Source apportionment and health effect of NO_x over the Pearl River Delta region in southern China

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Abstract:

As one of the most notorious atmospheric pollutants, NO_x not only promotes the formation of ozone but also has adverse health effects on humans. It is therefore of great importance to study the sources of NO_x and its effects on human health. The Comprehensive Air Quality Model (CAMx) modeling system and ozone source apportionment technology (OSAT) were used to study the contribution of NO_x from different emission sources over southern China. The results indicate that heavy duty diesel vehicles (HDDVs) and industrial point sources are the two major local NO_x sources, accounting for 30.8% and 18.5% of local NO_x sources, respectively. In Hong Kong, marine emissions contributed around 43.4% of local NO_x in 2011. Regional transport is another important source of this pollutant, especially in February and November, and it can contribute over 30% of ambient NO_x on average. Power plant point emission is a significant regional source in Zhuhai, Zhongshan and Foshan. The total emission sources are estimated to cause 2,119 (0–4,405) respiratory deaths

and 991 (0–2,281) lung cancer deaths due to long-term exposure to NO_x in the Pearl River Delta region. Our results suggest that local governments should combine their efforts and vigorously promote further reduction of NO_x emissions, especially for those sources that make a substantial contribution to NO_x emissions and affect human health: HDDV, LDGV, industrial point sources and marine sources.

Capsule Abstract: Result indicated that heavy duty diesel vehicle and industrial point source make a substantial contribution to ambient NO_x concentration in this region.

Key Words: NO_x, CAM_x, OSAT, C-R Function, Mortality

1 Introduction:

The Pearl River Delta (PRD) in southern China is experiencing dramatic economic growth due to the reform and opening-up policy. The rapid industrialization and urbanization associated with this growth have caused serious local and regional air pollution issues in the area, including ambient NO_x pollution. Developed countries have already paid attention to this pollutant and NO_x emissions at the beginning of this century had been reduced by 26.4% in Europe and 17.5% in the US compared to 1980 levels (Palmgren, 2004). Contrary to the decreasing trend in Western countries, emissions of NO_x in China have increased gradually for the past three decades. The annual growth rate for NO_x emissions was 7.3% between 1997 and 2006 (Stavrakouet al., 2008). Ambient NO_x is listed in the 12th five-year plan (years 2011–2015) of the Chinese government as a significant pollutant for emission control. In the PRD region, NO_x has been an issue of great concern and as far back as 2002, the Hong Kong and Guangdong governments agreed to reduce emissions of this pollutant, according to Pearl River Delta Air Quality Management Plan Mid-term Review (PRDAQMIG, 2007).

NO_x is a precursor of nitrates in particulate matter and like other ambient pollutants it can cause premature death by inducing lung cancer and respiratory disease. It is also an important precursor for ozone formation via a series of photo-oxidation reactions

with volatile organic compounds (VOCs). Ozone can cause acute respiratory problems and reduce lung capacity and as such it is extremely harmful to human health. It can also promote the oxidation of SO_x (NO_x) to sulfate (nitrate) by increasing the oxidation capacity of the atmosphere. When precipitation events take place, nitrates dissolves in rainwater and cause acid rain, which is also a notorious pollution problem in this region (Lu et al., 2015). The damage caused by acid rain can include changes in the leaching rate of nutrients from plants and soil, acidification of aquatic environments and corrosion of buildings. Therefore, after the achievement of SO_2 reduction during the 11th five-year plan, NO_x emission regulations have been further tightened [MEP, 2011] in the 12th five-year plan. The retention time of NO_x in the atmosphere is around one day and it can cross city borders and influence other places, depending on meteorological conditions. Therefore, it is important to look at city-based NO_x pollution so that NO_x control strategies can be formulated and the control effects can be further assessed.

One way of solving the environmental issues mentioned above, and improving the quality of life of residents in the PRD region, is to decrease ambient NO_x over this area. However, the air pollution problem is a regional issue and cannot be solved by single city. Cities need to identify where the NO_x is coming from so that the local governments can design NO_x control strategies.

Source apportionment techniques in emission-based air quality models can be used to track the sources of ambient pollutants by both region and source type. Zhang and Ying (2011) apportioned NO_x contributions to ozone formation by region and source category by using source apportionment techniques with CMAQ in Texas. Wang et al. (2014) integrated the particulate source apportionment technique (PSAT) with CAMx to track the sources of nitrate, sulfate, ammonium and elemental carbon in Shanghai during haze episodes. Wu et al. (2013) applied PSAT integrated with CAMx to track the power plant, industrial boiler, mobile, area and marine sources of respirable

particulate matter over the PRD region in April and December. Li et al. (2013) used the ozone source apportionment technique (OSAT) coupled with CAMx to evaluate ozone control policies systematically and found that local reductions from point sources, mobile sources and area sources can help to reduce episodic ozone concentrations. Until now, most source apportionment studies have mainly focused on ozone and respirable particulate matter. No NO_x source apportionment study has ever been conducted in China.

Since air pollutant can directly exert adverse health effect on human beings, some studies have been carried out in China to evaluate the impact. Huang et al. (2012) applied point observation data together with concentration response function to evaluate the health burden caused by PM_{10} in PRD region during 2006. They found that long-term exposure to PM_{10} caused 12,786 premature death in this region. Wang et al. (2015) applied WRF-CMAQ modeling system to calculate the $\text{PM}_{2.5}$ related mortality in Yangtze River Delta region. Their results showed that the short-term premature mortality due to $\text{PM}_{2.5}$ was 13,162 in 2010. Up to now, most of air pollutant related health burden estimation studies are focused on particulate matter in China, but few had paid attention to NO_x . Chauhan et al. (1998) found that nitrogen oxides increased the susceptibility to respiratory infections. Chaloulakou et al. (2008) stated that people suffering from respiratory diseases were sensitive to NO_2 at high concentration level. Clark et al. (2010) found that ambient NO is positively associated with a higher incidence of childhood asthma. Moreover, Nafstad et al. (2004) and Cao et al. (2011) calculated the relative risk of exposure to NO_x for lung cancer and respiratory disease via long-term cohort study. Hence, it is necessary and important to evaluate the health burden attributed to NO_x in PRD region.

In this study, we applied the WRF-SMOKE-CAMx air quality system integrated with OSAT to understand the contributions to ground NO_x from different source categories and region receptors in the PRD region. Section 2 describes the model domain setting,

choice of physical/chemical scheme and OSAT. In section 3, we present the evaluation of our chemical transport model and discuss the source apportionment result. We also assess premature deaths related to long-term NO_x exposure from different emission sources in this section. The overall study is summarized in section 4.

2 Model and Methodology:

2.1 Model description

The meteorology field in our study was simulated by Weather Research Forecast v3.2 (WRF) following the settings of Wu et al. (2013). We used the WRF single-moment 6-class scheme (Dudhia et al., 2008) for microphysics, the Grell–Devenyi ensemble cumulus parameterization scheme (Grell and Devenyi, 2002), the rapid radiative transfer model longwave radiation scheme (Mlawer et al., 1997), the Yonsei University PBL scheme (Hong et al., 2006), Dudhia’s (1989) shortwave radiation scheme and the Noah land–surface model (Chen and Dudhia, 2001). Observation data from Hong Kong meteorological stations were nudged to domain 3. For the CAMx v6.00 scheme setting, we applied Euler Backward Iterative (EBI) for the chemical solver, K-theory for vertical diffusion, the RADM scheme for aqueous phase chemistry, CB05 for gas phase chemistry, ISORROPIA 1.7 (Nenes et al., 1998) for the inorganic aerosol scheme and the updated SOAP scheme for secondary organic aerosol simulation.

As Figure 2 shows, the domain setting for the modeling system contains three nested meshes. The grid resolutions were 27 km for domain 1 (D1), 9 km for domain 2 (D2) and 3 km for domain 3 (D3). The domain for the meteorological field is larger than the chemical transport model domain, which can help to minimize the boundary effect in the simulation. The boundary condition for the outmost domain 1 was extracted from GEOS-Chem to better match the Asian background concentration. Domain 1

covered most of China and Japan, Thailand and Korea. Domain 2 covered the Guangdong Province and the smallest domain covered all the 10 major cities in the PRD region including Hong Kong and Macau Special Administrative Region.

For the emission inventory, INTEX-B 2006 regional emission inventories were used for the area outside the PRD region (domains 1 and 2) (Zhang et al., 2009). For the PRD region, a highly resolved emission inventory for 2010 was implemented (Zheng et al., 2009; Yin et al., 2015). Model of Emissions of Gases and Aerosols from Nature (MEGAN version 2.04) was used to process and map biogenic emissions (Guenther et al., 2006). Figure 1 shows the NO_x emissions in moles per second for domain 3. Shenzhen, Guangzhou and Hong Kong had more NO_x emissions than the other seven cities in this region. The emission data were distributed temporally and spatially using the Sparse Matrix Operator Kernel Emissions (SMOKE version 2.4) modeling system.

2.2 Ozone Source Apportionment Technology (OSAT)

OSAT hosted in CAMx can track the emission contributions to ozone, VOCs and NO_x at each grid cell from emissions “source grouping,” initial conditions, and boundary conditions by adding additional reactive tracer species. Here, a source grouping refers to a group that combines the source categories from a specific source region (ENVIRON 2015). This technique is inherently mass consistent with standard CAMx simulations. The tracer species update is based on the CAMx simulated NO_x concentration and the tracer contribution to the sum of the NO_x tracers in the former time step, as shown in equation (1) and (2). Where N_i is the NO_x tracer for source i , N_i^{new} is the tracer for source i in the new time step, N_i^{old} is the tracer for source i in previous time step.

$$\sum N_i = \text{NO} + \text{NO}_2 \quad (1)$$

$$N_i^{new} = N_i^{old} + \Delta NO_x \times \frac{N_i^{old}}{\sum N_i^{old}} \quad (2)$$

2.3 Source region and source category

Targeting a city-based analysis, we divided the D3 domain into 12 source receptor areas: Guangzhou, Shenzhen, Foshan, Dongguan, Huizhou, Zhongshan, Jiangmen, Zhuhai, and Zhaoqing, Hong Kong, the PRD water region and the region outside the Pearl River Economic Zone (PREZ) (see Figure 3). The mobile source includes seven types of vehicle in the emission inventory. Based on the amount of NO_x emissions in the whole region, as shown in Table 1, we chose HDGV (heavy duty gasoline vehicle), LDGV (light duty gasoline vehicle), LDDT (light duty diesel truck) and HDDV (heavy duty diesel vehicle) for tracking. The other three mobile sources, LDDV (light duty diesel vehicle), LDGT (light duty gasoline truck) and motorcycle, were combined with area source and labeled as A-M (Area-Mobile) because the NO_x emission of each single category is relatively small in the major cities when compared to the sources we chose to track separately. The other tracking source categories were biogenic emission, industrial point source (boiler), power plant and marine vessel. The source contribution from the local city is defined as the local contribution. Other sources within D3 but outside particular cities are defined as regional contribution and the sources outside the PRD region are defined as super-regional contribution. This can be expressed by the two equations below, where j is the source category, i is the source region, k is the receptor city and $S(i,j)$ represents the NO_x contribution from city i and source category j . $D3_total(k)$ is the contribution from all the cities in PRD region to city k and $D3_out(k)$ is the super-regional contribution to city k . S represents the local source when $i = k$ and it represents the regional source when $i \neq k$. BC represents the contribution from the boundary condition.

$$D3_total(k) = \sum_{i=1}^{11} \sum_{j=1}^8 S(i, j) \quad (3)$$

$$D3_out(k)=BC + IC + \sum_{j=1}^8 S(12, j) \quad (4)$$

2.4 The C-R function

In this study, we applied the concentration–response (C-R) function to calculate long-term respiratory mortality and lung cancer mortality caused by exposure to NO_x over the PRD region. This approach has been applied to estimate adverse health effect caused by ambient pollutants in US and Europe (Künzli et al., 2000; Hubbell et al., 2005). The C-R function for each grid cell can be expressed as Equation 5 where Δy represents the number of excess deaths caused by exposure to NO_x and y_0 is the baseline incidence rate for respiratory death and lung cancer death:

$$\Delta y = y_0(e^{\beta \Delta x} - 1)Pop \quad (5)$$

We applied the values from 2011 China Statistical Yearbook (www.stats.gov.cn/tjsj/ndsj/2011/indexeh.htm). The C-R function coefficient β is derived from the relative risk ($\beta = \ln(RR)/C$) reported in Cao et al. (2011). In their work, each 10 $\mu\text{g}/\text{m}^3$ of NO_x was associated with a 2.6% (95% CI: -0.2%, 5.6%) increase in respiratory mortality and a 2.7% (95% CI: -0.9%, 6.5%) increase in lung cancer mortality. *Pop* represents the population data, which we took from the Landscan project (Bright et al., 2011). The C-R function coefficient in the referenced study was for the population over 30 years old, hence, we derived the percentage of the population over 30 years old in different cities from Xie et al. (2011) and multiplied the factor by the Landscan population data for each city. Δx is the difference in average NO_x concentrations from different emission sources.

3 Results and Discussion

We performed numerical simulations for February, May, August and November 2011 to represent seasonal variation. These four months represent winter, spring, summer and winter respectively in this region. We realized that it would be better if the whole year can be simulated, however, due to the computational resource limitation, we have

to simulate four months to represent the whole year condition. The reason for us to simulate the year 2011 is mainly due to two reasons: 1, the year 2011 is not far from now; 2, the bottom-up inventory we used is based on 2010 PRD emission scenario.

3.1 Model evaluation

Table 2 lists the statistical results for 37 stations for surface temperature ($^{\circ}\text{C}$), wind speed (ms^{-1}) and wind direction (degree) in four months. The locations of the weather stations are shown by green triangles in Figure 3. The index of agreement (IOA) formulation for wind direction is different from the IOA formulation for other scalar quantities and follows Kwok et al. (2010). For ground temperature results, the IOA ranges from 0.73 to 0.82 and the mean bias ranges from -0.37 to 0.17. For the wind speed, the index of agreement ranges from 0.59 to 0.72 and the mean bias ranges from -0.05 to 0.45. Our meteorological simulation results are comparable to other studies (Huang et al., 2005; Wu et al., 2013; Li et al., 2013).

Table 3 compares observational data with the model simulation values. The locations of the air quality stations are shown in Figure 3. Data for NO_x , NO_2 and O_3 are available at 14 stations in Hong Kong. Another 23 stations in the PRD region only have NO_2 data for the model simulation period. These 37 stations were used to validate the hourly model results in this study. Seven statistics metrics—the mean observation value (Mean-OBS), mean model simulation value (Mean-SIM), root mean square error (RMSE), normalized mean error (NME), normalized mean bias (NMB), mean bias (MB) and index of agreement (IOA)—were used to evaluate the simulation results for these three ambient pollutants. Across the four months, the IOA of NO_x ranged from 0.54 to 0.65 and its NMB ranged from -0.22 to 0.09; the IOA of NO_2 ranged from 0.58 to 0.67 and its NMB ranged from -0.25 to -0.05; the IOA of O_3 ranged from 0.74 to 0.88 and its NMB ranged from -0.21 to 0.05. Figure 4 is the scatter plot of NO_x at the Kwun Tong site in the four months. As shown in this figure,

the CAMx simulation reproduced the temporal changes of NO_x fairly well and simulated and observed NO_x were mostly within 1:2-2:1 reference line. In general, the modeling system produces a good simulation of the magnitude and temporal variations of ambient NO₂, NO_x and O₃. Our simulation result is comparable to other modeling studies in this region (Huang et al., 2006; Kwok et al., 2010; Wu et al., 2012; Li et al., 2013; Lu et al. 2015).

3.2 Local, regional and super-regional sources

Table 4 shows the local, regional and super-regional contributions to NO_x in the PRD region in February, May, August and November 2011. On average, the local contribution ranges from 42.9% (November) to 60.8% (May), the regional contribution ranges from 35.5% (August) to 41.3% (February), and the super-regional contribution ranges from 3.7% (August) to 19.4% (November). The super-regional contribution is much less important for NO_x than for PM_{2.5} and ozone. Wu et al. (2013) found that the super-regional contribution of PM_{2.5} was over 30% in April and over 50% in December in the PRD region. Li et al. (2012) reported that in November, the average super-regional contribution of ozone in Hong Kong, Jiangmen and Guangzhou was around 71%. This is mainly because ozone and PM_{2.5} are typically secondary pollutants, which may or may not be formed from their primary precursors immediately after they are emitted. Also, the lifetime of ozone and PM_{2.5} is in the order of weeks (Taylor et al., 2013) whereas the lifetime of NO_x is approximately one day only (Jacob, 1999). A longer lifetime favors the long range transport of pollutants and the super-regional contributions of related pollutants in downwind areas will thus be increased.

Local NO_x contributions are particularly important and generally account for over 50% in the major cities of Guangzhou, Shenzhen and Hong Kong. Regional NO_x contributions for cities vary seasonally, ranging from 11.7% to 38.7%. For example,

both Zhuhai and Zhongshan are heavily influenced by regional contributions in February and November, as shown in Table 4. In these two months, these two cities are located downwind direction of Shenzhen, Dongguan and Guangzhou, in each of which the local emission of NO_x is large. Northerly winds bring NO_x from these three cities to Zhongshan and Zhuhai. In May and August, Shenzhen is heavily affected by regional sources of NO_x and the regional contribution reaches 36.3% and 44.1%, respectively. The prevailing wind direction in the PRD region is south-easterly in May and south-westerly in August, so NO_x is blown up from Hong Kong and influences Shenzhen.

3.3 Local source category analysis for each city

Local emissions are generally the dominant source of NO_x , the primary pollutant in the atmospheric environment. This section investigates the contributions from individual categories to the total local emissions. As shown in Figure 6, HDDVs contributed a substantial percentage of NO_x in all the cities, accounting for 12.9% to 58.1% of local contributions. Shenzhen was particularly heavily influenced by HDDVs (58%). Other cities, such as Guangzhou, Dongguan and Zhongshan, were also greatly affected, with around 30% of NO_x in these cities from HDDV. In less developed cities, such as Zhaoqing, HDDV had made a smaller contribution (13.9%). Three other mobile sources—HDGVs, LDGVs and LDDTs—did not contribute much ambient NO_x when compared to HDDV. The sum contribution of these three sources ranged from 7.7% (Hong Kong) to 27.5% (Guangzhou) across the ten cities and is smaller than the contribution from HDDVs in all the cities. This is mainly because the emission factor for HDDVs is around 4 times larger than the emission factor for the other three vehicle types in this region (Che et al., 2009). The average annual travel distance for HDDVs is also the largest of the four vehicle types.

As Figure 6 shows, industrial point sources accounted for a large proportion of local

NO_x emissions in Huizhou, Foshan, Jiangmen and Zhaoqing, having an average percentage of 18.7%, 30.3%, 25.4% and 44.8%, respectively. Power plants contribute much less than industrial point sources at ground level, and their average contribution across the ten cities is only 9.0% of local contributions. Marine sources are important in Hong Kong. As Figure 1 shows, a substantial amount of marine NO_x was clustered in southern and eastern Hong Kong waters. In Hong Kong, marine emissions contributed around 43.4% of local NO_x in 2011; local HDDVs only contributed 16.2%. In addition to anthropogenic sources, soil micro-bacteria can emit nitric oxide. In MEGA, this process is mainly controlled by temperature and soil moisture. According to our result, this natural source contribution to NO_x pollution ranges from 0.1% to 1.4% in the major cities of the PRD region except Huizhou (8.0%), Jiangmen (5.6%) and Zhaoqing (6.6%) in summer, where urbanization is low when compared to the major cities. Biogenic NO_x emissions are not negligible in these three cities.

3.4 Regional source category analysis for each city

Regional sources of NO_x are important in some cities and it is necessary to further analyze which city contributes most to pollution in a target city. In this section, we analyze the five cities which are heavily influenced by regional NO_x contributions: Foshan, Dongguan, Shenzhen, Zhongshan and Zhuhai. As Figure 7 shows, HDDVs are an important regional NO_x source in these five cities, especially Dongguan, which is located on the western side of Shenzhen. Although power plants did not contribute much to local NO_x pollution, they are an important regional source of NO_x. This is mainly because all the cities in the PRD region built their power plants at their borders, so NO_x coming from elevated power plants is easily dispersed to surrounding cities instead of influencing the source city. Marine emissions are an important regional source in Shenzhen, largely contributed by Hong Kong and PRD waters,

especially in summer when the predominant wind is southerly. Our result indicates that the city governments should control the number of HDDVs, because this source category contributes substantial amounts of ambient NO_x to regional and local environments over the PRD area.

As Table 5 shows, the regional contribution from Guangzhou to the other four cities (except Shenzhen) is more than 10% in each of the four months. In Foshan, the regional contribution of NO_x from Guangzhou reaches 78% of total regional contributions in November. As Figure 7 shows, Guangzhou accounts for over 60% of the regional HDDV and industrial point contributions of NO_x to Foshan in February, May and November. The city center of Guangzhou, which emits substantial amounts of NO_x (Figure 1), is close to the border with Foshan and NO_x emitted from the center is easily transported to Foshan. Hong Kong is an important NO_x contributor for Dongguan, Zhuhai and Shenzhen in May and August under the prevailing southerly wind. As Table 5 shows, NO_x from Hong Kong accounted for 60% of regional contributions to Shenzhen. As mentioned above, Shenzhen already suffered from substantial local NO_x emissions, so with the regional NO_x contribution from Hong Kong, the monthly NO_x concentration in this city in summer and autumn was the highest of all cities over the PRD region. As mentioned in section 3.2.1, regional NO_x contributions accounted for over 60% of NO_x in Zhuhai and Zhongshan in February and November due to the prevailing wind (Figure 5). In Table 5 and Figure 7, we can see that power plant point sources from Dongguan, and HDDV emissions from Shenzhen and Guangzhou are the main regional NO_x sources for Zhuhai and Zhongshan, where NO_x contributions from other cities are greater than the local NO_x contribution. Hence, besides reducing NO_x emissions in their own city, each city's government, and especially Zhuhai and Zhongshan, should communicate with the neighboring city that contributes most ambient NO_x pollution and combine their efforts to solve the issue.

The source apportionment results from OSAT are influenced by several uncertainties, which lead to biases in the NO_x pollution simulation. The NO_x source apportionment is also subject to uncertainties from the meteorological model performance and emission inventory, because the chemical reaction is relatively simple compared with other typical secondary pollutants (i.e., PM_{2.5}). The possible over-estimation of surface wind speed may lead to over-estimation of super-regional and/or regional contributions. Uncertainties in the inventory may lead to uncertainty in the contributions from different source regions. The understanding of NO_x emission sources and the bottom-up estimation of NO_x pollution is acceptable, with much smaller uncertainties than those for other pollutants (e.g., VOCs). In addition, instead of estimating by single grid, the results are estimates based on city and regional averages to reduce the possible over-estimation of the regional transport contribution. Therefore, we believe these uncertainties will not influence our major conclusion that local and regional sources are the main contributors to NO_x pollution in PRD cities.

3.5 Health impact of NO_x over the PRD region

Besides being an important precursor of ozone and particulate matter, NO_x can have direct adverse effects on human health. It is therefore of great importance to quantify mortality caused by ambient NO_x pollution from different sources and provide scientific references to local governments.

Estimates of premature mortality due to long-term exposure to NO_x from different sources in each PRD city is shown in Table 6. The results show an estimated 2,119 (0–4,405) premature deaths from respiratory causes annually and 991 (0–2,281) deaths from lung cancer death due to exposure to NO_x from the all the emission source categories in the overall PRD region. At the city level, annual premature mortality is highest in Hong Kong, with 773 (0–1,589) respiratory deaths and 361 (0–

821) lung cancer deaths. Premature mortality from these causes is lowest in Zhaoqing, which has lung cancer mortality of 9 (0–20) and respiratory mortality of 18 (0–39). HDDVs contributed most to premature mortality in this region and the number of deaths attributable to this source in the ten cities reached 684 (0–1,406) for respiratory causes and 320 (0–726) for lung cancer. The contributions from other source categories varied in the different cities. For example, marine emissions caused 154 (0–319) respiratory deaths and 72 (0–165) lung cancer deaths in Hong Kong. However, the contribution from marine sources is relatively low when compared to other sources in the other nine cities over the PRD region.

Although several studies have been carried out to calculate the health burden in PRD region, most of them were focused on all-cause mortality attributed to particulate matter (Huang et al., 2012; Xie et al., 2011; Ding et al., 2015). Only limited number of studies focused on air pollution caused respiratory mortality and lung cancer mortality estimation in this region. Lelieveld et al. (2013) calculated that the excess PM_{2.5} attributed lung cancer mortality was 3,530 and O₃ attributed respiratory mortality was 9,713 in this region. The short-term mortality caused by NO_x has not yet been calculated in this region by other studies. According to Shang et al. (2013), the acute excess risk of respiratory mortality caused by NO₂ is 1.62% for a 10µg/m³ increase of NO₂, which is larger than the values for PM_{2.5} (0.51%) and O₃ (0.73%).

There are three types of uncertainty in the estimation of premature mortality due to NO_x exposure. The first one is the C-R function coefficient (β), which was derived from epidemiological data from several different cities in China. Medical conditions vary in different locations, so uncertainty exists when we applied the coefficient to the whole of China. The second uncertainty is from the population data, which is based on sub-national level census, related algorithm and remote sensing data. Hence, the overall uncertainty for the whole dataset is hard to quantify. The last uncertainty is due to the baseline incidence rate, because lacking local data, we could only apply the

value for the whole of China (taken from the 2011 China Statistical Yearbook). Although uncertainty exists for the premature mortality estimation, there is no doubt that NO_x exposure is harmful to human health, and local governments should further enforce solid control policies for this pollutant as quickly as possible to improve air quality in this region.

4. Conclusion

In this study, we applied the WRF-SMOKE-CAMx modeling system coupled with OSAT to assess NO_x source apportionment in the PRD region. Our results show that unlike secondary ambient pollutants, the super-regional contribution of NO_x is relatively small when compared to local and regional sources. Local sources account for the largest contribution to NO_x pollution in the PRD region (42.9–60.8%), the regional contribution is relatively stable (35.5–41.3%), and the contribution from super-regional sources is limited. HDDVs and industrial point sources contributed 30.8% and 18.5%, respectively, to local NO_x sources in the cities within the PRD region. Marine sources did not influence most of the cities, but accounted for a substantial amount of NO_x in Hong Kong. Regional transport is another important source for this pollutant, especially in February and November, when the prevailing northerly wind transports the heavy pollutant to downwind cities (e.g., Zhongshan and Zhuhai). We used the concentration response function to estimate the long-term premature mortality caused by exposure to ambient NO_x over this region. To provide a scientific basis for pollution control policies, mortality due to eight major emission sources was analyzed. Among the different emission categories, HDDVs contributed most to premature mortality associated with NO_x exposure, accounting for around 42% of respiratory deaths and 41% of lung cancer deaths. In addition to local source contribution, regional contribution is a great concern for most of the cities in the region. Our results suggest that even though tighter NO_x emission regulations have been applied in the 12th five-year plan, local governments should combine their

efforts and vigorously promote further reduction of NO_x emissions, especially for those sources that make a substantial contribution to NO_x concentration and affect human health (i.e., HDDVs, LDGVs, industrial point sources and marine sources). Considering linear response, we can design how to achieve the required reduction percentage based on the source apportionment results. However, due to the nonlinear response between the local concentrations to emission control, it is hard to quantify the exact required reduction of the culpable sources to achieve the purpose without model sensitivity run. Scenario simulation by using brute force method or decoupled direct method needs to be carried out in the future to study how the ambient pollutant concentration responds to 10%, 20%, 30%, 40%, and even more NO_x emission reduction uniformly in the whole PRD region theoretically in the future. It is also important to consider available state-of-art control measures for different source sectors to design effective control strategies with minimum costs.

The OSAT results show a large difference compared to the apportionment solely based on the emission inventory (Table 1), which is not detailed enough to guide the implementation and evaluation of NO_x control regulations, especially because it does not consider the significant regional transport impact (35.5–41.3%) in the atmosphere. C-R function analysis based on the OSAT results, to quantify mortality caused by ambient NO_x pollution from different sources, provides more scientific references for local government guidance on air quality management.

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Table 1: Yearly NO_x Emissions in the PRD Region

Yearly NO _x Emission (ton/yr)	
Mobile:	
HDDV	1.9×10^5
HDGV	3.6×10^4
LDDT	2.3×10^4
LDDV	1.6×10^4
LDGT	1.2×10^4
LDGV	7.8×10^4
MC	8.4×10^3
Area:	9.6×10^4
Point:	
Industry	1.9×10^5
Energy	3.8×10^5
Marine:	1.1×10^5

Table 2: Hourly WRF performance matrix for temperature and wind in 2011

		RMSE	NMB	IOA	MB
Feb	Wind speed	1.8	0.29	0.7	0.1
	Wind direction	-	-0.16	0.88	-22.7
	Temperature	3	0.12	0.81	1.7
May	Wind speed	1.6	0.22	0.72	-0.05
	Wind direction	-	-0.07	0.9	-12.6
	Temperature	2.2	0.005	0.78	0.04
Aug	Wind speed	1.4	0.37	0.68	0.26
	Wind direction	-	0.05	0.88	3.6
	Temperature	2.2	-0.009	0.73	-0.37
Nov	Wind speed	2.1	0.45	0.59	0.45
	Wind direction	-	-0.22	0.89	-32.7
	Temperature	3.1	0.12	0.82	1.7

Table 3: Hourly CAMx model performance for NO_x, NO₂ and O₃ in 2011

		Mean-OBS	Mean-SIM	RMSE	NME	NMB	MB	IOA
Feb	NO _x	49.1	39.9	35.5	0.53	-0.22	-9.2	0.63
	NO ₂	32.0	27.7	18.4	0.47	-0.12	-4.3	0.67
	O ₃	30.7	27.3	16.2	0.41	-0.10	-3.4	0.74
May	NO _x	37.7	36.8	26.5	0.53	0.09	-0.9	0.65
	NO ₂	24.8	20.6	15.2	0.47	-0.05	-4.2	0.65
	O ₃	34.3	33.6	17.9	0.38	-0.01	-0.7	0.88
Aug	NO _x	39.5	35.7	29.0	0.56	-0.12	-3.8	0.54
	NO ₂	24.0	18.4	16.3	0.52	-0.24	-5.6	0.58
	O ₃	29.4	30.4	22.0	0.54	0.05	1.0	0.80
Nov	NO _x	41.2	32.1	28.7	0.49	-0.22	-9.1	0.63
	NO ₂	31.3	23.4	17.4	0.44	-0.25	-7.9	0.66
	O ₃	32.1	23.4	16.3	0.39	-0.21	-8.7	0.76

Table 4: Local, regional and super regional contribution of NO_x at 10 PRD cities

	Unit:ppb	HZ	GZ	FS	DG	JM	SZ	ZS	ZQ	HK	ZH	Avg
Feb	Local	1.7	11.3	12.0	12.3	2.9	19.8	7.3	2.3	25.8	3.4	9.9
		(53.9%)	(64.4%)	(52.3%)	(57.9%)	(28.1%)	(68.6%)	(30.1%)	(29.7%)	(84.4%)	(20.1%)	(53.8%)
	Regional	0.3	5.4	9.9	8.2	6.	8.1	16.4	4.2	3.6	13.0	7.6
		(8.5%)	(30.9%)	(43.0%)	(38.8%)	(65.8%)	(28.0%)	(67.5%)	(55.5%)	(11.7%)	(76.4%)	(41.3%)
	S-Regional	1.2	0.8	1.1	0.7	0.6	1.0	0.6	1.1	1.2	0.6	0.9
		(37.5%)	(4.7%)	(4.7%)	(3.3%)	(6.1%)	(3.4%)	(2.4%)	(14.7%)	(3.9%)	(3.5%)	(4.9%)
May	Local	1.6	8.9	8.4	10.2	2.3	16.7	5.3	1.8	14.1	4.3	7.4
		(42.5%)	(62.4%)	(59.1%)	(58.5%)	(46.3%)	(73.0%)	(38.7%)	(41.6%)	(77.5%)	(49.5%)	(60.1%)
	Regional	1.4	5.0	5.3	6.9	2.4	5.5	8.2	2.0	3.1	4.2	4.4
		(36.1%)	(34.7%)	(37.4%)	(39.4%)	(48.8%)	(24.0%)	(59.7%)	(45.8%)	(17.0%)	(48.0%)	(35.8%)
	S-Regional	0.8	0.4	0.5	0.4	0.2	0.7	0.2	0.6	1.0	0.2	0.5
		(21.4%)	(2.9%)	(3.5%)	(2.1%)	(4.9%)	(3.0%)	(1.6%)	(12.6%)	(5.4%)	(2.5%)	(4.1%)
Aug	Local	1.6	8.5	7.0	9.7	2.4	14.6	4.5	1.7	14.4	3.9	6.8
		(35.7%)	(59.0%)	(61.0%)	(55.0%)	(60.1%)	(68.0%)	(45.1%)	(41.7%)	(78.5%)	(61.2%)	(60.8%)
	Regional	2.2	5.5	4.2	7.6	1.3	6.3	5.3	1.9	3.2	2.3	4.0
		(49.2%)	(38.7%)	(36.2%)	(43.2%)	(32.8%)	(29.3%)	(52.9%)	(47.4%)	(17.3%)	(25.5%)	(35.5%)
	S-Regional	0.7	0.3	0.3	0.3	0.3	0.6	0.2	0.4	0.8	0.2	0.4
		(15.1%)	(2.3%)	(2.7%)	(1.8%)	(7.1%)	(2.7%)	(2.0%)	(10.9%)	(4.2%)	(3.3%)	(3.7%)
Nov	Local	1.4	7.7	9.5	8.7	2.5	14.7	5.8	2.0	11.9	3.8	6.8
		(26.6%)	(58.6%)	(44.4%)	(58.7%)	(17.7%)	(68.4%)	(24.4%)	(27.0%)	(61.9%)	(21.1%)	(42.9%)
	Regional	0.2	2.5	8.8	3.4	9.2	3.4	15.6	2.2	2.9	11.4	6.0
		(2.9%)	(19.4%)	(41.0%)	(22.9%)	(65.9%)	(15.8%)	(65.1%)	(30.2%)	(15.3%)	(64.0%)	(37.7%)
	S-Regional	3.6	2.9	3.1	2.8	2.3	3.4	2.5	3.2	4.4	2.7	3.1
		(70.5%)	(22.0%)	(14.5%)	(18.5%)	(16.4%)	(15.8%)	(10.5%)	(42.8%)	(22.7%)	(15.0%)	(19.4%)

Table 5: Regional contribution of NO_x from surrounding cities to five cities in the PRD region

		HZ	GZ	FS	DG	JM	SZ	ZS	ZQ	HK	ZH	PRDW
Feb	FS	0.6%	63.6%	0.0%	10.1%	4.6%	5.4%	6.5%	2.2%	3.4%	2.7%	1.0%
	DG	13.9%	20.4%	0.1%	0.0%	0.0%	53.4%	0.3%	0.0%	9.5%	0.1%	2.3%
	SZ	19.2%	1.8%	0.0%	38.7%	0.0%	0.0%	0.0%	0.0%	36.5%	0.1%	3.7%
	ZS	1.4%	35.0%	12.5%	21.6%	0.3%	11.6%	0.0%	0.0%	8.3%	6.5%	3.0%
	ZH	1.6%	22.1%	6.1%	20.2%	2.7%	15.1%	15.0%	0.0%	11.6%	0.0%	5.5%
May	FS	0.7%	56.9%	0.0%	9.3%	11.2%	2.2%	7.8%	3.7%	2.1%	5.3%	0.8%
	DG	8.8%	16.0%	0.2%	0.0%	0.3%	49.5%	0.9%	0.0%	17.6%	1.6%	5.2%
	SZ	8.2%	2.4%	0.1%	19.9%	0.1%	0.0%	0.2%	0.0%	59.4%	0.4%	9.2%
	ZS	0.9%	28.3%	10.3%	21.5%	2.9%	7.8%	0.0%	0.0%	6.3%	19.0%	3.0%
	ZH	1.1%	17.5%	5.0%	21.8%	6.7%	14.5%	15.7%	0.0%	10.9%	0.0%	6.9%
Aug	FS	0.2%	30.1%	0.0%	8.6%	23.7%	4.6%	8.3%	6.4%	3.2%	13.9%	1.1%
	DG	4.1%	18.4%	1.1%	0.0%	0.6%	45.8%	2.0%	0.0%	19.4%	2.2%	6.4%
	SZ	3.1%	4.6%	0.9%	15.9%	0.3%	0.0%	0.4%	0.0%	63.3%	0.9%	10.7%
	ZS	0.3%	19.0%	11.5%	13.2%	6.2%	8.5%	0.0%	0.3%	10.1%	26.7%	4.3%
	ZH	0.4%	11.5%	6.9%	11.1%	17.9%	9.4%	15.7%	0.7%	15.8%	0.0%	10.5%
Nov	FS	2.1%	78.4%	0.0%	12.3%	0.7%	1.0%	1.8%	3.0%	0.4%	0.3%	0.1%
	DG	35.3%	31.3%	0.7%	0.0%	0.1%	27.0%	0.1%	0.1%	4.5%	0.2%	0.7%
	SZ	34.2%	1.0%	0.3%	46.2%	0.1%	0.0%	0.0%	0.1%	16.8%	0.1%	1.4%
	ZS	2.5%	28.1%	9.3%	35.9%	0.1%	13.9%	0.0%	0.0%	4.4%	4.1%	1.6%
	ZH	2.4%	11.9%	2.5%	19.7%	2.0%	28.8%	12.7%	0.0%	16.6%	0.0%	3.5%

*Values in bold represent contributions of over 10%.

Table 6: Estimates of premature mortality due to NO_x from each source category in 10 PRD

Respiratory Mortality	HZ	GZ	FS	DG	JM	SZ	ZS	ZQ	HK	ZH
HDDV	7(0,15)	160(0,332)	51(0,106)	47(0,97)	17(0,35)	117(0,238)	19(0,39)	3(0,6)	254(0,519)	9(0,19)
HDGV	1(0,2)	56(0,117)	10(0,21)	6(0,14)	3(0,6)	13(0,28)	3(0,6)	1(0,2)	3(0,6)	1(0,3)
LDDT	1(0,2)	11(0,24)	6(0,12)	4(0,9)	2(0,4)	9(0,20)	2(0,5)	0(0,1)	80(0,168)	1(0,2)
LDGV	2(0,5)	56(0,117)	17(0,37)	18(0,37)	5(0,11)	32(0,67)	6(0,13)	1(0,2)	22(0,46)	3(0,6)
Power Plant	3(0,6)	58(0,123)	30(0,63)	24(0,51)	21(0,44)	14(0,30)	20(0,41)	2(0,5)	25(0,53)	8(0,17)
Industry	2(0,4)	66(0,138)	46(0,96)	11(0,23)	20(0,43)	4(0,8)	11(0,24)	7(0,14)	4(0,9)	4(0,8)
Marine	0(3,6)	9(0,18)	2(0,5)	6(0,13)	2(0,4)	17(0,36)	3(0,7)	0(0,1)	154(0,319)	6(0,12)
Biogenic	1(0,2)	1(0,3)	1(0,2)	1(0,2)	1(0,2)	0(0,0)	0(0,0)	0(0,0)	1(0,2)	0(0,0)
A-M	9(0,18)	102(0,213)	30(0,63)	21(0,45)	13(0,27)	42(0,87)	10(0,22)	5(0,10)	231(0,470)	12(0,24)
Lung Cancer	HZ	GZ	FS	DG	JM	SZ	ZS	ZQ	HK	ZH
HDDV	3(0,8)	75(0,171)	24(0,55)	22(0,51)	8(0,18)	55(0,123)	9(0,20)	1(0,3)	119(0,268)	4(0,10)
HDGV	1(0,2)	26(0,61)	5(0,11)	3(0,7)	1(0,3)	6(0,15)	1(0,3)	0(0,1)	1(0,3)	1(0,2)
LDDT	1(0,2)	5(0,13)	3(0,7)	2(0,5)	1(0,2)	4(0,10)	1(0,2)	0(0,1)	37(0,87)	0(0,1)
LDGV	1(0,2)	26(0,61)	8(0,19)	8(0,19)	2(0,5)	15(0,35)	3(0,7)	0(0,1)	10(0,24)	1(0,3)
Power Plant	1(0,3)	27(0,64)	14(0,33)	11(0,27)	10(0,23)	7(0,16)	9(0,22)	3(1,2)	12(0,28)	4(0,9)
Industry	1(0,2)	31(0,72)	21(0,50)	5(0,12)	10(0,22)	2(0,4)	5(0,12)	3(0,7)	2(0,4)	2(0,4)
Marine	1(0,3)	4(0,10)	1(0,3)	3(0,7)	1(0,2)	8(0,19)	2(0,4)	0(0,1)	72(0,165)	3(0,6)
Biogenic	0(0,1)	1(0,2)	0(0,1)	0(0,1)	0(0,1)	0(0,0)	0(0,0)	0(0,0)	0(0,1)	0(0,0)
A-M	4(0,10)	48(0,110)	14(0,33)	10(0,23)	6(0,14)	20(0,45)	5(0,11)	2(0,5)	108(0,212)	5(0,13)

Figure 1: Column sum of NO_x emissions (moles/second) for domain 3.

Figure 2: The 27km (D1), 9km (D2) and 3km (D3) domain setting for WRF and CMAQ.

Figure 3: Locations of meteorological observation stations and air quality stations. 'HZ' represents Huizhou, 'GZ' represents Guangzhou, 'FS' represents Foshan, 'DG' represents Dongguan, 'JM' represents Jiangmen, 'SZ' represents Shenzhen, 'ZS' represents Zhongshan, 'ZQ' represents Zhaoqing, 'HK' represents Hong Kong, 'ZH' represents Zhuhai. Red crosses indicate air quality stations with O_3 , NO_x and NO_2 data; blue circles indicate air quality stations with only NO_2 data; green triangles represent meteorological stations.

Figure 4: The scatter plot of model simulation (y-axis) and observation data (x-axis) for NO_x at Kwun Tong (unit:ppb). The 1:1 line, 2:1-1:2 line and 5:1-1:5 line are in black, red and green.

Figure 5: Monthly average surface wind speeds for February (a), May (b), August (c) and November (d) 2011 in the PRD region (unit: m/s).

Figure 6: Local source category contributions of NO_x at each city in February, May, August and November 2011

Figure 7: Regional category contribution for five cities in the PRD region. The x-axis is the top-3 regional contribution to the city in each month. Blue, red and green color bars represent the top three contributing from neighboring cities for the specific source category. Purple represents the contribution from the other seven cities for that specific category.