



Application of Stack Emissions Data from Tele-Monitoring Systems for Characterization of Industrial Emissions of Air Pollutants

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ABSTRACT

Real monitoring data are required for the validation of models or methods used to estimate industrial emission inventories of air pollutants. As such, the current study investigated the industrial emissions of four major and three other hazardous air pollutants in four subareas of the Yeongnam area in Korea over five years (2005 through 2009), using direct stack monitoring concentrations, which were obtained from tel-monitoring systems. Yearly variations in the industrial emissions of the target pollutants depended upon the subareas and chemical types. For example, the total suspended particulate (TSP) emissions in Geongnam increased for two consecutive years (2007 and 2008) and then, decreased in 2009, whereas the industrial CO emissions in Ulsan sharply increased since 2007. For TSP, primary metal manufacturing (PMM) and Power, electricity and gas supplying companies (PEGSC) were two major industrial emission sources. PEGSC exhibited the highest source for both SO₂ and NO_x emissions, followed by PMM and petroleum and petrochemical companies in descending order. For CO emissions, PMM exhibited the highest source, followed by municipal waste treatment and intermediate waste treatment facilities in descending order. Both NH₃ and HF were emitted primarily from two subareas (Ulsan and Geongbuk), which have fertilizer production companies. Both Geongnam and Geongbuk exhibited the highest and the second highest HCl emissions, presumably due to emissions during the acid treatment processes of various metals. Most of the industry categories, which are associated with fuel combustions or waste incineration, exhibited the highest and the second highest emission proportions for NO_x and SO₂, respectively. Maximum emissions were observed in the winter or summer season, while minimum emissions were observed in the spring or fall season, presumably due to increased energy utilizations for residential as well as industrial heating and cooling.

Keywords: Industrial category; Industrial source; Fuel-combustion; Waste incineration; Seasonal variation.

INTRODUCTION

Emission inventories on air pollutants provide fundamental information for understanding regional air pollution, for judging the efficiency of existing air pollution management programs, and for developing future air pollution control strategies. Major air pollutant sources include motor vehicles and point sources such as industries and domestic activities (Elbir and Muezzinoglu, 2004; Yang *et al.*, 2008; Shen *et al.*, 2010). In particular, many previous studies (Na *et al.*, 2001; Rehwagen *et al.*, 2005; Pal *et al.*, 2008; Ras *et al.*, 2009; Lee *et al.*, 2011) have indicated that more concern about air pollution was received in industrial areas primarily due to elevated air pollution levels in these areas. The industrial air pollution levels were ascribed to high

air pollutant emissions from the consumption of various energies, industrial facilities, and processes. As such, industrial emission inventories on air pollutants hold the key to effective air-quality management programs, providing essential tools for the planning of control strategies to achieve ambient air quality goals in industrial areas.

Industrial emission inventories on air pollutants can be established by employing emission calculations or by monitoring air pollutants emitted through stacks (Elbir and Muezzinoglu, 2004; Zheng *et al.*, 2009). Emission calculations using emission factors have mostly been utilized for estimating air pollutant emissions mainly because of lower costs or lack of stack monitoring data (Elbir and Muezzinoglu, 2004; Olcese and Toselli, 2004; Bhanarkar *et al.*, 2005). However, this estimation method has a disadvantage of having a lower accuracy for the emission data. To overcome the disadvantage of the estimation method, continuous monitoring data can be employed since source-monitoring activities provide a better accuracy for air pollutant emission estimation (Elbir and Muezzinoglu, 2004). Nevertheless, this method still

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has the disadvantage of high costs for collecting real monitoring data. As such, estimation methods validated with real monitoring data are strongly suggested to obtain emission inventories of air pollutants, which can compensate for the disadvantages of the two methods.

Emission data on industrial air pollutants determined by using stack-monitoring concentrations, which are utilized for the validation of emission estimation models or other theoretical methods, can hardly be found in published literature. The Korean ministry of environment (KME) required the Korean Environmental management corporation (KEMC) to establish an industrial stack tele-monitoring system (TMS) for major and a few hazardous air pollutants. Thus, TMSs have been installed in industrial stacks since 1988 as a part of an advanced air-quality management program. Recently, the KEMC has established four regional control centers each located in one of four subareas (Yeongnam, Honam, Jungbu, and Sudo areas). The centers are in charge of the collecting, analyzing, and transmitting the stack monitoring data, performing quality assurance and quality control programs on the monitoring data, examining any abnormal data, and providing technological support to industries (Choi, 2010). Consequently, the present study attempted to supplement the lack of industrial emission data determined by using direct stack monitoring concentrations, which were obtained from tele-monitoring systems (TMSs) for one of the four subareas, the Yeongnam area. The Yeongnam area is located in an area southeast of Seoul, which is the capital of Korea and consists of three metropolitan cities (Busan, Daegu and Ulsan), and two large provinces (Gyeongnam and Gyeongbuk).

METHODS

The number of TMSs installed in the five survey areas was determined based on the number of major industrial emission sources. The three metropolitan cities (Busan, Daegu, and Ulsan) have a population of 3.6 million with an area of 767 km², a population of 2.5 million with an area of 885 km², and a population of 1.2 million with an area of 644 km², respectively. The two provinces (Gyeongnam and Gyeongbuk) have 18 cities or counties with a population of 3.3 million and an area of 10,533 km², and 23 cities or counties with a population of 2.7 million and an area of 10,027 km², respectively. The total number of industrial stack TMSs installed in the Yeongnam area was 174: Busan, 19; Daegu, 8; Ulsan, 52; Gyeongnam, 51; and Gyeongbuk, 44. Ulsan had more TMSs compared with other subareas since it is a petrochemical industry-concentrated city and has a power plant. Meanwhile, Busan and Daegu are typical Korean metropolitan cities, and Gyeongnam and Gyeongbuk are typical Korean provinces. However, Gyeongnam has a power plant. Seven major and other hazardous chemical species (total suspended particulates (TSP), SO₂, NO_x, CO, NH₃, HCl, and HF) were continuously monitored through the TMSs. TSP is monitored using authorized particulate monitors and the other six species are monitored using authorized gas

analyzers, which are installed after the dehumidifiers and separators connected to sampling probes. Representative methods for the concentration measurements of target compounds were as follows: TSP, light-scattering or light-transmission method; SO₂, UV absorption method; NO_x and NH₃, UV or infrared absorption method; CO, infrared absorption method; HCl and HF, electrode or infrared absorption method. The stack concentrations of these pollutants were gathered according to industry categories over five years (2005 through 2009) to calculate stack emission rates. Thirty-minute average concentration data obtained from TMSs were regularly transmitted to the main computer in the Yeongnam TMS control center. Once the quality of the data was assured, the information was then transferred to the headquarters of the KEMC. We began our study with data from 2005 because the quality of the data for all the subareas for those years had been assured. The 30-min average air pollutant emissions were calculated by multiplying the monitored concentrations by the stack gas flow rate. There were 14 industry categories which were defined by the KEMC: chemical product manufacturing (CPM), food and beverage production (FBP), fertilizer production (FP), glass product manufacturing (GPM), heating and steam supplying companies (HSSC), intermediate waste treatment (IWT), mechanical products (MP), municipal waste treatment (MWT), non-metallic mineral manufacturing (NMMM), Power, electricity and gas supplying companies (PEGSC), primary metal manufacturing (PMM), petroleum and petrochemical companies (PPC), pulp and paper manufacturing (PPM), and wood and pulp manufacturing (WPM).

RESULTS AND DISCUSSION

Variations in Yearly Emissions in the Subareas for All Industry Categories

The general trends of major industrial air pollutant emissions were evaluated using the combined monitoring data of all industry categories for three metropolitan cities (Busan, Daegu and Ulsan) and two large provinces (Gyeongnam and Gyeongbuk) over 5 years. The variations in the yearly mean emissions of seven air pollutants according to the subareas are shown in Fig. 1(a) through Fig. 1(g). A yearly mean emission was calculated by averaging 30-min emission data obtained over a year. The yearly emission patterns varied according to both subareas and air pollutants. For TSP, overall emissions were the highest for Geongnam, second highest for Geongbuk, and third highest for Ulsan. Busan and Daegu had similar TSP emissions and the lowest emissions among the subareas. This order in TSP emissions is most likely due to electric arc furnaces (EAFs) used for PMM processes, which are associated with 83.8% of the industrial TSP emissions, whose details are described in the next section (Fig. 2(a)). The numbers of EAFs had the same order as the TSP emissions for the five subareas (Choi, 2010). In Geongnam, the TSP emissions increased for two consecutive years (2007 and 2008) and then, decreased in 2009. The increased TSP emissions are mainly attributed to the gradual increase

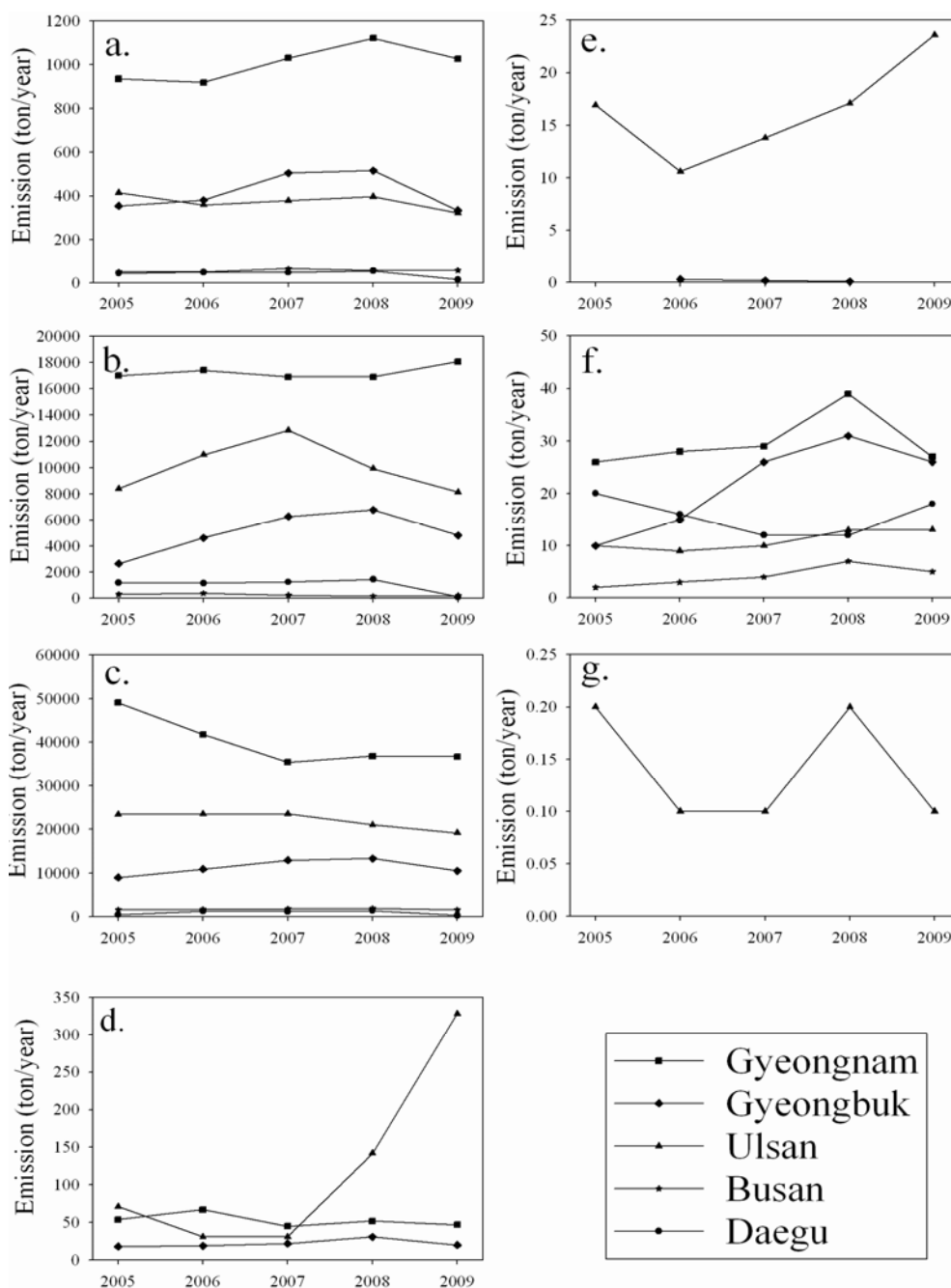


Fig. 1. Variations in the yearly mean emissions (average of 30-min emission data obtained over a year) of seven air pollutants for all industry categories according to subareas: a, TSP; b, SO_2 ; c, NO_x ; d, CO; e, NH_3 ; f, HCl; and g, HF.

of EAF number from four in 2006 to eight in 2007 and then to nine in 2009. However, the TSP emissions in Geongnam decreased although the number of EAFs was ten in 2009. This is closely linked to the strong enforcement of TSP emission regulations (KEMC, 2010). Since this strong enforcement was applied to other areas as well, TSP emissions in the other survey areas would also decrease in 2009.

SO_2 and NO_x are two of the major industrial-emitted pollutants in many industrialized cities (Bhanarkar *et al.*, 2005; Lucon and Santos, 2005; Zhang *et al.*, 2006; Kato and Akimoto, 2007; Zheng *et al.*, 2009). Similar to TSP,

Geongnam exhibited the highest emission for both SO_2 and NO_x . PEGSC are associated with 69.4 and 60.4 % of the industrial emissions of SO_2 (Fig. 2(b)) and NO_x (Fig. 2(c)), respectively. Thus, the highest emissions of SO_2 and NO_x in Geongnam are mainly attributed to the yearly combustion of 950 million tons of coal at the Hadong thermoelectric power plant for the energy production of 4,000 MW. Ulsan, which had the second highest emissions for both SO_2 and NO_x , also has a thermoelectric power plant having the yearly combustion of 40 million tons of heavy-oil at the Korean Nambu thermoelectric power plant for the energy production of 400 MW. Geongbuk, which

had the third highest emissions for both SO₂ and NO_x, consumed about 15 million tons of coal mostly in iron and metal industrial complex (IMIC) that included the Pohang Iron and Steel Co. Ltd., which is one of the most competitive steel companies in the world today along with about 1800 other non-ferrous metal production, iron, and steel Ferro-alloy manufacturing companies (Lee *et al.*, 2011). For Geongnam, the SO₂ emissions did not significantly vary over the years, whereas the NO_x emissions had decreased during the period from 2005 through 2007 and did not significantly vary after that. Similarly, Ulsan revealed a decreasing trend for NO_x emissions since 2007. The decreases of NO_x emissions in Geongnam and Ulsan appear to be due to the application of improved selective catalytic reduction (SCR) systems and low-NO_x-emission burners (LNB) for power generation (Choi, 2010). Ulsan also exhibited a decreasing trend for SO₂ emissions since 2007. This decrease is likely due to the increased utilization of low-sulfur containing fuels in many industries in Ulsan (UCER, 2010). Meanwhile, both SO₂ and NO_x emissions in Geongbuk and Daegu decreased in 2009, presumably due to the increased utilization of low-sulfur containing fuels and LNBs in many industries in Daegu (DMCER, 2010)

CO emission data were analyzed for three subareas only since they were not available for the remaining two subareas. It is noteworthy that the industrial CO emissions in Ulsan sharply increased since 2007, while they did not significantly increase over the years for both Geongnam and Geongbuk. PPM is related to 74.7 % of industrial CO emissions (Fig. 2(d)). According to the Ulsan city hall statistics, production yields increased dramatically for the Donhae Pulp and Paper Industry. As such, a possible explanation for the sharp increase in CO emissions in Ulsan is the increase in the incineration of wastes generated during pulp and paper manufacturing processes.

Both NH₃ and HF were emitted primarily from two subareas (Ulsan and Geongbuk), which have fertilizer production companies. Since NH₃ and HF are primarily emitted during production processes of urea fertilizers and multi-purpose fertilizers (Gilliland *et al.*, 2006; Sarawade *et al.*, 2010; Zhang *et al.*, 2011), respectively, their industrial emissions from these two subareas are attributed to the industries producing these two types of fertilizers. This assertion is supported by Figs. 2(e) and 2(g), which describe the contribution of CPM (85%) and FP (15%) to NH₃ emissions and of CPM to HF emissions (100%), respectively. According to the industrial statistics of the two areas, the yields in urea fertilizer production from CPM were about 10 times higher in Ulsan than in Geongbuk, thereby resulting in elevated NH₃ emissions in Ulsan. Since Ulsan was the only subarea that had multi-purpose fertilizer production companies, industrial HF emissions were reported for this subarea only. On the other hand, the yearly fluctuation in NH₃ and HF emissions in Ulsan appear to be due to the variations in yield of the fertilizers (UCER, 2010).

Based on the average emission values over 5 years, Geongnam and Geongbuk exhibited the highest and second

highest HCl emissions. HCl is generally emitted during acid treatment processes of various metals (Agrawal *et al.*, 2007; Agrawal and Sahu, 2009) and the numbers of acid treatment facilities for PMM were higher for Geongnam than for Geongbuk (Choi, 2010). Thus, the higher HCl emissions in Geongnam are attributed to PMM. This assertion is supported by Fig. 2(f), in that PMM contributes nearly half of the industrial HCl emissions. Meanwhile, the yearly fluctuations in HCl emissions in these subareas are presumed to be due to variations in acid applications for PMM processes.

Comparison of Industry Categories for Individual Pollutant Emissions in the Yeongnam Area

Fig. 2 represents the contribution of the industry categories to the emissions of the seven air pollutants in the Yeongnam area. For TSP, PMM (83.8%) and PEGSC (11.8%) were the two major industrial emission sources with a proportion of 95.6% (Fig. 2(a)). Machado *et al.* (2006) also reported that the electric arc furnace for PMM processes is a major source of particulate emissions. In addition, fossil-fuel combustion in fired power could emit significant amounts of airborne particulate matters (Zhang *et al.*, 2005; Koornneef *et al.*, 2010). PEGSC was a major emission source of both SO₂ and NO_x emissions, with proportions of 69.4 and 60.4%, respectively (Figs. 2(b) and 2(c)). Previous studies (Koornneef *et al.*, 2010; Nazari *et*

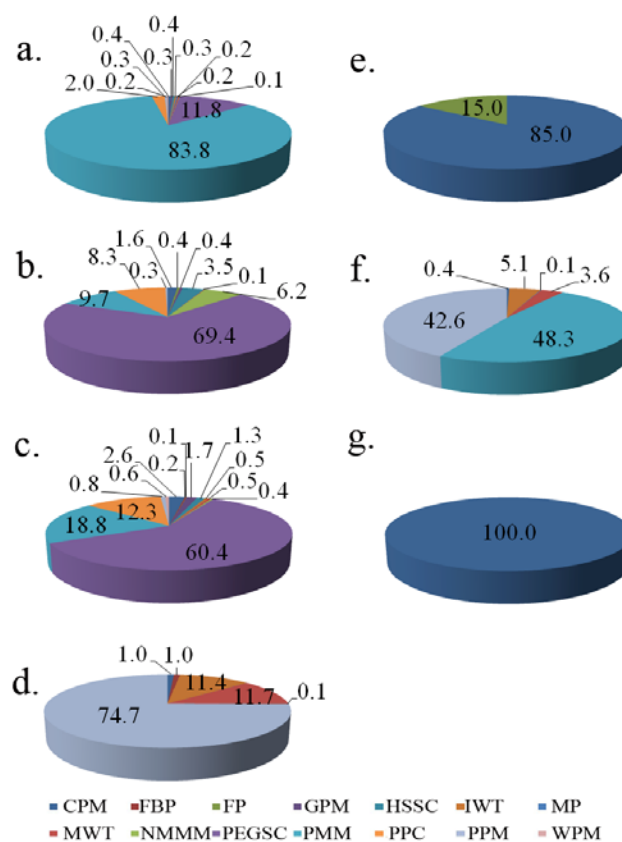


Fig. 2. Comparison of industry categories for the emission of seven air pollutants in the Yeongnam area: a, TSP; b, SO₂; c, NO_x; d, CO; e, NH₃; f, HCl; and g, HF.

al., 2010) also reported that fossil-fuel combustion in power plants is a major cause of these pollutants. PMM was revealed as the second highest source of both SO₂ and NO_x emissions. Sintering furnaces and electronic arc/sintering furnaces for PMM processes are important emission sources of SO₂ and NO_x, respectively (Li *et al.*, 2008; Song *et al.*, 2010). Cogeneration systems, which supply heat and power to PPCs, were the third highest source of both SO₂ and NO_x emissions. This result is supported by Tsay (2003), who reported that a cogeneration system is also an important source for these air pollutants. PPM was the highest source of CO emissions, followed by MWT and IWT in descending order (Fig. 2(d)). These are associated with the incineration of paper and pulp waste, municipal solid waste, and wasted oils, respectively (Choi, 2010). Meanwhile, the comparisons of industry categories for the other three pollutants (NH₃, HCl, and HF) have been described in the earlier section.

Pollutant Emission Proportions for Individual Industry Categories in the Yeongnam Area

Emission proportions of the seven air pollutants for individual industry categories in the Yeongnam area were shown in Fig. 3. This section attempted to provide qualitative information in determining priority pollutants for emission controls in individual industry categories. Since this type of investigations can hardly be found in the literature, this section describes only the current data, without comparing to other studies. Eleven of the 14 industry categories, which exhibited the highest emission proportions for NO_x, were as follows: CPM, FBP, GPM, HSSC, IWT, MWT, PEGSC, PMM, PPC, PPM, and WPM. As described earlier, these industry categories are all associated with waste incineration or fuel combustion. In addition, most of these industry categories had the second highest emission proportions for SO₂, mainly due to use of fossil fuels in these categories. Based on this result, but without considering any relative toxic effects of the target pollutants, it is suggested that NO_x and SO₂ are the two priority pollutants for emission controls in those industry categories. Two industry categories (FP and NMMM) had the highest emission proportions for SO₂ (Figs. 3(c) and 3(i)). This result is supported by a previous study (Choi, 2010), in that a significant amount of this pollutant is emitted during the manufacturing of sulfuric acid for both FP and NMMM. Meanwhile, MP had the highest and the second highest emission proportions for TSP (70.5%) and NO_x (26%), respectively (Fig. 3(g)). This is mainly attributed to TSP emissions during mechanical product manufacturing and to NO_x emissions from energy supplying boilers (Choi, 2010).

Monthly Variations in the Emissions of Total Pollutants in the Yeongnam Area

The seasonality in emissions of air pollutants is important when comparing emissions with time-specific measurements (Zhang *et al.*, 2009). As such, the present study also investigated monthly variations of the total industrial air pollutant emissions in the Yeongnam area by combining all

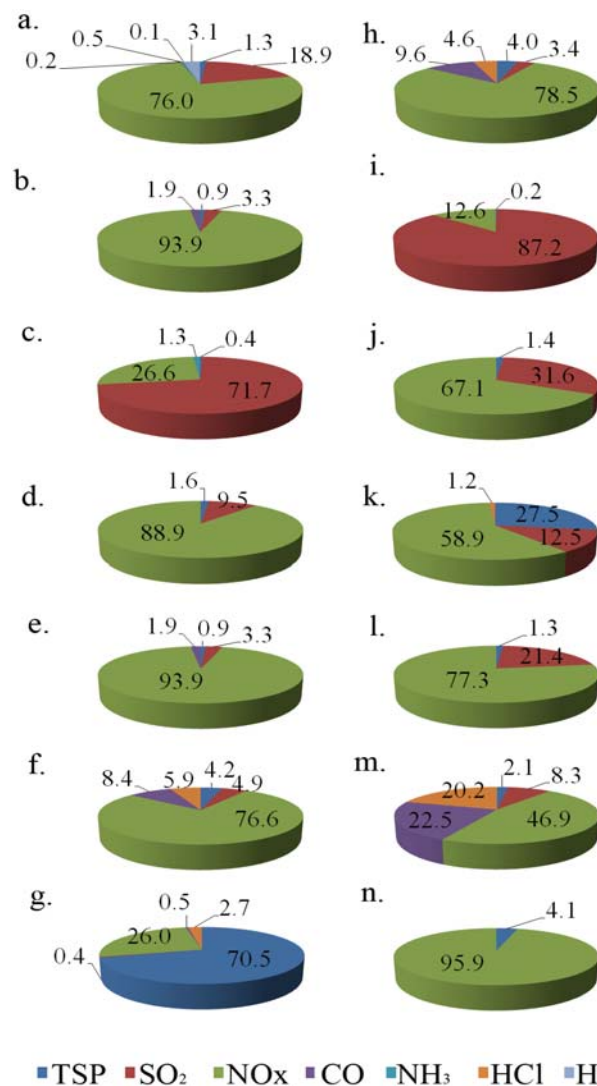


Fig. 3. Emission proportions of the seven air pollutants for individual industry categories in the Yeongnam area: a, CPM; b, FBP; c, FP; d, GPM; e, HSSC; f, IWT; g, MP; h, MWT; i, NMMM; j, PEGSC; k, PMM; l, PPC; m, PPM; and n, WPM.

the data from the five subareas. Fig. 4 represents a clear seasonal variation in the total industrial emissions of target air pollutants for each of the five survey years. Maximum emissions were observed in the winter or summer season, while minimum emissions were observed in the spring or fall season. This seasonal emission characteristic appears to be primarily due to increased energy utilization for residential as well as industrial heating and cooling in the cold and hot seasons, respectively. This assertion is supported by the finding that more than 60% (a 5-year average of 68,506 ton) of the total air pollutant emissions (a 5-year average of 114,094 ton) in the survey areas were attributed to PEGSC (Choi, 2010). However, Zhang *et al.* (2006) has reported that, unlike residential emissions, industrial emissions did not significantly exhibit a seasonal cycle. Unfortunately, we could not get all the information to understand the differences in the seasonal pattern

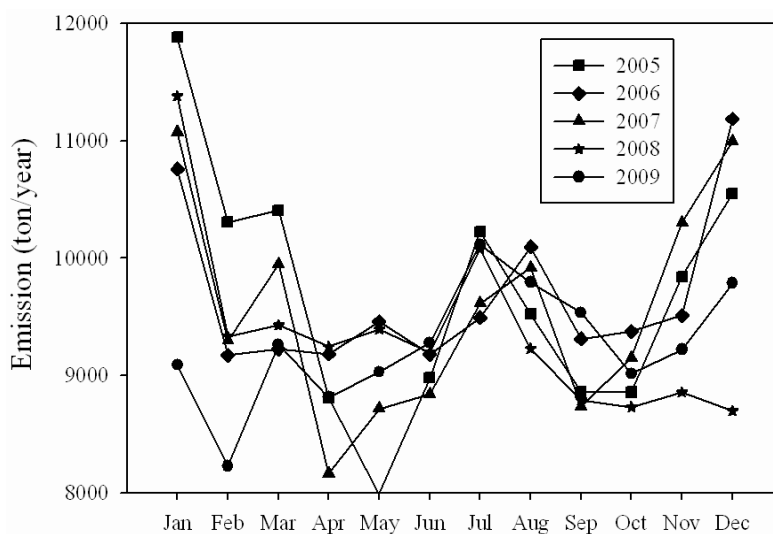


Fig. 4. Monthly mean (average of 30-min emission data obtained each month over five years, 2005 through 2009) variation of the total air pollutant emissions in the Yeongnam area.

between the two studies. Meanwhile, the monthly variations in total air pollutant emissions were not consistent for even the same season over the five survey years. For example, January's emissions was the highest in 2005, followed by 2008, 2007, 2006, and 2009, whereas December's emissions was the highest in 2006, followed by 2007, 2005, 2009, and 2008. Although the reason for this result is unclear, possible causes include irregular recent meteorological changes in even the same season and variations in industrial activities, which would change as the domestic and/or international economic situations change.

CONCLUSION

The present study was done to investigate not only the industrial emissions of four major and three other hazardous air pollutants using direct stack monitoring concentrations, which were obtained from TMSs, but also to provide real industrial emission data for the qualitative and semi-quantitative validation of emission estimation models or other theoretical methods. Yearly variations in the industrial emissions of the target pollutants depended upon the subareas and chemical types. For example, the TSP emissions in Geongnam increased for two consecutive years (2007 and 2008) and then, decreased in 2009, whereas the industrial CO emissions in Ulsan have sharply increased since 2007. For TSP, PMM and PEGSC were two major industrial emission sources. PEGSC was the highest source of both SO₂ and NO_x emissions, followed by PMM and PPC in descending order. For CO emissions, PPM was the highest source, followed by MWT and IWT in descending order. Industrial emissions of both NH₃ and HF were attributed to FP, and HCl emissions to acid treatment processes for PMM. There was a clear seasonal variation in total industrial emissions of target air pollutants for each of the five survey years, with the maximum emissions in the winter or summer season and the minimum emissions in the spring or fall season.

ACKNOWLEDGEMENT

This research was supported by the Research and Development Program through the Ulsan Regional Environmental Technology Development Center funded by the Korean Ministry of Environment.

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Received for review, March 2, 2011

Accepted, May 26, 2011