**Estimation of Long-Term Population Exposure to PM_{2.5} for Dense Urban Areas using
1-km MODIS Data**

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Abstract

The lack of long-term $PM_{2.5}$ measurements in developing countries makes it difficult to quantify the overall $PM_{2.5}$ pollution exposures and health impacts in these countries where the $PM_{2.5}$ concentrations are often very high. Moreover, it is also difficult for traditional fixed-site monitoring to capture the substantial spatial variability of $PM_{2.5}$ over dense urban areas or regions with significant topography. Hence, recent developments in satellite-based remote-sensing allowing the reconstruction of long-term, wide-area and high-resolution estimates of current and historical $PM_{2.5}$ concentration is an important step forward, allowing the quantification of the long-term pollution exposure of $PM_{2.5}$ in developing cities and in dense urban areas using the satellite-derived $PM_{2.5}$ data. In this study, instead of just looking at the spatial average $PM_{2.5}$ concentrations, we have studied the long-term population exposure of $PM_{2.5}$ by analyzing the population-weighted $PM_{2.5}$ concentrations at regional, city and district scales by combining $1\text{km}\times 1\text{km}$ satellite-derived $PM_{2.5}$ and population density data sets. The variation of population exposure to $PM_{2.5}$ across the urban areas in the Pearl River Delta (PRD) region from 2000 to 2014 was studied. Our result shows that the $PM_{2.5}$ concentrations over the PRD increased steadily from 2000 to 2004, remained at quite a high level through 2008 and then started to decline after 2009. More importantly, our analysis also shows that, at regional, city and district levels, the population-weighted mean $PM_{2.5}$ concentrations from data with 1 km resolutions are typically the highest, followed by the population-weighted mean $PM_{2.5}$ concentrations from data with 10 km resolutions and then the simple spatial $PM_{2.5}$ averages. This suggests that the use of simple spatial concentrations can lead to systematic underestimation of the overall population exposure and the associated health impacts. This systematic difference is related to the positive correlation between $PM_{2.5}$

pollutant concentration and population density, and shows the usefulness of using high-resolution satellite-retrieved $PM_{2.5}$ concentrations to quantify the overall population exposure. The higher population-weighted mean $PM_{2.5}$ concentration in comparison with simple spatial average indicates that, for more effective reduction of overall population exposure and protection of public health, control efforts must be further targeted at high-population high-pollution areas, and land-use and city planning should also encourage population to redistribute away from the highly polluted areas.

Keywords: MODIS, $PM_{2.5}$, Long-term Exposure, Regional Scale, City Scale, Districts Scale.

1. Introduction

Epidemiological studies in the United States (U.S.) and Europe have shown that exposure to ambient PM_{2.5} (fine particulate matter with aerodynamic diameter smaller than 2.5 µm) has significant adverse effects on morbidity and mortality (Dockery et al., 1993; Pope et al., 2002; Brunekreef and Forsberg, 2005; Beelen et al., 2008; Sacks et al., 2011; Hoek et al., 2013; Beelen et al., 2014). A systematic review of the association between long-term exposure to ambient pollution and chronic diseases conducted by Chen et al. (2008) concluded that long-term exposure to PM_{2.5} increases the cardiovascular mortality risk by 12% to 14% per 10 µg/m³ increase in PM_{2.5} concentration, independent of age, sex and geographic region.

On the other hand, even though many Asian cities have PM_{2.5} concentrations much higher than cities in U.S. or Europe, there are much less studies directly linking PM_{2.5} and long-term health effects in Asia, mainly because of the lack of PM_{2.5} measurements (Wong et al., 2015). Instead, most of the Asian studies (Cao et al., 2011; Zhang et al., 2011; Dong et al., 2012; Zhou et al., 2014) were based on PM₁₀ (particulate matter with aerodynamic diameter smaller than 10 µm) or Total Suspended Particulates (TSP, particulate matter with aerodynamic diameter up to 50 µm). For example, for cardiovascular mortality, Cao et al. (2011) found 0.9% increase for every 10 µg/m³ increase in TSP and Zhou et al. (2014) reported 1.8% increase for every 10 µg/m³ increase in PM₁₀. These cardiovascular mortality risks are much smaller than those noted earlier for U.S. and Europe for PM_{2.5}. Hence, the lack of in-situ measurements makes it difficult for us to independently quantify and understand the chronic health impacts of PM_{2.5} in rapidly developing countries where the concentrations and health effects are the

greatest (Cohen et al., 2005; van Donkelaar et al., 2010). To better quantify the long-term mortality or other health risks of PM_{2.5} in Asia and other developing regions, it is important to be able to get good long-term estimates of PM_{2.5} concentrations for health impact studies.

At the same time, traditional studies mostly relied on fixed-site monitors to quantify the pollutant exposure of the population and to assess the effectiveness of the governments' control policies (Hystad et al., 2011; Wang et al., 2014; Zhang et al., 2013; Zhong et al., 2013a). However, such monitors cannot capture the large spatial variability of the pollutant concentrations (Chen et al., 2006). Even with the enhanced monitoring of PM_{2.5} like what China had started in 2013 (with more than 1000 stations reporting by 2015), it is still difficult to have enough stations to fully capture the large spatial variability in pollutant concentrations. The situation is particularly bad for highly developed urban areas or regions with significant topography where the population is often clustered in small but highly concentrated areas. The use of average pollutant concentrations derived from scattered station measurements can lead to systematic errors in the estimate of overall exposure and hence in the health impact for the population.

Using satellite-derived surface extinction coefficients as proxy for PM_{2.5}, Wong et al. (2015) estimated the mortality hazard ratio for cardiovascular deaths per 10 µg/m³ increase in PM_{2.5} as 1.22. This is similar to a 22% increase, which is higher than the 12% to 14% noted by Chen et al. (2008) in their review. An important difference between Wong et al. (2015) and the earlier studies reviewed in Chen et al. (2008) was the use of high-resolution (1km×1km) satellite data to estimate the PM_{2.5} exposure for their cohort while the earlier studies used more sparsely located station data in the PM_{2.5} exposure estimation. In this study, we shall use

similar method to examine the differences in exposure estimates for cities with and without high-resolution PM_{2.5} pollution data.

Compared with fixed-site measurements, recent development in satellite-based remote sensing provides a useful alternative for estimate the long-term exposure to outdoor air pollution on large spatial scales (Brauer et al., 2012; van Donkelaar et al., 2010). Satellite-derived estimates of PM_{2.5} from Aerosol Optical Depth (AOD) are increasingly being incorporated into exposure studies and more broadly into health impact assessments (Anderson et al., Phase Three study groups, 2012; Crouse et al., 2012; Lim et al., 2012). Combining satellite-based AOD from MODIS (Moderate Resolution Imaging Spectroradiometer) and MISR (Multiangle Imaging Spectroradiometer) and coincident aerosol vertical profile from the global chemical transport model, van Donkelaar et al. (2010) and Brauer et al. (2012) estimated global PM_{2.5} concentration in the early to mid-2000s. By combining the satellite-retrieved PM_{2.5} concentration and population density, these studies show that in the early 2000s, about 25% to 32% of the global population lived in areas with annual PM_{2.5} concentration exceeding the World Health Organization (WHO) Air Quality PM_{2.5} Interim Target-1 (IT-1, annual mean of 35 µg/m³), and the situation is particularly severe in eastern and southern Asia. By combining MODIS AOD and PM_{2.5} concentration derived from newly available national PM_{2.5} network over China, Ma et al. (2014) showed that more than 96% of the Chinese population lives in areas that exceed the WHO IT-1 in 2013.

While most of the aforementioned assessments focused on global and national scales using satellite observations with spatial resolutions of 0.1°×0.1° or about 10km×10km (Brook

et al., 2013; Chen et al., 2013; Crouse et al., 2012; van Donkelaar et al., 2013; Evans et al., 2013; Hystad et al., 2011), there are new algorithms that can provide AOD (Li et al. 2004, 2005a; North et al., 2008; Lyapustin et al., 2011) and more recently PM_{2.5} data (Beloconi et al., 2016; Hu et al., 2014; Lin et al. 2015) at higher spatial resolution of 0.01°×0.01° (or 1km×1km). These higher resolution products provide new opportunities for looking at the PM_{2.5} problem at urban city and district scales where the spatial variation of PM_{2.5} is large, and can hence help better quantify the PM_{2.5} exposure in dense developing urban cities.

The Pearl River Delta (PRD) region is situated in the southern coast of China. It includes Hong Kong, Macau and 9 cities in China's Guangdong province. These Chinese cities were the first to develop when China started its modernization in the 1980s. With an area of about 8000 square kilometers and about 59 million people, the PRD region has been identified by the World Bank as the largest urban area in the world (World Bank, 2015). Unfortunately, with rapid development, the PRD region is also one of the most polluted city clusters in China (Zheng et al., 2015).

The Chinese national PM_{2.5} data was too short for long-term exposure analyses. However, as a result of joint collaboration efforts between Hong Kong and Guangdong, hourly PM_{2.5} ground measurements have been made in the PRD at a few stations since 2005 (Zhong et al. 2013a). These long-term PM_{2.5} station-based datasets across the PRD region provide a unique opportunity for us to study the impact of high-resolution PM_{2.5} data for exposure estimates for large urban populations, as well as for further validation of the accuracy of the high-resolution 0.01°×0.01° satellite-retrieved PM_{2.5} concentrations.

2. Data and Methods

2.1 Data

The PRD region, shown in Figure S1 of the Supporting Information, is located on the southern coast of China. With 59 million people in about 8000 square kilometers, it has one of the highest population densities in China. The PRD region is a large plane surrounded by mountains on three sides (west, north and east) and the Pacific Ocean to the south. The basin-shaped PRD confines pollutant dispersion when the background wind is weak. The climate of the PRD is affected by seasonal monsoons. The prevailing winds are from the north in winter and from the south in summer. The Chinese cities in the PRD were the first region in China to experiment free-market economic reform and thus had the longest and largest overall economic growth. Unfortunately, the rapid urbanization and industrialization in the PRD have caused heavy air pollution, which has been the main environmental concern of the public in the region, not only within Guangdong but also in the nearby Hong Kong. These concerns prompted the governments of Hong Kong and Guangdong to setup the PRD Regional Air Quality Monitoring Network (PRD-RAQMN) in 2005 to start monitor the air quality across the region (Zhong et al. 2013b). More importantly for the current study, even though $PM_{2.5}$ was not a criteria pollutant for Hong Kong or Guangdong at that time, it was also monitored at some of the stations of the PRD-RAQMN.

Primary data used in this study include (i) spectral data from the two MODIS instruments, Terra (since 2000) and Aqua (since 2002); (ii) station measurements of $PM_{2.5}$ over Hong

Kong (2000 to 2014) and the PRD (2005 to 2014); (iii) 3-hourly surface meteorological parameters, including visibility (L) and relative humidity (RH), from the global telecommunications system (GTS) of the World Meteorological Organization (WMO) from 2000 to 2014; and (iv) 1km $\times$ 1km LandScan population data developed by Oak Ridge National Laboratory (Bright et al., 2011).

Two MODIS instruments, Terra and Aqua, pass the equator at about 10:30am and 1:30pm local time, respectively. They provide daily AOD that covers nearly the entire globe. Terra provided AOD data from February 2000, and Aqua provided AOD data from July 2002. In this study, the 1-km AOD data in the PRD (111.5°E to 115°E, 21.5°N to 24°N) from 2000 to 2014 were used; these data were retrieved by the dark-target land algorithm from MODIS at 0.55 μ m and from our own look-up table, which was more compatible with the local conditions (Kaufman et al., 2002; Li et al., 2004, 2005a). A MODIS cloud mask with 99% cloud-free criteria is used to exclude cloudy pixels. Retrieval errors within 15% to 20% of sunphotometer measurements which has the same accuracy as MODIS standard aerosol products are found in Beijing and Hong Kong (Li et al., 2005a, 2005b). The retrieval AOD and PM₁₀ concentrations over Hong Kong and the PRD from 2001 to 2013 was validated with observations from the Aerosol Robotic NETwork (AERONET) and the governments' air quality monitoring stations, respectively (Li et al., 2015). Using similar AOD retrieval algorithm, Lin et al. (2015) also estimated the high-resolution (1km $\times$ 1km) PM_{2.5} distribution over China for 2013, and verified them with the newly released PM_{2.5} data across the country.

For this study, we have also obtained hourly PM_{2.5} data from 2000 to 2014 at air quality monitoring stations operated by the Hong Kong Environmental Protection Department and

data from 2005 to 2014 at a few stations of the PRD-RAQMN. As $PM_{2.5}$ was not a required criteria pollutant for monitoring before 2013, the temporal ranges of the $PM_{2.5}$ data were different at different stations. Since 2013, a much denser $PM_{2.5}$ network was established in the PRD by the Ministry of Environmental Protection of China, and by the end of 2014, there are nearly 100 hourly $PM_{2.5}$ monitoring sites in the PRD, as shown by the green points in Figure S1 in the Supporting Information.

The $PM_{2.5}$ concentration is monitored using the tapered element oscillating microbalance (TEOM) technique or with beta attenuation monitors (BAM or β -gauges). The principle disadvantage of the TEOM is the potential for volatilization of collected components such as ammonium nitrate and secondary organic compounds (Schwab et al., 2004); the BAM may also bias the measurements when large amounts of volatile components are present (Grover et al., 2006; Chung et al., 2001). However, the measurements uncertainties of the $PM_{2.5}$ data are generally within 10% (HKEPD, 2013). Because both methods are commonly used for routine monitoring purposes, they are used for validation of satellite-retrieved $PM_{2.5}$ concentrations in this study. The $PM_{2.5}$ data at 11am and 2pm were used to validate the satellite-derived hourly $PM_{2.5}$ data. The long-term averaged satellite-retrieved $PM_{2.5}$ data were adjusted with ratio of the ground-measured daily-average (based on 24 hours data) $PM_{2.5}$ concentrations to the average $PM_{2.5}$ concentrations at satellite passage times (11am and 2pm), and then evaluated by these ground-measured daily-average $PM_{2.5}$ concentrations.

Different from satellite-retrieval algorithms that are dependent on global numerical models, our retrieval algorithm uses surface weather observations to correct for the vertical distributions of aerosol and humidity. Hence, surface meteorological parameters, including

visibility and relative humidity, were obtained from weather reports available from the WMO GTS network, as shown by purple squares in Figure S1 in the Supporting Information. The meteorological data at 11am and 2pm were extracted to match the passage times of Terra and Aqua, respectively.

2.2 Satellite-Derived PM_{2.5}

The AOD represents a columnar characteristic of aerosols quantified by light extinction, whereas PM_{2.5} is a surface parameter to measure the gravimetric properties of particles. A description of the retrieval of ground-level PM_{2.5} concentration from the MODIS AOD was given by Lin et al. (2015) and will therefore only be briefly summarized here. Instead of using numerical models to simulate meteorological parameters, our algorithm is observational data-driven based on surface meteorological data (including visibility and relative humidity data) from WMO GTS stations. Furthermore, the AOD-PM_{2.5} model was built up based on physical understanding rather than pure regression techniques. There are generally two steps. In the first step, the concept of scale height H is incorporated into the vertical correction, which transforms the AOD to a surface aerosol extinction coefficient ($\sigma_a = \text{AOD}/H$). The visibility data provide accurate surface extinction coefficient information at specific GTS locations. Combined with satellite AOD, the information on the aerosol vertical distribution (H) at each GTS site could be derived. Assuming smooth variation in the aerosol vertical distribution (H) on a regional scale, the regional surface aerosol extinction coefficient could be derived from the satellite AOD. In the second step, the effects of hygroscopic growth, mass

extinction efficiency (MEE) and fine mode fraction (FMF) are taken into account in the humidity correction, which transforms the surface aerosol extinction coefficient into the PM_{2.5} concentration. The integrated humidity effects of hygroscopic growth, MEE and FMF are indicated by two parameters: the integrated humidity factor γ' and the integrated reference value K . Here, γ' shows the integrated humidity dependence from hygroscopic growth, MEE and FMF; K is an integrated reference value under the dry condition with RH of 40%. It equals to ratio of reference MEE and reference FMF of mixed aerosols under dry condition. Both values are associated with aerosol properties and could be estimated by simultaneous ground measurements of visibility, relative humidity and PM_{2.5} concentrations at the same locations. Based on the two location-dependent parameters γ' and K , the spatial distribution of that surface PM_{2.5} concentration can then be derived from the aerosol surface extinction coefficient σ_a , which is calculated in the first step:

$$PM_{2.5} = \frac{\frac{AOD}{H}}{K \cdot \left(\frac{1-RH}{1-RH_0}\right)^{-\gamma'}} \quad (1)$$

In this study, the γ' and K values are estimated with the use of simultaneous ground measurements of visibility, relative humidity and PM_{2.5} concentrations at three GTS stations in this region (Lin et al., 2015). Mean γ' and K value is 0.68 ± 0.03 and $6.80 \pm 0.13 \text{ m}^2/\text{g}$, respectively, at the three GTS stations in the PRD region. All input data (e.g., AOD, H , RH, γ' and K) are mapped onto grids with spatial resolutions of $0.01^\circ \times 0.01^\circ$. Ground-measured PM_{2.5} concentrations at all of the air quality monitoring stations were then utilized to evaluate the satellite-retrieved PM_{2.5} concentrations.

3. Results

3.1 Evaluation of PM_{2.5} Concentration

To match ground-measured PM_{2.5} concentrations, satellite-retrieved PM_{2.5} concentrations are extracted at the 1km×1km grids where the monitoring stations are located. Figure S2 in the Supporting Information shows the scatter plot comparisons; good correlation coefficient of 0.68 (N = 49265) is found for the hourly mean PM_{2.5} concentrations from ground and corresponding satellite-derived measurements for all PM_{2.5} observations from 2000 to 2014.

As each satellite passes over once a day for any specific times, to estimate long-term (e.g., monthly or annual) averages, the satellite-derived PM_{2.5} values must further be corrected for the difference between the 24-h daily average and the average of the hourly observations at the satellite overpass times (11am and 2pm). Hence, correction factors were derived at each monitoring station from the ratios of monthly means of the 24-h ground-based PM_{2.5} concentrations to monthly means of ground-based PM_{2.5} concentrations at the satellite overpass times; these monthly correction factors were then be interpolated onto all the grid points in our analyses domain using spline interpolation method. Spatial distributions of the monthly correction factors are shown in Figure S3 in the Supporting Information and the long-term satellite-retrieved PM_{2.5} concentrations have all been corrected using these correction factors. The correction factors smaller than 1 (larger than 1) indicates a possible overestimate (underestimate) for the daily mean PM_{2.5} level without this correction. The correction factors reflect the diurnal pattern of PM_{2.5} pollution, which may be related to both

local emission profiles and variation of meteorological conditions (Peng et al., 2011; Zhang et al., 2015). As shown in Figure S3, the correction factors are generally < 1 at most of the PRD urban area in spring and summer; while the correction factors are generally > 1 in autumn and winter especially over the northern parts of PRD.

Figure 1 shows the time-series comparisons of the monthly averaged $\text{PM}_{2.5}$ concentration from ground and satellite measurements at two stations: (a) Tsuen Wan, and (b) Yuen Long. The significant cyclic variations are associated with the seasonal evolution of the East Asian Monsoon system (Yuan et al., 2013). In summer, a large surface low-pressure system develops over Asia, leading to southerly or southwesterly prevailing winds that bring a clean oceanic air mass from the sea. In winter, the inland temperature becomes lower than that of the ocean, leading to northerly or northeasterly surface winds in the PRD that transport particles to Hong Kong from the highly polluted PRD center area. Significant correlations of 0.83 ($N = 120$, $\text{RMSE} = 7.70 \mu\text{g}/\text{m}^3$) and 0.91 ($N = 119$; $\text{RMSE} = 6.58 \mu\text{g}/\text{m}^3$), respectively, were found at the two stations. The deviations between the satellite-based mean $\text{PM}_{2.5}$ concentration and the ground-based mean $\text{PM}_{2.5}$ concentration were $-2.50 \mu\text{g}/\text{m}^3$ and $0.94 \mu\text{g}/\text{m}^3$ for Tsuen Wan and Yuen Long, respectively, which shows good agreement between the satellite and ground measurements.

Figure 1. Comparison of monthly PM_{2.5} concentrations from ground measurements and satellite measurements at (a) Tsuen Wan and (b) Yuen Long from 2005 to 2014.

Figure S4 in the Supporting Information shows the evaluation of the monthly mean PM_{2.5} concentration from the satellites at all of the ground-based air-quality stations from 2000 to 2014. A good correlation coefficient of 0.82 (N = 3094) is found, which is comparable to 0.85 (N = 20) for the monthly mean of the satellite-retrieved PM_{2.5} concentration in Northern Italy (Di Nicolantonio et al., 2009). Figure S5 in the supporting Information shows the evaluation of the annual mean PM_{2.5} concentration from the satellites at all of the ground-based air-quality stations from 2000 to 2014. A good correlation coefficient of 0.77 (N = 231) is obtained, which is comparable to those of 0.77 to 0.82 in U.S. and 0.74 to 0.85 in China (van Donkelaar et al., 2010, 2013; Geng et al., 2015).

Since there are large seasonal variations in both the satellite-derived and the station-based PM_{2.5} concentrations, we also want to make sure the significant correlation between the two time-series is not just because of their being phase-locked with the seasonal cycle. Hence, we have also verified that the correlations between the satellite-derived and station-based PM_{2.5}

time-series are still significant after removing both the seasonal and annual variations. For example, student t-test results in Table S1 of the Supplemental Information show that the correlation between the detrended monthly mean satellite-derived and station-based PM_{2.5} concentration time-series (with both seasonal and annual cycle removed) for the Tsuen Wan air quality monitoring station is 0.69, which is still statistically significant at the 99% confidence level.

Nevertheless, the correct seasonal trend of the satellite-derived PM_{2.5} concentrations does reflect the capability of the retrieved product in reproducing the seasonal variation of the station-based PM_{2.5} concentrations (which is not the case for satellite-based AOD). Hence, we shall keep using the full, not the seasonal and annual detrended, time-series in the calculation of subsequent verification statistics.

Table 1. The validation statistics for the monthly and annual means for satellite-retrieved PM_{2.5} using all available ground observations.

	RMSE ($\mu\text{g}/\text{m}^3$)	ME ($\mu\text{g}/\text{m}^3$)	MAE ($\mu\text{g}/\text{m}^3$)	MPE (%)	MAPE (%)	R
2000-2014 (monthly)	11.2	-3.5 ± 10.6	8.3 ± 7.5	-2.0 ± 30.7	22.3 ± 21.2	0.82 (N=3094)
2000-2012 (monthly)	11.0	-4.3 ± 10.1	8.2 ± 7.3	-4.9 ± 30.3	22.4 ± 21.0	0.76 (N=1164)
2013-2014 (monthly)	11.3	-3.0 ± 10.9	8.4 ± 7.6	-0.3 ± 30.7	22.2 ± 21.3	0.84 (N=1930)
2000-2014 (annual)	6.2	-1.5 ± 6.0	5.0 ± 3.6	-3.1 ± 14.7	12.6 ± 8.2	0.77 (N=231)
2013 (annual)	5.5	-0.7 ± 5.4	4.7 ± 2.9	-0.5 ± 12.0	10.6 ± 5.7	0.78 (N=76)
2014 (annual)	6.1	0.3 ± 6.1	4.8 ± 3.8	1.1 ± 15.1	12.3 ± 8.9	0.74 (N=86)

For a more comprehensive evaluation, statistics for verification of the monthly and the annual means including the root mean square error (RMSE), mean error (ME), mean absolute error (MAE), mean percentage error (MPE), mean absolute percentage error (MAPE) and correlation coefficient (R) for satellite-retrieved PM_{2.5} using all the available ground observations are listed in Table 1. For the monthly means, since there are many more stations after 2013, we have listed in the verification statistics for all stations in the first row, as well as separate comparisons at available stations in 2000-2012 and 2013-2014 in rows 2 and 3, respectively. Nevertheless, the verification statistics for all stations in all years and those for the separate data sets before and after 2013 are very similar, suggesting that the retrieval accuracy is quite stable for the monthly means. The deviations between the satellite-retrieved and ground-based monthly mean PM_{2.5} concentrations are well within about $\pm 40\%$. For the annual means, the verification statistics for all available stations were listed in row 4, and those for 2013 and 2014 were shown separately in rows 5 and 6, respectively. Again, the verification statistics in rows 4, 5 and 6 are quite similar, suggesting that the retrieval accuracy is also stable for annual means. The deviations between the satellite-retrieved and ground-based annual mean PM_{2.5} concentrations are well within about $\pm 20\%$.

3.2 Yearly Variation of PM_{2.5} Concentration

Figure 2. Yearly variation in the annual average of satellite-retrieved and ground-observed concentrations of $PM_{2.5}$ in the PRD. The peak $PM_{2.5}$ concentrations were observed in 2004 and 2007-2008, which is consistent with other studies based on ground measurements.

Figure 2 shows the year-to-year variation of the satellite-retrieved annual $PM_{2.5}$ concentrations at 1-km resolution from 2000 to 2014, and the station $PM_{2.5}$ concentrations in the PRD are shown by circles. Higher concentrations of $PM_{2.5}$ appeared near the center of the

PRD and lower values in the surrounding areas. Meanwhile, the satellite measurements of the peak $PM_{2.5}$ concentrations were observed in 2004 and 2007-2008 at the center of the PRD. This trend is consistent with studies based on emissions and ground measurements of PM. For example, Lu et al. (2013) showed that PM_{10} emissions increased continuously by 76% from 2000 to 2007 but slightly decreased from 2007 to 2009. With the use of pollution measurements from the PRD-RAQMN network, Zhong et al. (2013) concluded that PM_{10} concentrations declined by 13.5% when the annual concentrations were compared within a 6-year period from 2006 to 2011 and peaked in 2007. Wang et al. (2014) indicated that the annual average $PM_{2.5}$ concentration ranged from $49.1 \mu g/m^3$ in 2000 to $64.3 \mu g/m^3$ in 2010, with a peak of $84.1 \mu g/m^3$ in 2004 in the PRD based on published ground $PM_{2.5}$ data from various sources including published papers, statistical yearbooks and the Internet. Zhang et al. (2013) found a peak PM_{10} concentration in 2003-2004 and a decline of approximately 30% in the annual PM_{10} concentration from 2004 to 2009 in Guangzhou on the basis of data from nine ground sites measured by the Guangzhou Environmental Monitoring Center. Hence, the satellite-derived $PM_{2.5}$ values show reasonable year-to-year variations compared with other sources.

3.3 Long-Term $PM_{2.5}$ Exposure Assessment in the PRD and HK

3.3.1 PRD

Figure 3(a) shows the long-term average satellite-retrieved $PM_{2.5}$ concentrations from

2000 to 2014 in the PRD with a spatial resolution of $0.01^{\circ} \times 0.01^{\circ}$. Substantial variations in the $PM_{2.5}$ concentrations can be noted. Much higher $PM_{2.5}$ concentrations are observed in the center of the PRD, with orange and red colors showing annual average $PM_{2.5}$ concentrations higher than $35 \mu g/m^3$ (WHO Interim Target-1 for $PM_{2.5}$; please refer to Table S2 for details of WHO Air Quality Guidelines, Interim Targets and Chinese National Ambient Air Quality Standards for $PM_{2.5}$)

The long-term averaged $PM_{2.5}$ concentration in Hong Kong for the same period is plotted on a subgraph with a different color scale to highlight the spatial variability across Hong Kong. This plot shows that higher $PM_{2.5}$ concentrations in the central urban areas (with very tall buildings and heavy traffic) and in the northwest, which is adjacent to the PRD and most susceptible to pollutant transport from the PRD (Yuan et al. 2006).

To quantify public exposure, we need to combine the average pollutant concentration data with population data with similar spatial resolution. Here, the LandScan $1km \times 1km$ population data developed by Oak Ridge National Laboratory is used (Bright et al., 2011). Figure 3(b) shows the distribution of population density in the PRD and Hong Kong with the same spatial resolution as the $PM_{2.5}$ concentration. The total population of the PRD and Hong Kong was estimated at about 59 million, of whom about 7.3 million reside in Hong Kong. Most areas in the region have population densities greater than 1000 people/km^2 , which is much higher than the urban criteria set by some other countries. The United States Census Bureau defines an urban area as having a population density of at least 1,000 people per square mile (386 people/km^2). Statistics Canada defines an urban area as having a population density of no less than 400 people/km^2 .

Figure 3. (a) Average satellite-retrieved $PM_{2.5}$ concentrations from 2000 to 2014 in the PRD and in Hong Kong (inset). (b) The distribution of population density at same spatial resolution in the PRD and in Hong Kong.

Comparison between the spatial distributions of $PM_{2.5}$ concentration and the population density in Figure 3 show that areas with higher population densities are often associated with higher $PM_{2.5}$ concentrations. In other words, as more people are living in highly polluted areas, the percentage of the population living in less polluted “rural” areas is smaller. Hence, the spatial average of the $PM_{2.5}$ concentration would underestimate the mean pollution exposure, and the population-weighted average would be a better indicator of the $PM_{2.5}$ pollution exposure to the public.

For the PRD, the spatial average of PM_{2.5} concentration from 2000 to 2014 is $38.0 \pm 7.6 \mu\text{g}/\text{m}^3$, while the population-weighted mean PM_{2.5} concentration for the same period is much higher at $43.9 \pm 7.8 \mu\text{g}/\text{m}^3$. To evaluate the effect of spatial resolution, the high-resolution PM_{2.5} concentration and population density data are averaged into grids with lower resolutions of $0.1^\circ \times 0.1^\circ$ and the results are shown in Figure S6 of the Supporting Information; the corresponding population-weighted mean PM_{2.5} concentration at this lower resolution is estimated to be $42.9 \pm 8.1 \mu\text{g}/\text{m}^3$. It is higher than the simple spatial average PM_{2.5} concentration, but lower than the estimate with datasets at higher spatial resolutions of $0.01^\circ \times 0.01^\circ$. Hence, satellite-retrieved PM_{2.5} concentration with higher spatial resolution can give more accurate estimate of the pollution exposure to the public.

The PRD consists of Hong Kong, Macau and 9 Chinese cities in Guangdong. There is no verification site in Macau so it is not considered in this study. To show the resolution effect for different cities, the three estimates of PM_{2.5} exposures for the entire PRD and individual cities are plotted in Figure 4 (and the corresponding averages are listed in Table S3 of the Supplemental Information). In particular, the blue bars in Figure 4 show the spatial average PM_{2.5} levels in PRD and the individual cities, the green bars show the population-weighted mean PM_{2.5} with data at 10 km resolution, and the red bars show the population-weighted mean PM_{2.5} with data at 1 km resolution. The cities are ordered by their population-weighted mean PM_{2.5} concentrations from data at 1 km resolution. In term of population-weighted mean PM_{2.5} concentration, the most polluted city in the PRD is Foshan (FS) and the least polluted city is Hong Kong (HK). Furthermore, Figure 4 shows that for nearly all of the cities, the red bars are higher than the green bars, and the green bars are higher than the blue bars.

This is because the pollution concentrations and the population densities are positively correlated. Ideally, it would be better to have more people reside in less polluted areas, but that is certainly not the case in most of the cities in the PRD. The only city with the spatial mean equal to the population-weighted means is Zhuhai (ZH), which is indicative of better city planning in Zhuhai (in term of air pollution exposure) when compared to other cities in the PRD. In contrast, the city with largest difference between the spatial mean and the population-weighted mean is Guangzhou (GZ), suggesting that the city is least successful in cleaning up the densely populated districts or in encouraging its residents to live in less polluted districts.

Figure 4 also shows that the WHO Air Quality $PM_{2.5}$ IT-1 is exceeded in all PRD cities except for HK, and the WHO IT-2 is exceeded in all PRD cities including HK. These results highlight the severity of air pollution and $PM_{2.5}$ exposure problems in the region, especially at its center city clusters.

Figure 4. Population-weighted mean PM_{2.5} concentrations from data at 1 km resolutions (red bars), population-weighted mean PM_{2.5} concentrations from data at 10 km resolutions (green bars), and spatial mean PM_{2.5} concentrations (blue bars) in different cities in the PRD from 2000 to 2014.

With the use of the 1-km satellite-retrieved PM_{2.5} data, cumulative percentage of the population (0%-100%) at different exposure levels of PM_{2.5} concentrations in the PRD and its component cities from 2000 to 2014 are shown in Figure 5, with details in Table S3 of the Supplemental Information. The results show that the WHO AQG, IT-3 and IT-2 for PM_{2.5} are exceeded for 100% (59 million) of population in the PRD, while the WHO IT-1 is exceeded by 84.0% (49.5 million) of the population. Based on satellite PM_{2.5} observation, van Donkelaar et al. (2010) estimated that 50% of the population in eastern Asia was exposed to PM_{2.5} concentrations exceeded WHO Air Quality PM_{2.5} IT-1. Result in this study confirms that the PRD is one of the most polluted region in the eastern Asia.

As shown in Figure 5, a larger proportion of the population is exposed to high PM_{2.5} concentrations in Foshan (FS), Dongguan (DG), Guangzhou (GZ), and Zhongshan (ZS), which is consistent with the previous discussion that central PRD is more polluted. More than 90% of populations are exposed to PM_{2.5} levels that exceed WHO IT-1 in all cities except Jiangmen (JM, 79.8%), Huizhou (HZ, 56.1%) and Hong Kong (HK, 26.4%). While highlighting the severity of the pollution problem in the PRD, these results also showed the ability of the high-resolution satellite-derived data in the quantification of the pollution exposure problem down to the city level in densely populated regions like the PRD.

Figure 5. Cumulative percentage of the population at different $PM_{2.5}$ exposure levels in the PRD (black line) and its component cities (dashed lines) from 2000 to 2014. WHO IT-1 is exceeded by 84.0% and 26.4% of the populations in the PRD and HK, respectively.

3.3.2 HK

With the high spatial resolution of the $PM_{2.5}$ concentration data, long-term levels of $PM_{2.5}$ exposure could also be analyzed at district scale within a city. Taking advantage of the new remote-sensing data at 1-km resolution, we have analyzed the $PM_{2.5}$ exposure in Hong Kong at district level. Hong Kong has 18 districts; the population-weighted average concentration of $PM_{2.5}$ (red) and the spatial average concentration of $PM_{2.5}$ (blue) for the 18 districts from 2000 to 2014 are shown in Figure 6 and the details are listed in Table S4 in the Supplemental information.

First, we note that the population-weighted mean concentrations (red bars) for any district are typically higher than the simple spatial average (blue bars) for the same district, showing positive correlation between population density and pollutant concentration in most of the districts. Second, while there are clear differences in the PM_{2.5} concentrations in the districts, all districts in Hong Kong have annual concentrations higher than the WHO IT-2 (25 µg/m³), which is a long way from the WHO AQG (10 µg/m³), highlighting the severe PM_{2.5} pollution problem in Hong Kong (even though it is already the least polluted city in the PRD). Third, the population-weighted mean PM_{2.5} concentrations at three of the districts (Kowloon City, Yuen Long, and Yau Tsim Mong) are higher than the WHO IT-1 (35 µg/m³), suggesting that these districts are of particular concern in term of population exposure to PM_{2.5}.

Figure 6 also shows that the differences between the population-weighted average (red bars) and the spatial average (blue bars) concentrations can be quite different for different districts. In particular, the two concentrations are almost the same at some districts (e.g., Yau Tsim Mong and Islands districts), but the population-weighted average can also be higher than the spatial average by almost 4 µg/m³ (e.g., North and Tai Po districts). This shows that there are significant differences in spatial correlations between population density and PM_{2.5} concentration in the different districts, with districts showing higher population-weighted averages having higher positive spatial correlation between population density and pollution distribution. In other words, these districts are having more people living in relatively more polluted areas (either because of the lack of effective land-use planning or simply the high density is too crowded for effective management of pollution).

This result highlights the fact that the overall population exposure can be lowered either

by reducing the mean pollutant concentration or by decreasing the correlation between population density and pollutant concentration. The first is well-known and is often the target of local pollution control programs. The second is less discussed and it can be effected by either identifying pollution hot-spots in high population area and have targeted control programs to reduce the pollutant level at these hot-spots (which should be more cost effective than trying to reduce pollutant concentrations everywhere), or encouraging the public to relocate to less polluted areas through city planning and land-use development.

Figure 6. Population-weighted mean PM_{2.5} concentration and spatial mean PM_{2.5} concentration in different districts in Hong Kong from 2000 to 2014.

4. Discussion

An extensive body of epidemiological studies has shown that exposure to ambient PM_{2.5} can be linked to significant and adverse health effects. To better quantify the adverse health impacts of PM_{2.5}, exposure estimates that can better incorporate the spatial variability of PM_{2.5} concentrations are needed. Traditional epidemiological studies relied upon fixed-site government monitors to estimate exposure levels. However, it is difficult and too costly for such monitor networks to fully capture the spatial variability of PM_{2.5}, particularly over dense urban areas or region with significant topography. At the same time, the lack of extended PM_{2.5} measurements in developing countries makes it difficult to study and quantify the long-term health impacts of PM_{2.5} in these countries where the pollutant concentrations are often much higher and the potential health effects are much greater than those in developed cities in the U.S. or Europe. Hence, recent developments in satellite-based remote sensing allowing the reconstruction of long-term and wide-area estimates of historical PM_{2.5} concentration is an important step forward, allowing the quantification of the long-term health impacts of PM_{2.5} in developing countries and dense urban areas using satellite-derived PM_{2.5} data.

Up to the present, most satellite-based exposure studies relied on satellite data with spatial resolution of 0.1°×0.1° (10km×10km). Products at these resolutions are good for looking at larger regional scale variabilities, but they are less useful for characterizing the spatial variations of PM_{2.5} within dense urban city areas like the PRD. This motivated us to try to apply the higher resolution satellite-derived PM_{2.5} data with spatial resolution (0.01°×0.01°) to quantify the population exposure of PM_{2.5} at regional, city and even district scales in the PRD.

Evaluations of satellite-retrieved $PM_{2.5}$ concentrations against ground-based station data show that the percentage errors between the annual mean $PM_{2.5}$ from satellite and ground observation are within $\pm 20\%$. The deviations are likely to result from uncertainties of satellite-based AOD, γ' and K values, and input visibility and RH data. In this study, constant γ' and $\alpha_{ext,10}$ were applied throughout the entire study period. Because these two values are associated with aerosol properties, we expect that estimations of $PM_{2.5}$ concentration can be further improved if we take into account the temporal variations of γ' and K . However, this requires more $PM_{2.5}$ observations but most $PM_{2.5}$ observations are available only after 2013 and too short for temporal analyses of γ' and $\alpha_{ext,10}$. Therefore, we recommend further studies to be conducted to examine effects of temporal variations in γ and $\alpha_{ext,10}$ in a few years' time after more data become available. Further, verification statistics are found to be comparable for the two periods from 2013 to 2014 (with a lot of verifying stations) and from 2000 to 2012 (with much less verifying stations), suggesting that the retrieval method is stable when applied to the past when there was only a small number of verifying stations.

As noted in Section 3.1, the satellite-derived $PM_{2.5}$ values must also be corrected for the systematic difference between the 24-hour daily average and the average of the hourly observations at the satellite overpass times (11am and 2pm). This systematic bias can either be positive or negative. As shown in Figure S7, the negative bias (e.g. Hong Kong) indicates a possible overestimate of $PM_{2.5}$ concentrations without this correction, while the positive bias (e.g. Guangzhou) indicates a possible underestimate of $PM_{2.5}$ concentration without this correction. We found that the difference between the corrected and un-corrected estimates can be as large as about 10% (see Figure S7). The results shows that 10% potential bias could be

introduced in long-term exposure estimates of $\text{PM}_{2.5}$ without consideration of the different sampling time of the satellite and the ground-based measurements.

Because of the effects of cloud and bright surface, some satellite-based AOD data are missing. Satellite data set in 2014 is used as an example for examination of this effect. Figure S8(a) in the Supporting Information shows the spatial distribution of sample size of the available satellite observations in 2014 in the region. Sample sizes are around 300 in most of the PRD region. Larger percentages of the satellite data are missing at the center urban areas of the PRD, which is probably due to stronger effects of cloud or bright surface. To evaluate the effect of missing data, Figure S8(b) shows the spatial distribution of the relative deviation between annual mean of the ground-observed $\text{PM}_{2.5}$ concentration at the times for which satellite observations are available (sample sizes are around 300) and at all of the 2 hours (11am and 2pm) for which satellites pass over the region (sample size = 365 x2). Mean relative deviation of $2.5 \pm 5.8\%$ at all of the ground stations indicates that the effect of the missing satellite data on long-term $\text{PM}_{2.5}$ estimates is not large in the region.

5. Conclusions

In this study, $\text{PM}_{2.5}$ concentration with a spatial resolution of 1 km was retrieved in the PRD region from 2000 to 2014. The deviations between satellite-retrieved and ground-based monthly (annual) mean $\text{PM}_{2.5}$ concentrations are well within about $\pm 40\%$ ($\pm 20\%$). Instead of focusing on the average $\text{PM}_{2.5}$ concentration, this study focuses on the long-term population exposure of $\text{PM}_{2.5}$ at regional, city and district scales by analyzing the population-weighted

mean PM_{2.5} concentrations calculated using 1km×1km satellite-derived PM_{2.5} and population density data. Overall, the long-term population-weighted mean PM_{2.5} concentration from 2000 to 2014 at the PRD is $43.9 \pm 7.8 \mu\text{g}/\text{m}^3$, and the cities with the highest ($53.3 \pm 4.2 \mu\text{g}/\text{m}^3$) and lowest ($32.5 \pm 3.0 \mu\text{g}/\text{m}^3$) population-weighted mean PM_{2.5} concentrations are Foshan and Hong Kong, respectively.

Using the newly available satellite-derived dataset, the variations of the PM_{2.5} concentrations from 2000 to 2014 was assessed. Our result shows that the PM_{2.5} concentrations over the PRD increased steadily from 2000 to 2004, remained at quite a high level through 2008, and then started to decline after 2009. The initial increase in pollutant concentrations up till 2008 is in line with government reports showing failure to control emissions of SO₂, smoke and dust during the 10th FYP (China MEP, 2008). The reduction in pollution in recent years since 2009 may be associated with the more effective air pollution control policies applied in the area in recent years (Lu et al., 2013). However, the pollution problem is severe in the region, with the WHO AQG, IT-3 and IT-2 for PM_{2.5} being exceeded for nearly all (59 million), and the WHO IT-1 being exceeded for 84.0% (49.5 million) of the population in the PRD. For the individual cities, more than 90% of the population is exposed to PM_{2.5} concentrations exceeding WHO IT-1 in all the cities except for Jiangmen (79.8%), Huizhou (56.1%) and Hong Kong (26.4%).

Our analysis also showed that at regional, city and district levels, the population-weighted mean PM_{2.5} concentrations from data with 1 km resolutions are typically the highest, followed by population-weighted mean PM_{2.5} concentrations from data with 10 km resolutions and then the simple spatial PM_{2.5} averages. This suggests positive correlation between PM_{2.5}

concentration and population density in most cities, and that the use of simple spatial average (or data with lower spatial resolution) can lead to a systematic underestimation of the average exposure. Hence, this underscores the utility of using high-resolution satellite-retrieved PM_{2.5} concentrations to quantify the overall population exposure and health impact estimates. The high-resolution satellite-derived PM_{2.5} data allowed us to compare the population exposure between different districts in the same city (Hong Kong) and also showed that, amongst the different cities in the PRD, Zhuhai (ZH) had been the most successful while Guangzhou (GZ) had been the least successful in encouraging its residents to stay in less polluted areas.

The higher population-weighted mean PM_{2.5} concentration in comparison with simple spatial average indicates that more control efforts must be targeted at the high-population high-pollution areas for the more effective reduction of overall pollution exposure and protection of public health. Beyond the tail-pipe solutions that target individual emission sources, we must also consider land-use planning to put more people in less populated areas, as well as demand-side management measures (e.g., ventilation assessment, electronic road pricing, low-emission zones and pedestrianization) to improve ventilation and reduce the high pollutant concentrations in the most densely populated part of the city.

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