氏　　名	TAREK MOHAMED NASER
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学位論文題目	SPATIAL AND TEMPORAL ANALYSIS OF TRAFFIC-RELATED AIR POLLUTANTS AT DIFFERENT DISTANCES FROM MAJOR ROADS IN JAPAN （日本における主要道路からの自動車関連大気汚染物質の距離別時空間解析）
論文審査委員	委員長　教　授　坂本　和彦 委　員　教　授　吉門　　洋 委　員　准教授　藤野　　毅 委　員　准教授　王　　青躍

論文の内容の要旨

In urban environment, traffic emission is one of the most important sources of air pollution. Carbon monoxide, nitrogen oxides, hydrocarbons, particulate matter, and polycyclic aromatic hydrocarbons are known as traffic-related pollutants. The particle mass from motor vehicles is distributed between fine (aerodynamic diameter < 2.5 μm, e.g., $PM_{2.5}$) and ultrafine (aerodynamic diameter < 0.1 μm) particles, and is composed primarily of carbonaceous materials such as organic carbon and elemental carbon (EC) (Kittelson *et al.,* 1998, Kleeman *et al.,* 2000). Studies have shown that adverse health effects increase with proximity to roads, suggesting that motor vehicle emissions are responsible (Nicolai *et al*., 2003). Recently, in a first human experimental study examining the vascular effects of breathing pure ultrafine particles (UFP, ～25 nm size, and 95% EC), EC was found to alter pulmonary endothelial function, which is an important factor in the development of hypertension and heart failure (Shah *et al.,* 2008). Therefore, to address concerns about potential health risks near busy roads, the behavior of traffic-related pollutants should be clarified, particularly of those harmful constituents such as EC.

Knowledge of emission factors (EFs) is a key element in the exposure assessment. EFs of traffic-related air pollutants under the general traffic condition can be estimated from roadside monitoring data by making some assumption on the dispersion behavior (Hueglin *et al.,* 2006; Ning *et al.,* 2008). Among traffic-related pollutants, EF of EC that is more laborious to measure in dynamometric tests has been estimated by this method (Jiang *et al.,* 2005; Hueglin *et al.,* 2006; Kirchstetter *et al.,* 2008; Ning *et al.,* 2008). EF of EC can be estimated using pre-determined EF of NOx and assuming that the average atmospheric dilution of EC is identical to that of NOx (Hueglin *et al.,* 2006). We shall adopt this assumption using the Gaussian plume model to obtain EF of EC under more strictly chosen atmospheric conditions.

During our pilot research, the fate of traffic-related emissions on the urban particulate matter ($PM_{2.5}$ and PM_{10}) was investigated. The chemical compositions of $PM_{2.5}$ and PM_{10} and associated high-molecular-weight polycyclic

aromatic hydrocarbons (PAHs) were investigated during winter and summer ($PM_{2.5}$ and PM_{10} only) at a roadside and an urban background site in Saitama, Japan. The average concentrations of $PM_{2.5}$ found exceeded the United States Environmental Protection Agency standards during both periods. Carbonaceous components (elemental and organic carbon) were abundant in both the observed and calculated (by means of a mass closure model) chemical composition of $PM_{2.5}$. Traffic-related pollutants (elemental carbon and high-molecular-weight PAHs) were strongly associated with $PM_{2.5}$ rather than with larger particles. The mass concentrations of $PM_{2.5}$, as well as those of EC and PAHs associated with the particles, at the two sites were strongly correlated. Comparison of our data with source profile ratios indicates that diesel-powered vehicles were probably the main source of the measured PAHs. Our results emphasize the risks associated with human exposure to $PM_{2.5}$ in urban areas. These risks are expected to be particularly high in areas located near traffic roads with high volumes of diesel-powered vehicles. These results highlight the need for the establishment of standards for $PM_{2.5}$ in Japan. To establish these standards, the characteristics of traffic-related emissions should be well studied near major urban roads with long-term monitoring period.

Due to these above results we investigated the characteristics of traffic-related air pollutants monitored simultaneously near major roads in Japanese cites for one year. The monitoring campaign is a part of an epidemiological study (SORA project, Study On Respiratory disease and Automobile exhaust) carried out by Japanese Ministry of the Environment for the planned period of 2005-2009 in Japanese urban areas (Kanto, Chukyo, and Kinki). The monitored pollutants are nitrogen oxides (NOx), black carbon (BC), from which EC is calculated, suspended particulate matter (SPM), and $PM_{2.5}$. The present work focuses on these aspects: (1) evaluation of the conventional Gaussian plume model in built up areas, (2) determination of EF for automobile-originated EC that has been less understood than those of NOx or SPM and (3) investigating the factors that might affect the dispersion of traffic-related air pollutants, particularly the atmospheric stability, and traffic characteristics. Traffic-related air pollutants (SPM, $PM_{2.5}$, NOx, and BC) are simultaneously and continuously monitored at various distances from major roads at 10 sites with heavy traffic in six Japanese prefectures (Tokyo, Chiba, Kanagawa, Aichi, Hyogo, and Osaka) of Japanese urban areas (Kanto, Chukyo, and Kinki). We examine the data of one-year period (Jan. 1st ～ Dec. 31st, 2006) at all sites where the emission from the road traffic can be estimated relatively accurately. These sites are chosen by recent traffic census as neighborhoods of the highest DEP (Diesel Exhaust Particles) emitting roads in Japan. The road structure ranges from surface only, elevated only to surface and elevated.

For wind directions near perpendicular to the target roads, neutral atmospheric stability and sufficiently high wind speed, the observed data are compared with the prediction by the Gaussian plume model. Except for two sites with significant emission from the side street or with high densely built buildings, the Gaussian plume model agrees well with the observation for NOx and EC if concentrations are normalized to remove the background values and to minimize the estimation errors of the emission rate.

Background-subtracted NOx and EC are shown great dependence on road-normal wind direction and are strongly correlated at all analyzed sites as well. The emission factor of EC is estimated by evaluating the ratio $(C_{NOx} - C_{NOx,\ background}) / (C_{EC} - C_{EC,\ background})$. We assumed that the emission factor of EC is proportional to that of PM. Good agreement between observation and prediction by the Gaussian plume model was obtained with proportionally constant $(EC/PM)_v = 0.4$. Although this value depends on the correctness of the emission factors of PM and NOx and the Gaussian plume model, it is comparable to the results of recent chassis dynamometer tests reported by JAMA (NIES, 2007).

The influence of atmospheric stability on the traffic-related pollutants was also investigated. Traffic-related pollutants (NOx, EC, SPM, and $PM_{2.5}$) showed relatively lower concentrations during stable conditions than those

observed during other conditions (neutral and unstable). At most of sites, the atmospheric stability has influenced the concentration to increase in the following order; stable conditions < neutral conditions < unstable conditions. This order was obviously clear for near road-normal wind conditions. However, at the further distances, the concentrations showed relatively higher values during stable conditions than those monitored during unstable conditions. Under neutral stability condition, better agreement between calculated and observed decay is resulted at most of the sites.

論文の審査結果の要旨

当学位論文審査委員会は、当該論文の発表会を平成 21 年 2 月 6 日に公開で開催し、約 40 分の発表の後、本論文に関する詳細な質疑を行い、論文内容を審査した。

以下に論文内容を示すとともに、学位論文審査の結果を要約する。

Traffic-Related Air Pollutants and Their Dispersion near Roads (Literature Review)

Through this chapter recent researches concerning the characteristics of traffic-related air pollutants are reviewed. Due to the importance of the spatial variation of these pollutants, we focused on their decay trend with the distance from the road. Various parameters that can affect that decay such as wind direction, wind speed, and road features have been discussed.

Emission Factors of Traffic-Related Air Pollutants (Literature Review)

In this chapter, the determination methods of emission factors (EFs) of black carbon (BC), or elemental carbon (EC), which is more laborious to measure in dynamometric tests, are discussed. Among these methods, tunnel measurements and estimation by roadside measurements are focused. Moreover, we discussed the variation in the ratio EC/PM for vehicle emissions among different study environments along with the factors affecting that variation. We concluded that roadside studies, where background concentrations are subtracted and assumed models are applied to account for the dispersion of exhaust gas plume, are the best alternative method in estimation of emission factors traffic-related pollutants.

Tracing the Fate of Traffic-Related Emissions on the Particulate Matter (PM2.5 and PM10) Measured At a Roadside and an Urban Background Area in Saitama

Through this chapter we are investigating the fate of traffic-related emissions on the urban particulate matter by conducting field study at a roadside and an urban background site in Saitama, Japan. The chemical compositions of PM2.5 and PM10 and associated high-molecular-weight polycyclic aromatic hydrocarbons (PAHs) were investigated. Carbonaceous components (elemental and organic carbon) were abundant in both the observed and calculated (by means of a mass closure model) chemical composition of PM2.5. Traffic-related pollutants (elemental carbon and high-molecular-weight PAHs) were strongly associated with PM2.5 rather than with larger particles. Comparison of our data with source profile ratios indicates that diesel-powered vehicles were probably the main source of the measured PAHs. Our results emphasize the risks associated with human exposure to PM2.5 in urban areas. These risks are expected to be particularly high in areas located near traffic roads with high volumes of diesel-powered vehicles.

Analysis of Traffic-Related Air Pollutants at Different Distances from Major Roads in Japan

We investigate the characteristics of traffic-related air pollutants (NOx, EC, SPM, and PM2.5) monitored simultaneously near major roads in ten Japanese cites for one year. The monitoring campaign is a part of an epidemiological study (SORA project, Study On Respiratory disease and Automobile exhaust) carried out by Japanese Ministry of the Environment for the planned period of 2005-2009 in Japanese urban areas (Kanto, Chukyo, and Kinki). The present chapter focuses on two aspects: (1) evaluation of the conventional Gaussian plume model in built up areas, (2) determination of EF for automobile-originated EC that has been less understood than those of NOx or

SPM. For wind directions near-perpendicular to the target roads, neutral atmospheric stability and sufficiently high wind speed, the observed data are compared with the prediction by the conventional Gaussian plume model. Except for a site with high densely built buildings and another with unaccounted suspected emission source, the Gaussian plume model agrees well with the observed NOx and EC concentrations normalized by the values at the stations closest to the road. Under the assumption that the emission factor of EC is proportional to that of PM (total particulate matter at emission) and also using emission factor of NOx, the emission factor of EC is estimated by evaluating the ratio $(C_{NOx} - C_{NOx,\ background}) / (C_{EC} - C_{EC,\ background})$.

The novelty of our work lies in the improved reliability of the results. Gross agreement with the previous work is merely a fortunate outcome. The improved reliability is achieved by the long monitoring period and the dense distribution of monitoring stations. As mentioned in the manuscript, the statistical variation is quite large even after removing the effects of the temporal variations of weather and traffic. Hence, sufficiently long (but not too long as explained below) evaluation period is necessary. Since previous works were at most for a month (one case had one-year monitoring period, but the stations were very sparse), the reliability of the results were significantly lower than ours.

The Influence of Atmospheric Stability on Traffic-Related Pollutants

In this chapter, the influence of atmospheric stability on the dispersion of traffic-related pollutants (pollutant decay and concentration rose) is discussed, as well as on NOx/EC ratio. At most of sites, the atmospheric stability has influenced the concentration to increase in the following order; stable conditions < neutral conditions < unstable conditions. This order was obviously clear for near road-normal wind conditions very close to the road. However, at the further distances, the concentrations showed relatively higher values during stable conditions than those monitored during unstable conditions. Under neutral stability condition, better agreement between calculated and observed decay is resulted at most of the sites. Both observed and calculated values of the ratio NOx/EC are not influenced by atmospheric stability.

上記のように、本研究では、まず道路近傍における自動車から排出される粒子状物質（SPM）中の炭素成分、特に微小粒子、有機炭素（OC）、元素状炭素（EC）、多環芳香族炭化水素（PAHs）に注目して、道路近傍と一般環境の比較を行った。次に道路を走行する自動車の排ガスの影響を調べるため、道路直近ならびに道路からの異なる距離に設置した測定点において、NOx、EC、SPM の連続モニタリングを行った。これらの研究結果として、道路近傍ならびに一般環境の SPM は測定成分濃度を利用したマス・クロージャーモデルにより十分説明できること、PAHs は微小粒子領域に存在し自動車排ガスからの影響が大きいこと、バックグラウンドの値を考慮した NOx/EC 濃度比を利用すれば自動車からの NOx/SPM を求められること、成分濃度は大気安定度に影響を受けるが NOx/EC で考えればその影響を除けること、などを明らかにした。これらの研究成果により、道路からの距離に応じた自動車排ガスからの健康影響を評価するための粒子状物質などの暴露レベルの推定は十分実現可能であると結論している。

以上に要約したように、本論文では学術的価値の高い成果が示されており、博士（学術）の学位を授与するにふさわしい内容を備えていると判断し、当学位論文審査委員会は合格と判定した。