論 文 の 内 容 の 要 旨

論文題目　A Study on Elemental Carbon in China
(中国における元素状炭素の研究)

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This thesis reports results obtained by the author in studies of ambient atmospheric elemental carbon (EC) in Beijing over a period of three years from winter 2005 to summer 2008. Throughout the text considerable attention is devoted to temporal variation focusing in EC and its dominant sources in Beijing.

By use of a semi-continuous thermal-optical analyzer with high-time resolution of one-hour, levels of EC concentrations in air were determined at Peking University in Beijing (PKU), China. Simultaneously, gaseous components, CO and CO_2, meteorological data, and traffic properties were measured. The measurement of those species was also conducted at the Yufa site, about 53 km south of the PKU site, in the summer of 2006 as a part of the Campaigns of Air Quality Research in Beijing and Surrounding Region 2006 (CAREBeijing 2006). Comparing with the obtained data at the Yufa site, the observed data was found to represent the values in the region within 50 km from the PKU site.

The annual mean of EC concentration was 6.9 μgC m^{-3}, which was about 1.5-4 times higher than the level in other Asian urban areas, such as Seoul and Tokyo. The concentrations of the observed species decreased with increasing near-surface wind speed (WS). The concentrations reached low and stable levels at a WS of about 4 m s^{-1}. The concentrations at stronger winds were considered to the background levels, and those in southerly air were much higher than northerly air. Under low-wind speed conditions (WS $\leq$ 2.0 m s^{-1}) of each species are interpreted to be more strongly controlled by regional-scale air mass affected by emissions from Beijing.

The slopes of the CO-CO_2, EC-CO_2, and EC-CO correlations in the weak-wind regime (WS $\leq$ 2.0 m s^{-1}) are used to estimate major EC and CO sources in Beijing. The median EC, $\Delta EC/\Delta CO_2$ (except for summer), and $\Delta EC/\Delta CO$ (except for winter) increased in the late evening and remained high until early morning. These diurnal variations indicate regular sources of EC throughout the year. According to the traffic volume data measured on the North-fourth ring road, the total vehicles were high during daytime, while the numbers of heavy-duty diesel trucks (HDDTs) and their fraction to total vehicles increased at 2200 LT and remained high during nighttime. The nighttime increase in the traffic of HDDTs is due to the traffic regulations, which allow trucks to enter the city only during nighttime (2200-0600 LT). These results indicate dominant contribution of exhaust from diesel vehicles to the nighttime enhancement of EC.

In winter, the CO showed higher concentrations compared with those in other seasons.

Especially, the nighttime CO and $\Delta CO/\Delta CO_2$ ratio were largely higher than those in the other seasons. CO emissions associated with coal or biofuel combustion for domestic heating are not the main cause of the high CO in winter, considering the lack of expected increases in EC in winter above the levels in the other seasons. Instead, the increase in the CO emissions from the exhaust of gasoline vehicles at low temperature is likely the dominant cause. The time required for catalysts to be heated to temperatures high enough for efficient CO removal is elongated at low temperatures, leading to higher CO emissions. The similarity in $\Delta EC/\Delta CO_2$ between winter and the other seasons indicates no significant additional EC emissions in winter. These results suggest that the vehicular sector is one of the most dominant sources to impact on the diurnal and seasonal variation of CO and EC in Beijing.

EC, CO, and CO_2 showed no significant decrease on the weekend. In addition, the diurnal variations of EC, CO, CO_2, and $\Delta EC/\Delta CO$ were similar between weekdays and weekends, confirming the absence of the differences in the diurnal variation of emission activity. This finding shows no significant weekend effect in Beijing.

A series of aggressive measures was launched by the Chinese government to reduce pollutant emissions from Beijing and surrounding areas during the Olympic Games. Observations showed significant decreases in concentrations of EC and CO during the Olympic season 2008, relative to the summer seasons in 2006 and 2007. The median concentration of EC was 2.22 $\mu gC\ m^{-3}$ in summer 2008; about 2.5 and 3.9 $\mu gC\ m^{-3}$ (53 and 63%) lower than for summer in 2007 and 2006, respectively. The median concentration of CO in summer 2008 was 660 ppbv, which was reduced about 16 and 33% compared with those in summer 2006 and 2007, respectively. Under weak-wind speed ($WS \leq 1.0\ m\ s^{-1}$) conditions, large decrease for EC concentrations during the Olympics would have to be related to the significant reductions of EC emissions in the Beijing urban area. Especially, large reduction of the nighttime EC concentrations and the change in the diurnal variation of $\Delta EC/\Delta CO$ ratios without the nighttime enhancement in summer 2008 strongly reflected the decrease in the traffic of nighttime heavy-duty diesel trucks by traffic restrictions. These indicate that control measures for Olympics targeted at the vehicle would sector were effective in reducing EC emissions in Beijing resulting in a significant source. This result suggests that the dominant EC source in Beijing is the transportation sector, especially diesel-fueled vehicles.

The observed slopes of the CO-CO_2-EC correlations for one year, four seasons in 2005-2006, were used to evaluate the emission ratios of these species derived from the emission inventory by *Streets et al.* [2003] in the Beijing area. The comparison suggests that *Streets et al.* [2003] largely overestimate the contributions of the emissions from domestic sectors with high CO/CO_2 and EC/CO_2 emission ratios and/or underestimate those from non-domestic sectors, especially vehicle emissions, in the Beijing study area. These results indicate that the observed slopes are useful parameters in assessing the reliability of emission inventories.