppbv. These results indicate the dominant contribution of regional transport to the ambient  $O_x$  in Hong Kong.

Figure 1b also shows opposite trends in the regional background  $O_x$  and local  $O_x$  in Hong Kong. The regional background  $O_x$  have increased at  $0.52 \pm 0.55$  ppbv/year in the past decade, though the trend was only of marginal statistical significance ( $p = 0.06$ ). A single-tailed Student's  $t$  test suggests that the probability that the trend is positive is about 0.97. The increasing amplitude also agrees well with that determined for surface  $O_3$  at Hok Tsui (a background site in Hong Kong;  $0.58$  ppbv/year) during the 1994–2007 period.<sup>16</sup> On the contrary, the locally produced  $O_x$  showed a clear and statistically significant downward trend ( $p = 0.05$ ) but with a smaller rate ( $-0.25 \pm 0.25$  ppbv/year). The above analyses reveal clearly that the local  $O_x$  has been reduced but the transported regional  $O_x$  has increased more significantly, and their combined effect resulted in a rise in  $O_3$  and a nondecrease in  $O_x$  in Hong Kong.

**3.2. Trends in  $O_3$  Precursors.** It is of interest to find the cause of reduction in the locally produced  $O_x$  in Hong Kong. For this purpose, we examined the trends of major  $O_3$  precursors obtained at AQMS. Figure 2a clearly displays the statistically significant decreasing trends in the mean concentrations of CO,  $NO_x$ , and  $NO_2$  observed at all available AQMS (i.e., 9 for  $NO_x$ , 11 for  $NO_2$  and 4 for CO) in the autumn months between 2002 and 2013: CO showed a mean rate of



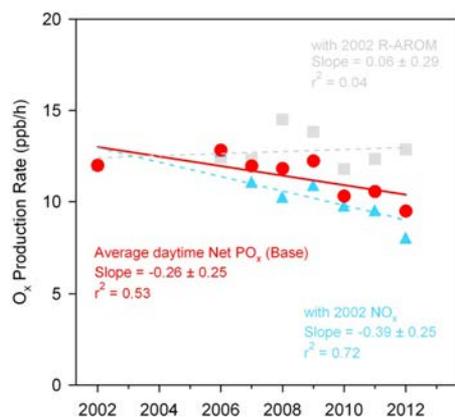
**Figure 2.** (a) Trends in autumn mean (September–November)  $NO_x$ ,  $NO_2$ , and CO observed at all available AQMS in Hong Kong. (b) Trends for major anthropogenic hydrocarbons measured at Tung Chung (the most polluted AQMS). The fit lines indicate linear regressions with slopes ( $\pm 95\%$  confidence intervals) and  $R$  squares annotated.

change of  $-12.5 \pm 10.9$  ppbv/year;  $NO_x$  and  $NO_2$  decreased by  $1.55 \pm 0.49$  and  $0.31 \pm 0.31$  ppbv/year, respectively. The trends at individual AQMS were also downward and statistically significant, despite the large variance in change rates at different sites (see SI Figure S6 and S7). The decrease in ambient concentrations of CO and  $NO_x$  is unsurprising, considering the persistent and step-by-step efforts made by the Hong Kong government in the last few decades to control and cut emissions from transportation, power plants and other industrial activities (see SI Figure S2 for reduction in the estimated emissions).

In Hong Kong, photochemical  $O_3$  production is generally more sensitive to VOCs.<sup>20</sup> Figure 2b presents the average concentrations of major NMHCs measured at Tung Chung (the only AQMS with long-term VOC observations) in the autumn months of 2002–2012. To facilitate analysis and interpretation, the anthropogenic NMHC species were categorized into four groups, namely, reactive aromatics (R-AROM; including all aromatics except for benzene), reactive olefins (R-OLF, including all alkenes other than ethene; note that the ethene data obtained from the real-time GC measurements were not reliable and that ethene only presents a minor contributor to  $O_3$  production in Hong Kong.<sup>20</sup> Thus, ethene was not considered in this analysis), alkanes with  $\geq 4$  carbons (C4HC) and hydrocarbons with low reactivity (LRHC; including ethane, propane and benzene). The ambient levels of reactive aromatics clearly declined over the period of study ( $p = 0.012$ ), with a mean change rate of  $-0.20 \pm 0.14$  ppbv/year. This is an expected result of the strict VOC-control measures undertaken by the Hong Kong government. In April 2007, for instance, Hong Kong implemented the VOC Regulation, which was designed to control the VOC content (mainly aromatics) of architectural paints/coatings, printing inks, and six broad categories of consumer products. This Regulation was amended in October 2009 to extend control to additional products with high aromatic-VOC content. In comparison, despite the various control measures implemented by the government, we observed no significant trends in the concentrations of other NMHC groups (i.e., R-OLF, C4HC, and LRHC). The use of liquefied petroleum gas, which was not under effective control in Hong Kong until 2013, may be responsible for the sustained levels of certain VOC species (e.g., alkenes and butanes). Overall, these observations confirm that the ambient levels of CO,  $NO_x$ , and aromatic VOCs in Hong Kong have decreased in response to local control efforts. In the next section, we further evaluate their contributions to the reduced local  $O_3$  production by the OBM analysis.

**3.3. Changes in Local Ozone Production.** Figure 3 presents the trends in the OBM-calculated daytime-average (09:00–18:00 LT) net  $O_x$  production rates at Tung Chung for the years of 2002 and 2006–2012, during which the VOC measurements were available. Consistent with the observed decline in the locally produced  $O_x$  concentrations, the in situ net  $O_x$  production rate showed a clear and statistically significant downward trend ( $-0.26 \pm 0.25$  ppbv/h/year,  $p = 0.04$ ; see the red solid circles). This finding confirms the reduction in local ozone production in Hong Kong.

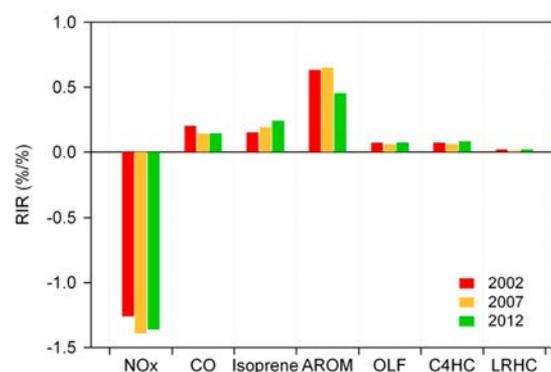
To evaluate the impact of the  $O_3$  precursor changes on local  $O_3$  production, a number of sensitivity model runs were conducted for the 2006–2012 period, with 2002 as a baseline. The base models for 2006–2012 were rerun with most of their parameters unchanged but with constraints to target  $O_3$  precursors based on the 2002 data (i.e., assuming no change



**Figure 3.** Trends in daytime-average net  $O_x$  production rate calculated by the OBM at Tung Chung. The red circles indicate the true trends from the base models, and the gray squares and sky-blue triangles show the “trends” if the reactive aromatics or  $NO_x$  unchanged with respect to the year 2002. The fit lines denote linear regression with slopes ( $\pm 95\%$  confidence intervals) and  $R$  squares annotated.

in each target precursor since 2002). The differences in the net  $O_x$  production rates between the sensitivity and base runs can thus be ascribed to the changes in target precursors. The results are also presented in Figure 3. It is clear that if the levels of reactive aromatics had remained the same from 2002 onward, the net  $O_x$  production rates between 2006 and 2012 would have been significantly higher than those determined from the base runs (see gray squares), and the trend of decreasing in situ  $O_x$  production would have been broken. For example, if the levels of R-AROM had not decreased between 2002 and 2012 (despite decreasing levels of the other species), the net  $O_x$  production rate in 2012 would have been  $\sim 12.9$  ppbv/h, which is much higher than the result given by the base model ( $\sim 9.5$  ppbv/h), and comparable to the 2002 result ( $\sim 12.0$  ppbv/h). This indicates that the reduction of R-AROM should be the key factor leading to decreasing local  $O_3$  production in Hong Kong. In contrast, if the  $NO_x$  concentration had remained at the level observed in 2002, the net  $O_x$  production rates between 2006 and 2012 would have been even lower than the base results (see blue triangles), and a more significant decrease in local  $O_3$  production would have been established at Tung Chung. In comparison, variations in ambient CO, R-OLF, C4HC, and LRHC levels had little effect on local  $O_3$  production, as indicated by the comparability of the sensitivity and base model results for  $O_x$  production rate when these precursors were examined (figure not shown).

It is also critical to the future  $O_3$  pollution control to determine whether the chemical regime of local  $O_3$  production has altered in response to the changing levels of ozone precursors. We calculated by the OBM the relative incremental reactivity (RIR) of major  $O_3$  precursors at Tung Chung in 2002, 2007, and 2012, to identify the changes in local  $O_3$  production regime over the past decade. RIR is defined as the ratio of decrease in  $O_x$  production rate to decrease in precursor concentrations, and can be used to estimate the effects of a given emission reduction on ambient  $O_3$  concentrations.<sup>32</sup> The OBM-calculated RIRs for the major  $O_3$  precursors are shown in Figure 4. Two aspects of the figure are of interest. First, there was no qualitative change in the  $O_3$  production regime derived at Tung Chang between 2002 and 2012.  $O_3$  production was found to be VOC-limited, particularly sensitive to reactive aromatics, and  $NO_x$  generally had a negative influence on



**Figure 4.** OBM-calculated RIRs for major  $O_3$  precursors at Tung Chung in 2002, 2007, and 2012.

attempts to control  $O_3$  production (due to the titration effect). These characteristics are consistent with those determined at five sites (including Tung Chung) in Hong Kong in 2002.<sup>20</sup> Second, the RIR of R-AROM showed a considerable quantitative decrease between 2007 (RIR = 0.65) and 2012 (RIR = 0.45). This is attributable to the reduction in reactive aromatics in Hong Kong (see Figure 2b). Based on the analysis, it is concluded that further reducing the levels of reactive aromatics is currently still the most effective way of minimizing local  $O_3$  production in Hong Kong.

#### 4. DISCUSSION AND IMPLICATIONS

As evidenced by the present study, although the control measures implemented by the Hong Kong government have effectively mitigated local  $O_3$  formation, these efforts have been negated by the increase of regional influences. The rise in regional background  $O_3$  levels is believed to be due to the increase in  $O_3$  levels in the PRD region, along with the contributions of “super-regional” transport from the upwind regions beyond the PRD, that is, eastern China. Over the last few decades, the anthropogenic emissions of  $NO_x$  and VOCs have sharply increased across eastern China due to economic expansion and increasing energy use.<sup>33–35</sup> Indeed, it has been confirmed that the atmospheric  $O_3$  levels in several of the most urbanised regions of eastern China, namely the North China Plain, the Yangtze River Delta and the PRD, have shown an upward trend in the past decade.<sup>13,16,36–38</sup> This increase in “background”  $O_3$ , due mainly to regional (PRD-originating) and super-regional (eastern China based) contributions, has offset the reduction of local ozone production in Hong Kong, leading to an overall increase in ambient  $O_3$  levels. However, without local control efforts would have definitely resulted in a more significant increase in ambient  $O_3$  levels in Hong Kong. To successfully control its ozone problem in the future, therefore, Hong Kong must collaborate closely with mainland China, not only the PRD but also the entirety of eastern China.

This study also has important implications for efforts to reduce secondary pollution in other Chinese cities. The findings of recent studies have indicated that ozone pollution is becoming more extensive and more severe in the major urban-industrialized regions of China,<sup>12,14,39</sup> and the ozone problem will be undoubtedly among China’s key environmental management targets in the near future. In the past, China’s air-pollution control has been limited to a few megacities (Beijing, Shanghai, and Guangzhou), partly due to more imminent needs to clean its air for hosting the 29th Summer Olympics, the 40th

World Expo., and the 16th Asian Games, respectively. Although these measures have helped to lessen primary pollution, they do not seem to have been sufficient in reducing ozone and other secondary pollutants.<sup>40,41</sup> Even the unprecedentedly stringent pollution-control measures implemented in the Beijing Municipality before the Olympics in summer of 2008 did not significantly reduce ozone and sulfate aerosol pollution, due to the strong influence of regional pollution sources south of Beijing.<sup>41</sup> The long-term data analyzed in the present study in Hong Kong vividly illustrate the importance of dealing with pollution sources over a large geographical area. As many other cities in central and eastern China are closer to regional sources of pollution, it is vital to develop and implement coordinated multiregional policies to mitigate ozone pollution and other secondary problems in these cities.

## ■ ASSOCIATED CONTENT

### ● Supporting Information

Table S1; Figures S1–S7; description of the measured hydrocarbon and OVOC species; description of the observation-based box model; results of sensitivity modeling analysis with real-time alkenes measurements; impact of the HONO treatment on the OBM analyses. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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### Notes

The authors declare no competing financial interest.

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