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Evaluating Vehicle Emission Control Policies using on-Road Mobile Measurements and Continuous Wavelet Transform: a Case Study during the Asia-Pacific Economic Cooperation Forum, China 2014

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Abstract. Vehicle emissions are major sources of atmospheric pollutants in urban areas, especially in megacities around the world. Various vehicle emission control policies have been implemented to improve air quality. However, the effectiveness of these policies is unclear, due to a lack of systematic evaluation and sound methodologies. During the Asia-Pacific Economic Cooperation (APEC) Forum, China 2014, the

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Chinese government implemented the strictest vehicle emission control policy in the country's history, which provided an opportunity to evaluate its effectiveness, based on our recently developed method. To evaluate the vehicle emission decline, we used a mobile research platform to measure the main air pollutants ($PM_{2.5}$, black carbon (BC), SO_2, CO, NO_x and O_3) on the 4th ring road of the city of Beijing, combined with a continuous wavelet transform method (CWT) to separate out 'instantaneous emissions' by passing vehicles. The results suggested that our measurements captured the spatial distribution and variation of atmospheric pollutant concentrations on the 4th ring road. The 'instantaneous concentration' decomposed by the CWT method represents on-road emissions better than other methods reported in the literature. With this method, we found that the daytime vehicle emission of CO and NO_x decreased by 28.1 and 16.3 %, respectively, during the APEC period relative to the period before APEC, and by 39.3 and 38.5 %, respectively, relative to the period after APEC. The night-time vehicle emissions of CO and NO_x decreased by 56.0 and 60.7 %, respectively, during the APEC period relative to the period after APEC. Because vehicle emissions of NO_x and CO contribute considerably to the total emissions of these pollutants in Beijing, the vehicle emission control policy implementation was extremely successful in controlling air quality during APEC 2014, China.

Keywords: On-road mobile measurement, Continuous wavelet transform, Asia-Pacific Economic Cooperation Forum, Air quality

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1 Introduction

Particle and trace gases emissions from vehicles contribute significantly to urban air pollution (Amato et al., 2016; Milando et al., 2016). Due to the increase in vehicle numbers and the lack of control policies, vehicle emissions have become an important issue for urban air quality management, especially in megacities (Denier van der Gon et al., 2013; Cyrys et al., 2014; Kelly and Zhu, 2016). The megacity of Beijing has been experiencing severe air pollution problems over the last three decades (Streets et al., 2007; Tang et al., 2009; Han et al., 2013; Wang et al., 2014). Not only have the concentrations of the main pollutants reached extremely high levels during pollution episodes but the frequency of polluted days has also been high (Guo et al., 2014; Tian et al., 2014). Vehicle emissions are a major pollution source in the complex emission budget of Beijing, arising from the rapid increase in the number of vehicles in use, which is now near six million (Beijing Traffic Management Bureau 2015, Kelly and Zhu, 2016). Positive matrix factorization (PMF) results in a volatile organic compound (VOC) source appointment study showed that vehicle exhaust contributed 27.82 ppb (35 %)，8.17 ppb (17 %) and 19.65 ppb (14 %) before (18 - 31 October), during (3 - 12 November) and after APEC, China 2014 (13 - 22 November), respectively (Li et al., 2015). The Fifth-Generation Penn State/NCAR Mesoscale Model - Community Multiscale Air Quality (MM5-CMAQ) simulation suggested that road transport contributed 27.4 ± 4.8 % of $PM_{2.5}$ to Beijing's atmosphere in 2011 (Cheng et al., 2013).

To improve air quality, a vehicle emission control policy was implemented by the

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Beijing government in the early 2000s, and more comprehensive policies have been in place since the 2008 Beijing Olympic Games (Wang et al., 2009; Kelly and Zhu, 2016). These policies have improved the air quality to a certain degree, but they have also inconvenienced the residents of the city (e.g., alternating restrictions on driving based on the last digit of the car license number) and generated enormous economic losses. Due to the enormous negative influence of vehicle emissions on daily life and economic development, the effectiveness of vehicle emission control policies has attracted much attention. Because of the inconsistent results from different station measurements and model simulations, the efficiency of various vehicle emission control policies is uncertain (Zhang et al., 2009; Wang et al., 2010; Zhou et al., 2010).

Measurements from a curbside station used by Zhou et al. (2010) suggest that vehicle emissions of NO_x and CO decreased by 49.0 and 44.5 %, respectively, during the 2008 Olympics, relative to the pre-control period. However, another study reported that the decreases of vehicle emissions of NO, NO_2, NO_x and CO were about 53, 27, 30 and 23 %, respectively, based on measurements made at a height of 20 m at the China Meteorological Administration station near the 3rd ring road of Beijing (Zhang et al., 2009). The use of different measurement stations is likely to be the reason for the inconsistent results between these two studies. The CMAQ model results indicate that mobile source NO_x and non-methane VOC (NMVOC) concentrations were reduced by 46 and 57 % during the 2008 Olympics, relative to the pre-control period (June 2008) (Wang et al., 2010). Emission inventories have a large impact on model simulation

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results. Overall, the inconsistent results are mainly due to the representation of vehicle emissions by different station measurements and model simulations.

A mobile research platform has many advantages for measuring on-road air pollutants directly and capturing fresh emissions from passing vehicles. Because of these advantages, mobile research platforms have been widely used for mapping the spatial and temporal distributions of air pollutants, estimating the emission factors of on-road air pollutants, identifying and apportioning sources of air pollutants in areas at various geographical scales and evaluating the influence of specific point sources or regional transport on air quality (Bukowiecki et al., 2002; Kolb et al., 2004; Fruin et al., 2004; Zavala et al., 2009; Wang et al., 2009, 2011; Thornhill et al., 2010; Hudda et al., 2013; Sun et al., 2014; Weichenthal et al., 2015). For example, the spatial and temporal distributions of trace gases and aerosol parameters in Switzerland have been characterized (Bukowiecki et al., 2002). On-road emission factors of ammonia in New Jersey and California, USA, have been estimated by mobile research platform measurements (Sun et al., 2014). Additionally, the transport and distribution of SO_2 in and around the city of Beijing have previously been characterized by our group (Wang et al., 2011).

On-road concentrations of pollutants are comprised of instantaneous emissions from passing vehicles, on-road accumulative concentrations and the environmental background concentration. The concentration from instantaneous vehicle emissions is the most appropriate to evaluate the effectiveness of vehicle emission control policies

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because there is less influence from meteorological conditions and other factors. Early studies used several methods to separate out vehicle instantaneous emissions from the on-road concentration, but they have proved to be complicated or it has been difficult to obtain real 'instantaneous emissions' (Bukowiecki et al., 2002; Kolb et al., 2004; Jiang et al., 2005; Thornkill et al., 2010; Riley et al., 2014).

Measurement from a stationary site far from the road has been used to represent the environmental background concentration, and then the difference between the on-road and environmental background concentrations can be regarded as the vehicle emission (Riley et al., 2014). This method can only approximate the environmental background concentration and is limited by the availability of suitable stationary sites. In addition, this method cannot distinguish instantaneous emissions from the on-road accumulative concentration. Bukowiecki et al. (2002) used the fifth-percentile of a five-minute moving average of on-road concentrations as the on-road background concentration, incorporating the on-road accumulative concentration and environmental background concentration. The remaining concentration was then regarded as the concentration of instantaneous emissions. This method is affected by vehicle volume, vehicle speed, road condition and other factors. Jiang et al. (2005) used the fifth-percentile of a three-minute moving average of on-road concentrations as the "baseline", then parked the mobile research platform far away from the road to measure the "plume threshold", which was the difference between the "baseline" and 95 % of the data points. By adding the "plume threshold" to the "baseline", the upper boundary

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of the on-road background concentration was obtained. This modification did not overcome the essential problems in Bukowieck's method, which relies on various parameters, such as vehicle volume, vehicle speed and road condition. Thornkill et al. (2010) applied PMF receptor modelling to on-road measurements to quantify diesel and gasoline-vehicle emissions. However, the vehicle emission concentration consists of both an instantaneous emission component and the on-road accumulative concentration. Subjective judgement of the "gentle" and "peak" components of a raw signal has also been used to represent the on-road background concentration and the instantaneous concentration (Kolb et al., 2004). A clear line between the "gentle" and "peak" components is required, but most of the time it is difficult to distinguish these two components precisely. In this paper, we report the use of a continuous wavelet transform (CWT) method to separate out instantaneous emission concentrations. A CWT has been widely used in signal decomposition (Tian et al., 2014; Kang et al., 2007; Domingues et al., 2005). The signal can be decomposed into low and high frequency components (Torrence and Compo, 1998). In our study, a CWT was used to decompose the signals of on-road measured pollutants. Because instantaneous emissions from vehicles are random and change rapidly, the high frequency component of the CWT decomposition represents the instantaneous emissions, while the low frequency component is the sum of the on-road accumulated and environmental background concentrations.

The 21st Asia-Pacific Economic Cooperation (APEC) Meeting was held in Beijing, China, between 5 and 11 November 2014. To avoid poor air quality during the APEC

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period, the government formulated a series of air quality control policies in Beijing and neighbouring regions (Table 1), which were even stricter than those of the Beijing 2008 Summer Olympics (Wang et al., 2009). These policies were aimed at reducing emissions from vehicles, power plants, construction sites and factories. The vehicle emission control policy removed more than 50 % of vehicles from the roads of Beijing city. This provided a unique opportunity to assess the effectiveness of the vehicle emission control policy. In our study, an on-road mobile research platform was used to measure the spatial distribution of trace gases and particles on the 4th ring road of Beijing before (28 October - 2 November), during (3 - 12 November) and after (13 - 22 November) the APEC meeting. To accurately evaluate the effectiveness of the vehicle emission control policy, we used a continuous wavelet transform method to separate out instantaneous emissions from vehicles. The effectiveness of the vehicle emission control policy was analysed and discussed.

2 Experimental methods

2.1 Mobile research platform and instruments

A mobile research platform was used to characterize the spatial and temporal distribution of pollutants (NO_x, SO_2, CO, black carbon (BC) and $PM_{2.5}$) on the 4th ring road of the city of Beijing in the periods “before APEC”, “APEC” and “after APEC”. The details of our mobile research platform were described in a previous study (Wang et al., 2009). Briefly, the mobile research platform was built in a diesel vehicle (IVECO

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Turin V). Two sets of uninterruptible power systems (UPSs) were installed to ensure the continuous operation of the instruments, and an isokinetic inlet system was designed to enhance the sampling efficiency. The onboard instruments used to measure gaseous and particulate pollutants were purchased commercially: NO-NO_2-NO_x, CO, O_3 and SO_2 (ECOTECH, Australia), particle number concentrations (PNC) (TSI 3091, USA), particle mass concentrations (Grimm Dust monitor 1.108, Germany) and BC (MAAP, THERMO, USA). Meteorological parameters and GPS data, such as temperature, relative humidity, barometric pressure, and latitude and longitude, were also recorded. In addition, an automobile data recorder was used to record the road traffic situation. The gas analysers (NO-NO_2-NO_x, CO, O_3 and SO_2) were calibrated every week. The traveling speed was maintained at 60 ± 5 km h^{-1} during on-road measurements.

2.2 Air quality control policies and measurement strategy

Table 1 summarizes the air quality control policies before, during, and after the 21st APEC meeting in Beijing. The policies implemented were stricter than those implemented for the Beijing 2008 Summer Olympics. The vehicle emission control policy was applied to a broader area and extended to neighbouring regions (Hebei, Tianjin and Shandong). Industrial emission control policies limited the operation of, or halted, more than 10,000 factories, mostly in the North China Plain (Beijing, Inner Mongolia, Shandong, Shanxi, Hebei, Tianjin and Zhejiang).

To characterize on-road air quality and evaluate the effectiveness of air quality control policies, the 4th ring road (65.3 km long) was chosen as the sampling route (Fig.

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1). In Beijing, most air pollution control policies are initially implemented within the 4th ring road. For example, the city has adopted a vehicle emission control policy that alternately limits the number of vehicles on workdays within the 5th ring road (not including the 5th ring road) based on the last digit of the license plate number since the 2008 Beijing Olympics. In addition, just before APEC, the city completed a shift from coal to natural gas as the fuel used for power generation within the 4th ring road. Our measurements were made twice each day, with one daytime and one night-time measurement. Figure 1 shows the measurements route. For each measurement trip, we travelled anticlockwise on the 4th ring road from PKU station at 10:00 AM (local time) for the daytime measurement, and at 1:00 AM for the night-time measurement. We chose 10:00 AM for the daytime measurement to avoid the morning rush hour and the rapid development of the boundary layer. We chose 1:00 AM for the night-time measurement to measure the emissions from trucks, which were allowed into the city between 12:00 AM to 6:00 AM. The daytime measurement trips, usually took around 90 min while the night-time trips lasted for around 70 min.

Figure 1. here

2.3 Continuous wavelet transform

The CWT was developed from the Fourier transform, and is considered to be a "mathematical microscope" that is widely used in image analysis, transient signal analysis and communication systems (Tian et al., 2014; Domingues et al., 2005; Kang et al., 2007). The CWT is defined by the following equation (Domingues et al., 2005):

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$$\mathcal{W}_f^{\psi}(a,b) = \int_{-\infty}^{\infty} f(u)\,\bar{\psi}_{a,b}(\mathrm{u})\mathrm{du} \qquad \mathrm{a} > 0, \tag{1}$$

where

$$\psi_{a,b}(u) = \frac{1}{\sqrt{a}}\psi(\frac{u-b}{a})$$

is the so-called mother-wavelet; the parameter *f* represents the time series of a signal; and *a* and *b* refer to the scale and translation parameters, respectively; and $\bar{\psi}_{a,b}(u)$ and $\psi_{a,b}(u)$ are the conjugate complexes.

The One-Dimensional Stationary Wavelet Analysis Tool (SWT Denoising 1-D) function of MATLAB (R2014a) tool cabinet was chosen to do the CWT decomposition in this study. The sym4 wavelet was chosen as the mother-wavelet, decomposition level 5 was chosen and a threshold was selected to ensure the smoothness of the result. A sensitivity test indicated that the decomposed instantaneous emission was insensitive (less than 6 %) to the selected mother-wavelet and the decomposition level (from 2 to 8). The result of the CWT decomposition included some negative values in the high frequency component, which was due to the mother-wavelet and the noise reconstituted by the CWT vibrating near to zero. To reconcile this problem, the high frequency component was moved up to reference a special REF-line and the low frequency component was moved down in a homologous manner (Fig. S1). This move made both the low and high frequency components more reasonable.

3. Results and discussion

3.1 Meteorological conditions

Figure 2 shows the temporal variation of meteorological parameters, including wind speed, wind direction, temperature and relative humidity, observed during the sampling period from 28 October to 22 November 2014. Three wind rose plots are displayed for the three separate periods, i.e., the period before APEC (28 October – 24 November), APEC period (3 November – 12 November), and the period after APEC (13 November – 22 November). The temporal variation of temperature (T) and relative humidity (RH) during the three periods are shown accordingly. Meteorological parameters were measured at Fengtai station, which is an urban site (Figure 1).

Figure 2. here

As shown in Figure 2, the APEC period and the period before APEC experienced a high frequency of northwest winds, with high wind speeds; the prevalent wind directions in the period after APEC were northeast and southwest. Beijing is surrounded by emission hot spots, with industrial areas to the south, and the North China Plain and the megacity of Tianjin to the southeast. In contrast, broad areas of desert and grassland are located to the north and northwest of Beijing. Therefore, in terms of the air quality in Beijing city, north winds are considered to be favourable for the diffusion of pollutants and south winds are favourable for the transport of pollutants (Zhang et al., 2014). The temperature declined with time, and the daily minimum temperature dropped below 0 °C after 8th November. Low temperatures might lead to coal burning in households in the suburban areas of Beijing, which constitutes a major source of air pollutants in the winter (Lv et al., 2016; Yang et al., 2016). In summary, meteorological conditions have an important influence on the background levels of air pollutants via

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various mechanisms, such as the transport and diffusion of pollutants, and the emergence of new pollutant sources.

3.2 Temporal and spatial variations of on-road pollutants

Figure 3 shows the temporal variation of the daily average concentrations of $PM_{2.5}$, SO_2, NO_x, BC, CO and O_3 for both daytime and night-time measurements on the 4th ring road of Beijing over the whole measurement period. A significant difference between the daytime and night-time average concentrations was observed. The daytime average concentrations (mean ± 1 SD) of $PM_{2.5}$, BC, SO_2, CO, NO_x and O_3 were 28.8 ± 21.1 μg m^{-3}, 1.8 ± 1.0 μg m^{-3}, 8.2 ± 4.6 ppb, 1.8 ± 0.9 ppm, 306.0 ± 104.8 ppb and 5.4 ± 6.0 ppb, respectively, during the period before APEC; 11.8 ± 5.3 μg m^{-3}, 1.2 ± 0.7 μg m^{-3}, 6.9 ± 4.9 ppb, 1.2 ± 0.6 ppm, 215.1 ± 61.1 ppb and 7.8 ± 5.2 ppb, respectively, during the APEC period; and were 40.4 ± 35.5 μg m^{-3}, 6.1 ± 2.9 μg m^{-3}, 18.4 ± 9.5 ppb, 2.6 ± 1.4 ppm, 350.5 ± 82.8 ppb and 3.4 ± 1.5 ppb, respectively, during the period after APEC. In comparison, the night-time average concentrations of $PM_{2.5}$, BC, SO_2, CO, NO_x and O_3 were 22.1 ± 15.5 μg m^{-3}, 3.5 ± 2.7 μg m^{-3}, 5.3 ± 2.7 ppb, 1.5 ± 1.0 ppm, 304.8 ± 113.3 ppb, and 5.5 ± 7.6 ppb, respectively, during the APEC period, and 66.2 ± 54.1 μg m^{-3}, 8.1 ± 3.6 μg m^{-3}, 20.2 ± 8.3 ppb, 3.3 ± 1.9 ppm, 792.2 ± 228.4 ppb, and 3.5 ± 1.1 ppb, respectively, during the period after APEC.

Figure 3. here

The difference in the concentrations measured in the daytime and night-time could not be fully attributed to the shift of mixing layer height. Specific reasons, such as the influence of background and traffic emissions, are discussed below. Overall, these values were in the range of previous on-road observations in the city of Beijing (Wang et al., 2009; Wang et al., 2012). The concentrations of NO_x and BC were measured in the winter of 2010 on the 6th ring road, with peak values reaching 800 ppb and 10 μg m^{-3}, respectively (Wang et al., 2012). In addition, NO_x and CO concentrations were measured before the 2008 Beijing Olympics on the 4th ring road, with peak values of 110.8 ± 30.0 ppb and 2.4 ± 0.7 ppm, respectively (Wang et al., 2009).

Both daytime and night-time measurements show similar temporal trends in this study. Generally, the concentrations of all pollutants except for O_3 declined during the APEC period compared to the periods before and after APEC, with the highest concentration occurring during the period after APEC (Figs. 3 & 4). For example, daytime average concentrations of on-road $PM_{2.5}$, BC, SO_2, CO, NO_x and O_3 during the APEC period decreased by 60, 33, 16, 33, 29 and -46 % relative to the period before APEC, but decreased by 71, 80, 63, 55, 35 and -129 % relative to the period after APEC, respectively. Stationary measurements at a site on the Peking University campus showed similar temporal variations for both the daytime and night-time average concentrations of these air pollutants (Fig. S2). Only the concentration of O_3 increased in the APEC period, which may be because Beijing has a NO_x-controlled regime for O_3 formation. The decline in average NO_x concentration during the APEC period led to an increase in the average O_3 concentration at the same time, while the significant increase in O_3 concentrations on November 6th and 12th were the result of an inflowing air mass

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from northwest China. A simultaneous decline in the concentrations of all other pollutants and the strong northwest wind indicated the inflow of an air mass. However, even with the strict vehicle emission control policy and a significant decline in vehicle volume in Beijing during the APEC period, the daily average NO_x concentration was not always lower than during the periods before and after APEC. The possible reasons for this could be either a shift in background concentrations or favourable diffusion conditions on certain days during the periods before and after APEC. In either situation, the on-road measurement itself was not appropriate for the direct evaluation of the effectiveness of the vehicle emission control policy.

Figure 4. here

The temporal trends in the concentrations of the different species could be attributed to specific reasons other than the vehicle emission control policy. The sharp increases in the concentrations of all pollutants except for O_3 during the period after APEC were mainly due to coal burning in the heating season, especially the unregulated burning in suburban households within Beijing, as the temperature dropped (Fig. 2). Because residential coal burning is the main source of BC in Beijing (Bond et al., 2013), the mass ratio of on-road background BC to CO (NO_x) can be used an indicator of the prevalence of residential coal burning. The ratio of the on-road background concentration of BC to CO (NO_x) (M(BC/CO) or M(BC/NO_x)) increased dramatically in the period after APEC (Fig. 5), which provides further evidence of the contribution of residential coal burning to background concentrations. Unfavourable meteorological

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conditions, such as the prevalence of a southerly wind (Fig. 2), might be another reason for the rapid increase in the concentration of primary pollutants in the period after APEC.

Figure 4 shows the spatial variations of the average concentrations of pollutants along the north, west, south and east sections of the 4th ring road (Fig. 1) in the periods before, during and after APEC. There were large spatial gradients in the concentrations of all pollutants observed in the measurements made in a single trip, which were mainly due to the number of vehicles on the road and wind direction. It is worth noting that no consistent spatial distribution of the average concentration of pollutants was found during the three periods in our measurements (Fig. 4). The average concentration on the whole of the 4th ring road was thus used for further analysis.

Figure 5. here

3.3 Evaluation of the vehicle emission control policy

3.3.1 Results of the CWT decomposition

To verify the effect of the CWT method in the signal decomposition of on-road measurements of air pollutants, a simulation experiment was conducted (Fig. S3). The CWT successfully decomposed the simulated high frequency signal, which had been previously added to the simulated low frequency signal. This gives us confidence that the high frequency instantaneous concentrations could be separated from the on-road concentration. Figure 6 shows the observational data decomposition both with the CWT

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method and the moving five-minute fifth-percentile method; the results of these two methods were similar in terms of magnitude and trend. An excellent correlation was also found for both the on-road background (R^2 = 0.99) and instantaneous concentrations (R^2 = 0.97) decomposed with the two methods, with slopes of 0.97 and 1.1, respectively (Fig. S4). This comparison indicates the agreement of both methods in signal decomposition. However, the on-road background signals decomposed with the CWT were smoother and without saltation, and better reflected the variation of the on-road background signal, as compared to the results decomposed with the moving five-minute fifth-percentile method. Moreover, the CWT decomposed vehicle instantaneous concentrations of NO_x, CO, SO_2 and O_3 were relatively lower than the on-road background concentration (Figs 7 & S5), which is reasonable as stated below. The CWT decomposed instantaneous concentrations of NO_x and CO were strongly correlated (Table S1), because they had a vehicle emission origin.

Figure 6. here

Figures 7 and S5 show the temporal variation of the average instantaneous and on-road background concentrations of CO, NO_x, BC, $PM_{2.5}$, SO_2 and O_3 decomposed with the CWT method in both daytime and night-time measurements for the whole study period. The average instantaneous concentrations of CO, BC, $PM_{2.5}$, SO_2 and O_3 were far less than the on-road background concentrations, whereas the instantaneous concentration of NO_x was comparable to the on-road background concentration. This was actually considered reasonable for several reasons. First, unlike CO and NO_x, direct

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vehicles emission are not a major source for BC, $PM_{2.5}$, SO_2 and O_3 on the expressways in Beijing (Hao et al., 2005; Bond et al., 2013). Therefore, the instantaneous concentrations of these pollutants should be lower than the on-road background concentrations. Second, the accumulative concentration of vehicle direct-emission pollutants should be high on-road and CO is chemically inert; thus, the on-road background concentration of CO naturally accounts for a large proportion of the overall concentration. Third, the NO_x analyser, CO analyser and GRIMM enabled our measurement to capture the full spike of instantaneous emissions of NO_x, CO and $PM_{2.5}$ from passing vehicles. However, for SO_2, BC and O_3 measurements, the full spike was unlikely to be captured if it was shorter than the time resolution of the SO_2 (120 s), BC (60 s) and O_3 analysers (60 s). Therefore, the instantaneous concentrations of BC, SO_2 and O_3 decomposed by the CWT tended to underestimate the instantaneous emission of passing vehicles. Finally, the instantaneous concentration decomposed by the CWT method did not represent the instantaneous emission of vehicles on the 4th ring road. The difference between the instantaneous concentration and instantaneous emission was acceptable for the evaluation of policy effectiveness because only the relative changes of instantaneous emissions were meaningful in the evaluation.

Figure 7. here

3.3.2 Effectiveness of the vehicle emission control policy

Table 2 shows the percentage decline in the instantaneous concentrations of CO, NO_x, BC and $PM_{2.5}$ in the APEC period relative to the periods before and after APEC in both

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the daytime and night-time. For the daytime, the decline in the instantaneous concentration of CO, NO_x, BC and $PM_{2.5}$ during the APEC period were 28.1, 16.3, -3.5 and 23.5 % relative to the period before APEC, and 39.3, 38.5, 44.6 and 24.0 % relative to the period after APEC, respectively. For the night-time, the decline in the instantaneous concentration of CO, NO_x, BC and $PM_{2.5}$ during APEC period were 56, 60.7, 7.2 and 62.1 % relative to the period after APEC, respectively. The percentage decline in the total concentration and on-road background concentration of CO, NO_x, BC and $PM_{2.5}$ were show in Table S2. As stated above, the absolute values of the instantaneous concentrations of $PM_{2.5}$ and BC were small (Fig. 7), thus their decline percentage provided less information about the effectiveness of vehicle emission control policies.

For the daytime, we observed large variation of the instantaneous concentration of NO_x and CO throughout the periods before, during and after APEC. The large decline during APEC could be explained by the stringent vehicle emission control policies that reduced on-road vehicle numbers. In the periods before and after APEC, a 20 % decline in the number of vehicles on the road was expected, based on the policy to control the number of cars driving on road on weekdays using the last digit of the license plate. During the APEC period, the odd/even plate number rule for traffic control was implemented in Beijing, thus, more than 50 % of all potential vehicles were banned from driving on roads in Beijing during the APEC period. As such, a relative decline of 38 % (=(80-50)/80) in the overall vehicle volume on the roads was calculated for the

APEC period, relative to the periods before and after APEC. We noticed the change of the traffic volume based on the average time taken for one full trip measurements on the 4th ring road, it was 72 min during APEC period, shorter than that in the periods before (87 min) and after APEC (92 min). The decline of instantaneous concentration of CO and NO_x during APEC period relative to the period before APEC was lower than the decline relative to the period after APEC. This could be attributed to the additional control policies implemented in the period before APEC, such as diesel vehicles that did not meet the National Vehicle Emission Standard III were strictly banned from driving on road, and vehicles from other provinces were restricted from entering Beijing. Thus, more vehicles (especially high emission diesel vehicle) were under controlled in the period before APEC relative to the period after APEC.

It is worth to note that the decline of instantaneous concentration of NO_x and CO during APEC period relative to the period before APEC were different. This might be due to the different emission factors of NO_x and CO between gasoline and diesel vehicles, while different control policies were aimed at diesel and gasoline vehicles. Lang et al (2012) reported that gasoline vehicle is the dominant vehicle type in Beijing (account for more than 90 %) and the main contributor to vehicular emission of CO (close to 60 %), while diesel vehicle account less than 10 % of the total vehicle population in Beijing, contributed about 70 % to the total vehicular emission of NO_x. The decline of instantaneous concentration of CO (28.1 %) in APEC period relative to the period before APEC was significantly, mainly due to the stringent control of

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gasoline vehicles emission during APEC period. In comparison, the decline of instantaneous concentration of NO_x (16.3 %) in APEC period relative to the period before APEC was less than the decline of CO, this could be due to two reasons: 1) as stated above, additional control policies for diesel vehicles were implemented in the period before APEC; 2) the increase in diesel buses for public transit needs during the APEC period. The small increase of instantaneous concentration of BC also indicated the increase use of diesel vehicles in the APEC period relative to the period before APEC.

The decline of instantaneous concentration of CO and NO_x in the APEC period relative to the period after APEC in daytime were 39.3 % and 38.5 %, respectively. Same as above, the decline of instantaneous concentration of CO was mainly due to control of gasoline vehicles, and it is about the same as the decline of the number of gasoline vehicles allowed driving on road (38 %). The decline of instantaneous concentration of NO_x was mainly due to the decline of diesel vehicle emission, also evidenced by the decline of instantaneous concentration of BC.

For the night-time, the relative decline in instantaneous concentration was 56.0 % for CO and 60.7 % for NO_x in the APEC period relative to the period after APEC, which was significantly higher than the daytime decline (Table 2). The difference between daytime and night-time results could be explained with two reasons. First, the ratio of the number of diesel vehicles to gasoline vehicles was different between the daytime and night-time. The vehicular emission of CO was dominated by gasoline vehicles, and

NO_x by diesel vehicles, the ratio of the instantaneous concentrations of NO_x to CO (M(NO_x/CO)) could be used to indicate the ratio of the number of diesel vehicles to gasoline vehicles. The ratio M(NO_x/CO) was higher in the night-time than daytime (Fig. 5), indicates a higher proportion of diesel vehicles in the night-time. Moreover, even though the vehicle volume in the night-time was much less than in daytime, as evidenced by the video recorder installed in our mobile research platform, we found the instantaneous concentrations of CO, NO_x, BC and $PM_{2.5}$ in the night-time were higher than those in daytime in the APEC period and the period after APEC (Fig. 7). This could be mainly attributed to higher proportion of diesel vehicles driving in the night time, the emission factors of diesel vehicles are 1-2 times higher than those of gasoline vehicles (Zhang et al., 2014; Lang et al., 2012). Secondly, the declines of the emission of gasoline and diesel vehicle during APEC period relative to the period after APEC were different due to the different vehicle emission control policies (Table 1). Diesel vehicles were strictly controlled in the night-time during the APEC period, only a portion of diesel vehicles (e.g. support cargo trucks which meet the National Vehicle Emission Standard III) were permitted to drive on roads, while no such control policies implemented in the period after APEC. Strict control of diesel vehicles (especially those with high emission of air pollutants) during APEC period favoured the decline of diesel vehicle emission. Because of the lack of night-time measurement in the period before APEC, the effect of control policy in night-time in the period before APEC cannot be evaluated.

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4. Conclusion

We used a mobile research platform to measure on-road air pollutant concentrations on the 4th ring road of Beijing before, during and after the 21st APEC from 28 October to 24 November, 2014. A signal decomposition method, namely a continuous wavelet transform, was introduced to separate out instantaneous concentrations to represent the instantaneous emissions from passing vehicles. The relative changes of instantaneous emission were then used to evaluate the effectiveness of the vehicle emission control policy during the APEC period. Our study suggests that the daytime vehicle emission of NO_x and CO declined by around 38 - 39 % in the APEC period, relative to the periods when the vehicle reduction policy was only based on the last digit of the license plate number in the city of Beijing. The observed effectiveness was consistent with what was expected from the details of the vehicle emission control policy, which suggested the representability of our measurements and the rationality of the CWT decomposed instantaneous concentration for the effective assessment of a vehicle emission control policy. Due to the contribution of on-road emissions to the NO_x and CO emission budget in Beijing, it was concluded that the vehicle emission control policy implemented during the 21st APEC period was crucial to the successful control of emissions.

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Data availability. The data of mobile and stationary measurements are available upon requests.

Author contribution. T. Zhu and Z. Tan designed the experiments. T. Zhu secured the research grants. Z. Tan, Y. Zhu, Y. Han, Y. Fang, Y. Wang and P. Liang carried out the experiments. Y. Wang and Z. Tan applied the continuous wavelet transform method in decomposition of on-road measure signals. J. Wang managed the data. Q. Wang compiled emission control policies implement during APEC. L. Meng and Y. Wang provided the data of meteorological parameters. Z. Tan analyzed the data with contributions from all co-authors. Z. Tan prepared the manuscript with helps from T. Zhu, C. Ye and Y. Li.

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Figure Captions:

Fig. 1 The measurement route on the 4th ring road in Beijing (map Version: Google map 2014). The anti-clockwise trip was started from the monitoring station in the campus of Peking University (PKU station). The yellow line shows the main road. The red star shows stationary stations. The black dotted line separates the 4th ring road into the North, the West, the South and the East parts.

Fig. 2 Meteorological parameters (Wind speed, wind direction, temperature, and relative humidity) measured during sampling period at the Fengtai meteorological station. Three wind rose plots were for the period before APEC (from 24th October to 2nd November) (a), APEC period (from 3rd November to 12th November) (b) and the period after APEC (from 12th November to 22nd November) (c). Blue shaded area shows the APEC period.

Fig. 3 Temporal variations of the concentrations of air pollutant ($PM_{2.5}$, SO_2, NO_x, BC, CO and O_3) averaged along the 4th ring road of Beijing. The solid dots and the open dots represent average concentrations of daytime measurement beginning at 10:00 AM and night-time measurement beginning at 1:00 AM, respectively. Those missing dots were caused by instrument failure.

Fig. 4 Box whisker plots of the concentrations of selected traffic-related air pollutants ($PM_{2.5}$, BC, SO_2, CO, O_3, and NO_x) averaged along the four parts (north, west, south and east) of the 4th ring road of Beijing, in the periods before, during, and after APEC. The box part represents the central 50 % of the data, the lower and upper edge represent the 25th and 75th percentile, respectively. The whiskers in the plot represent the error bars.

Fig. 5 The temporal variations of the mass ratios between CO, NO_x and BC during daytime and night-time measurements. The black solid dots represent mass ratios between instantaneous concentrations decomposed by CWT method, the open dots represent mass ratios between on-road background concentrations decomposed by CWT method. The left panel is for daytime and the right panel is for night-time measurements.

Fig. 6 Background signals of on-road measured NOx concentration decomposed by a continuous wavelet transform (CWT) and the moving five-minute fifth-percentile method. The grey line is the original pollutant concentration (Original), the red line is the on-road background concentration decomposed from the original concentration by the CWT (Background_CWT) and the black line is the on-road background concentration decomposed from the original concentration by a moving five-minute fifth-percentile method (Background_5%).

Fig. 7 Temporal variations of the average instantaneous concentrations and on-road background concentrations of CO, NO_x, BC, and $PM_{2.5}$ decomposed from on-road measurements results by CWT for both daytime and night-time measurements. The solid dots and the open dots represent instantaneous concentration and on-road background concentration, respectively. Middle area shows the APEC period.

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Table 1. Air pollution control measures implemented before, during and after the 21th APEC.

Periods	Starting Dates	Control measures	Detail of measures
Before APEC	1 Oct. 2014	Early warning	Enhancing air quality monitoring.
	1 Nov. 2014	Coal-fired control	Reduce coal-fired power plant productions; operate units were required to reduce emission by more than 30 % in Beijing; Postpone centralized heating.
During APEC	3 Nov. 2014	Traffic control	The odd/even plate number rule for traffic control in Beijing, Tianjin, Hebei and Shandong; ”Yellow label vehicles” and 70 % of official vehicle were banned from Beijing’s roads; Trucks were limited to drive inside the 6th Ring Road between 6 AM and 24 PM. Restricted vehicles with license plate of other province entering Beijing.
	3 Nov. 2014	Industrial Emission Control	148 factories were required to be halted or reduce production in Beijing; More than 10,000 factories were required to reduce production or to be halted in Hebei, Tianjin, Shandong, Shanxi and Inner Mongolia.
	3 Nov. 2014	Dust pollution control	Dust emission factories and outdoor constructions were shut down or reduced in activities in Beijing and surrounding region; Increasing road cleaning and water spraying in Beijing.
	6 Nov. 2014	Emergency measures	State-owned enterprises were required to reduce more productions, 40 % of coal-fired boilers were shut down in Beijing; more special pollutant emission factories were required to reduce production around Beijing.
After APEC	13 Nov. 2014		Lifting of regulations adopted from 13 November; continuous controlling 20 % of private cars based on the last digital of plate number.

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Table 2. The percentage declines of instantaneous concentrations of traffic-related pollutants during APEC period relative to the periods before and after APEC.

	Compared period	CO (%)	NO_x (%)	BC (%)	$PM_{2.5}$ (%)
Day	Before APEC	28.1	16.3	-3.5	23.5
	After APEC	39.3	38.5	44.6	24.0
Night	After APEC	56.0	60.7	7.2	62.1
Day-night average	After APEC	50.1	53.6	25.8	49.4

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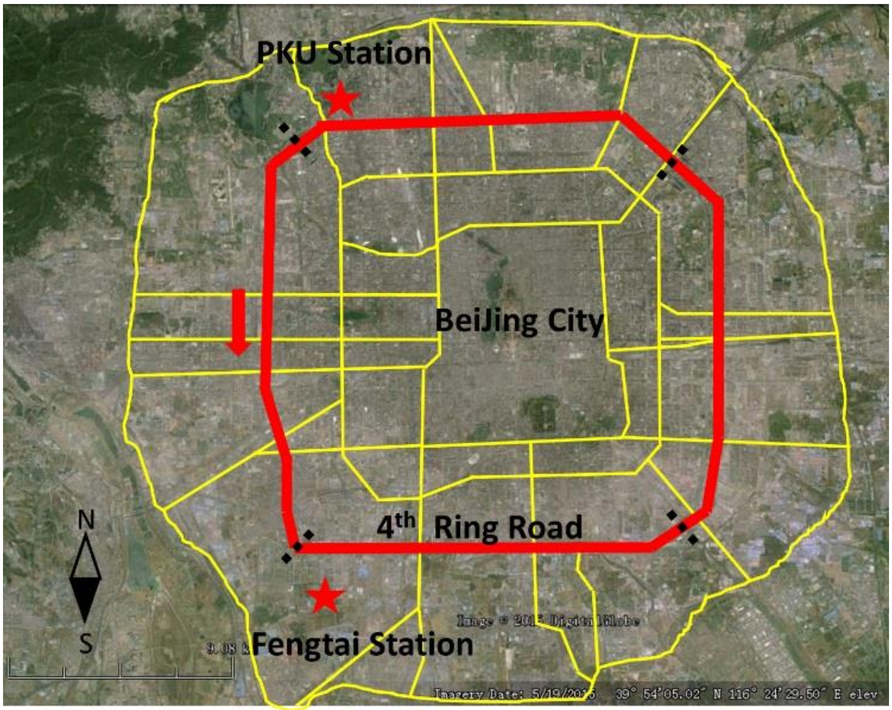

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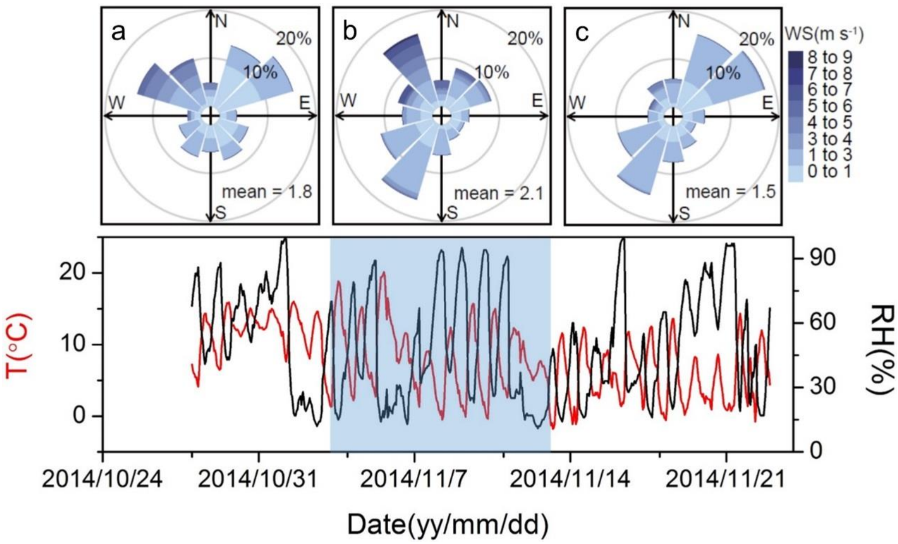

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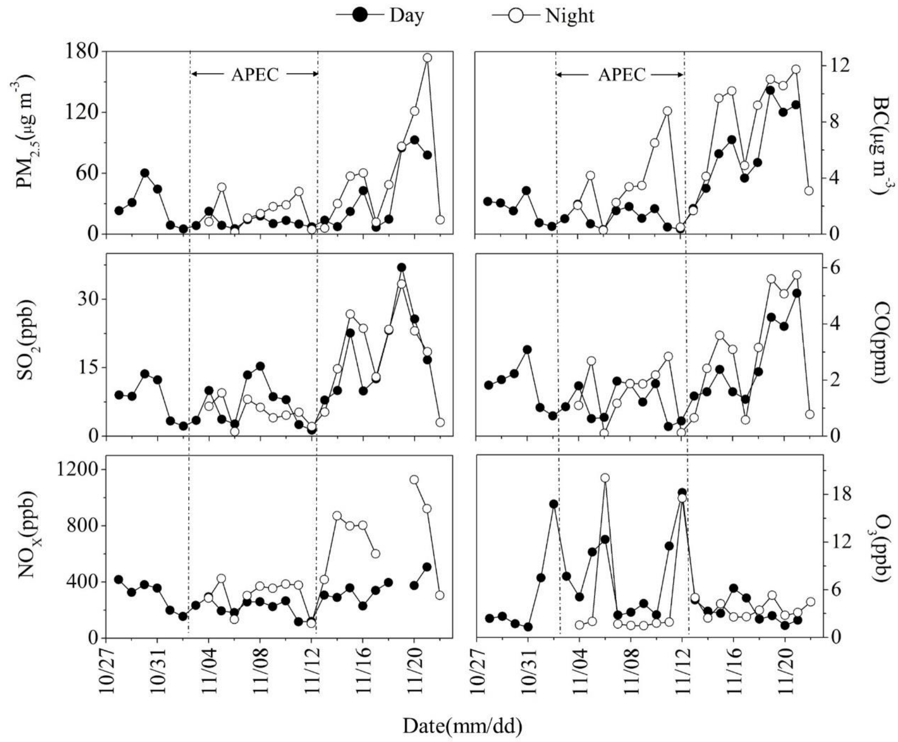

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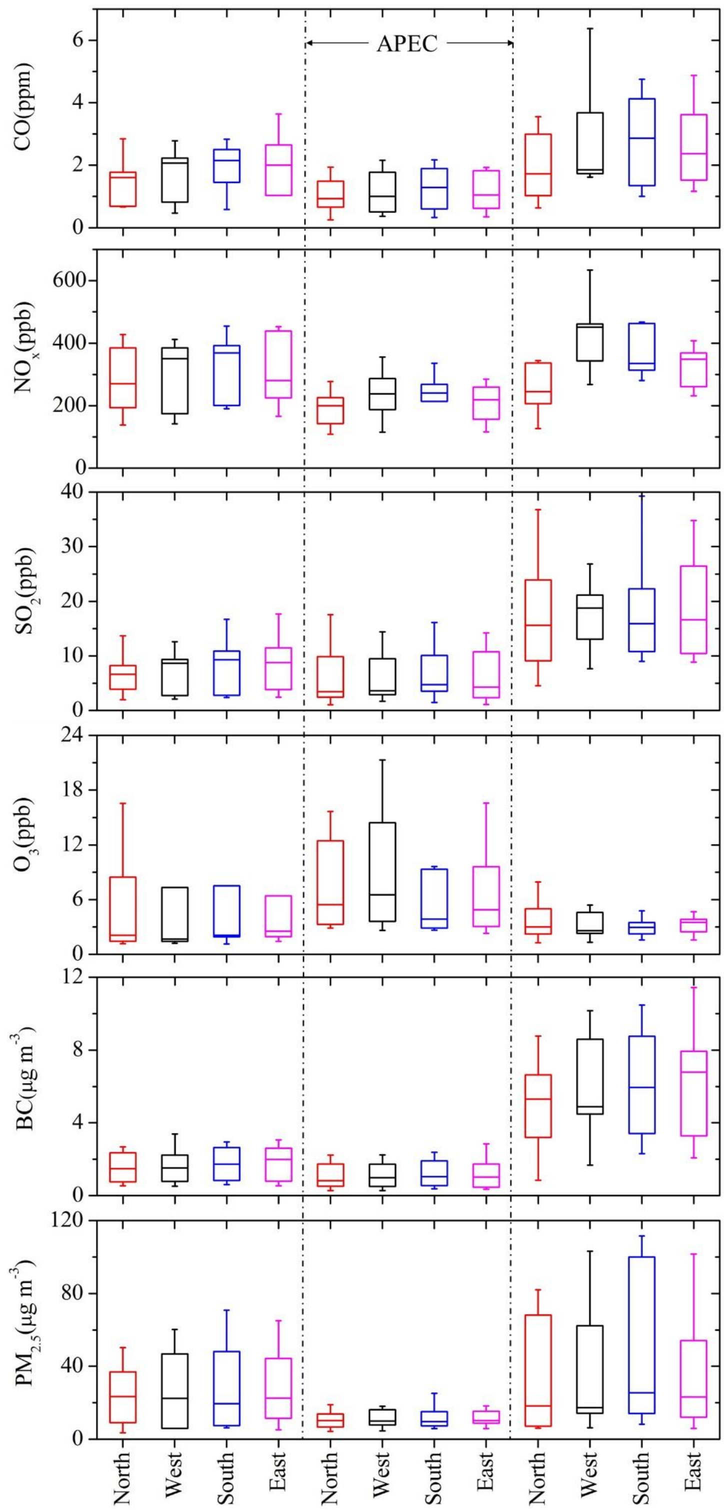

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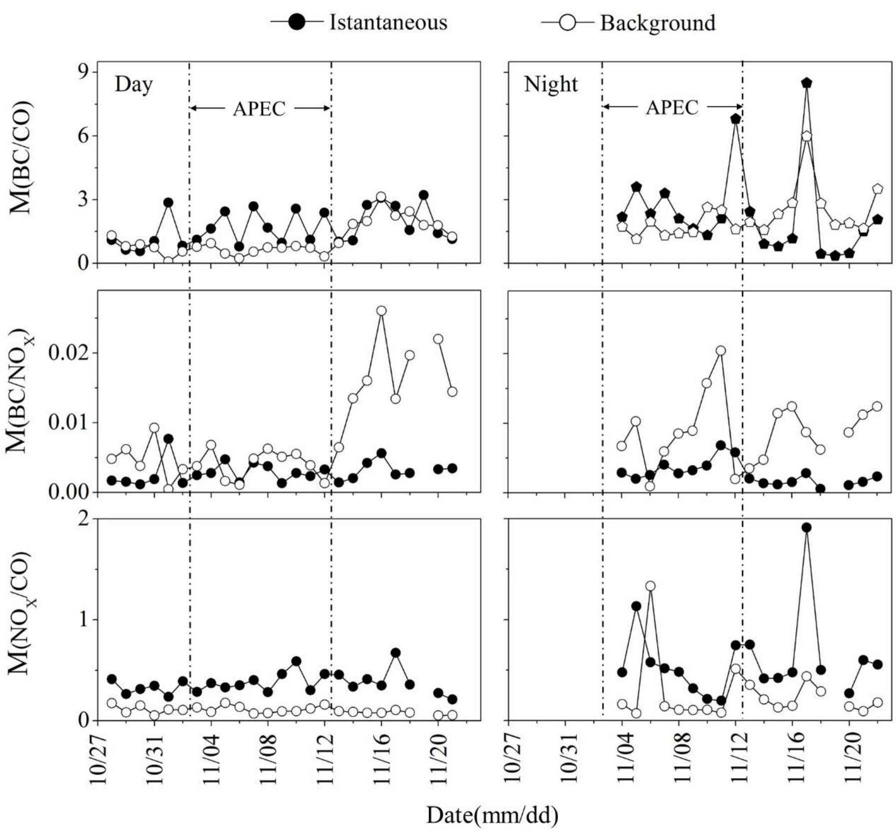

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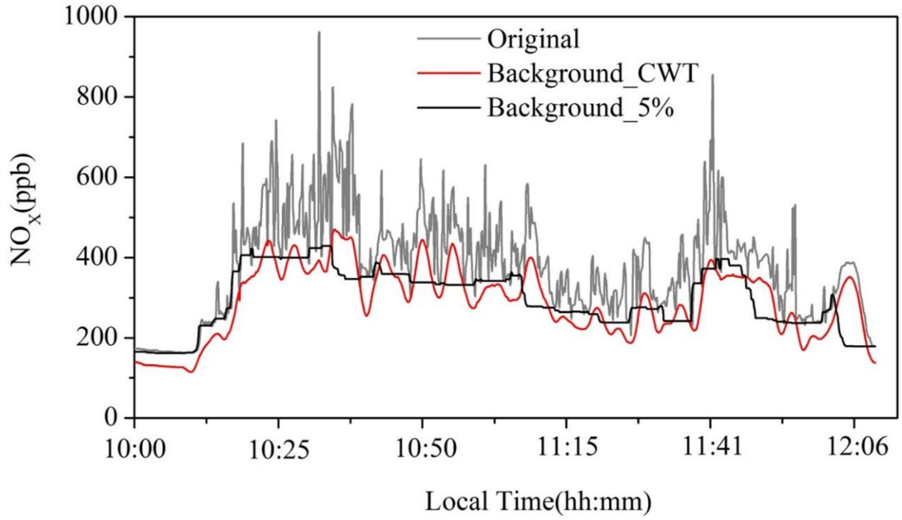

Fig. 6 Background signals of on-road measured NOx concentration decomposed by a continuous wavelet transform (CWT) and the moving five-minute fifth-percentile method. The grey line is the original pollutant concentration (Original), the red line is the on-road background concentration decomposed from the original concentration by the CWT (Background_CWT) and the black line is the on-road background concentration decomposed from the original concentration by a moving five-minute fifth-percentile method (Background_5%).

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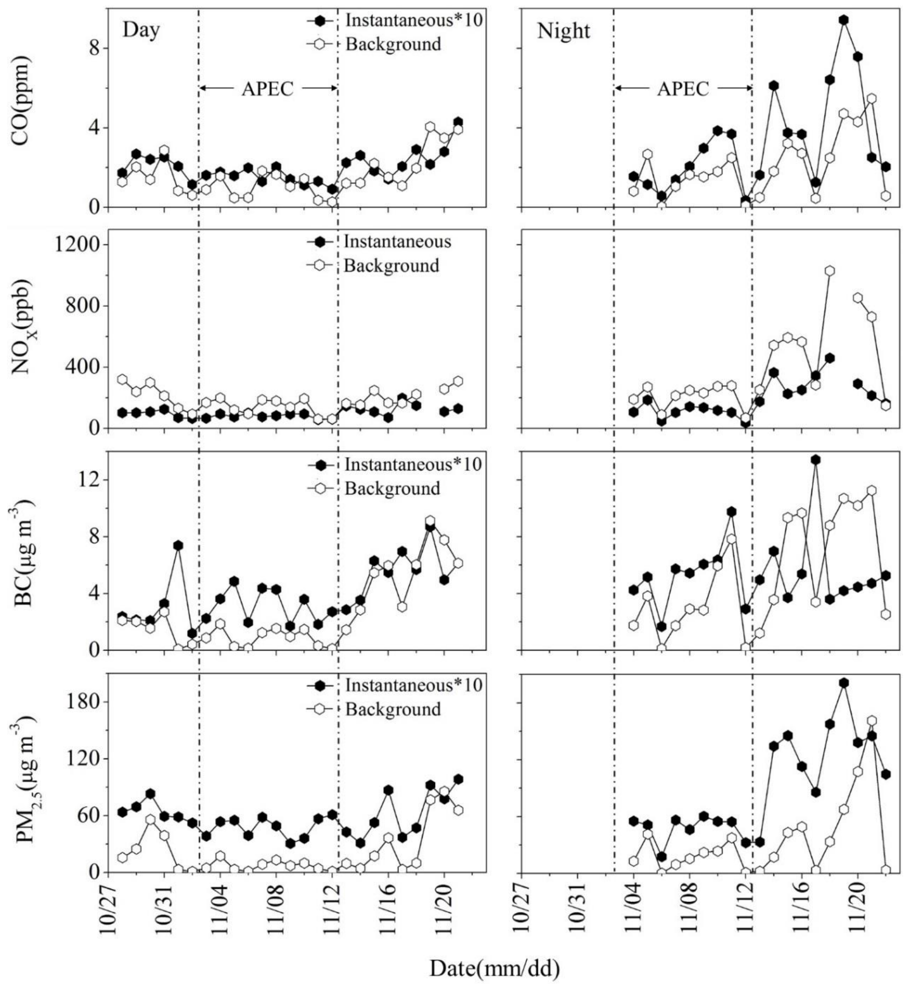

Fig. 7 Temporal variations of the average instantaneous concentrations and on-road background concentrations of CO, NO_x, BC, and $PM_{2.5}$ decomposed from on-road measurements results by CWT for both daytime and night-time measurements. The solid dots and the open dots represent instantaneous concentration and on-road background concentration, respectively. Middle area shows the APEC period.