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雑誌名 | 地理の科学報告 第7シリーズ
巻 | 34
号 | 2
ページ | 71-81
発行年 | 1984-12
URL | http://hdl.handle.net/10097/45131
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Introduction

Air pollution due to sulfur dioxide had been a serious problem in the Southern Kanto region, Japan. After the Air Pollution Control Act in 1968, however, levels of SO$_2$ concentrations have decreased considerably. Fig. 1 shows the trend in the regional mean SO$_2$ concentration in the Southern Kanto region during the 1965-1980 period. A gradual downward trend is seen after a maximum value in 1967. The regional SO$_2$ level dropped from 0.056 ppm in 1967 to 0.016 ppm in 1980. There are two reasons for the decrease in recent years. 1) Reduction of SO$_2$ emissions because of emission-control actions such as replacement of high-sulfur with low-sulfur fuel, desulfurization of fuel and fuel gas desulfurization. 2) Dispersion from tall stacks.

![Fig. 1 Trend in the regional mean SO$_2$ concentration in the Southern Kanto region derived from thirteen sites where SO$_2$ concentration was measured continuously during the 1965-1980 period.](image_url)
Dispersion from tall stacks does not reduce emissions, but it considerably dilutes ground level concentrations. After the Air Pollution Control Act in 1968, stack height has been raised.

Decrease of SO$_2$ concentrations are evident in other source areas (Garnett 1980; van Dop and Kruizinga 1976). It also became clear that air pollution due to sulfur is an international, or interstate problem rather than a problem in solely the source areas as stacks become higher. In Europe, a large portion of sulfur originates from emission sources in neighboring countries (Elliasen and Saltbones 1983; Johnson et al. 1978; Ottar 1978; Rodhe 1972). This enhanced the interests of medium- or long-range transport of sulfur (Galvin et al. 1978; Perhac 1978; Rahn et al. 1980; Schiermeier et al. 1979) and its effect on acid rain (Likens et al. 1979).

Though levels of SO$_2$ concentration have decreased significantly in the last ten years, the extreme elevated SO$_2$ concentrations which exceeds the Japan 1-hour SO$_2$ criterion of 0.1 ppm frequently occur in the Southern Kanto region which covers the most densely populated and largest industrial area in Japan. This paper reports the result of statistical analysis of those extremely elevated SO$_2$ concentrations in the Southern Kanto region in the late 1970's, and discusses their relationships to meteorological conditions.

Data Description

Hourly ground-level sulfur dioxide data for the period April 1974-March 1979, were acquired from the telemetered Air Pollution Network System, which consists of nearly 300 sampling sites covering the Southern Kanto region (Tokyo, Kanagawa, Saitama and Chiba Prefecture). In this report, 115 sites were selected so that these sites are located relatively uniformly and have few missing measurements. Fig. 2 shows the location of the selected sites. Data during five winter seasons (December-February) were analyzed in this report, and other seasonal data were used to describe seasonal variations of concentrations.

Meteorological elements used in this report are daily maximum wind speed, daily maximum temperature and daily minimum temperature at the Tokyo Meteorological Station, shown on Fig. 2 by an solid square, which lies in downtown Tokyo. These meteorological data were extracted from the Monthly Report of the Japan Meteorological Agency.

Emissions within the Southern Kanto Region

To provide emission data for air quality studies, the Air Quality Bureau Environment Agency (1977) conducted detailed emission surveys and estimated anthropogenic SO$_2$ emissions from point and area sources inventories in the Southern Kanto region, using a 6 km grid system. Fig. 3 shown the SO$_2$ emissions in winter.
The Southern Kanto region covers the largest industrial area and the most densely populated area in Japan. Industrial areas located in coastal reclaimed zones west (Keihin-Industrial area) and east (Keiyo-Industrial area) of Tokyo-Bay consist of power plants, iron and steel mills, oil refineries and petrochemical plants. In the surrounding area, the Kashima-Industrial area is located northeast of the study region. The stack height in those industrial areas is generally high. In particular, the Keiyo and Kashima areas were recently developed, where large plants are located and stack heights are almost >100 m. From Fig. 3, it is clear that those industrial areas are emitting very high values and they are the major sources of pollutants.

Large cities that border the coast of Tokyo Bay such as Tokyo, Yokohama, Kawasaki, Urawa and Chiba have a total population in excess of 25 million. In such an urbanized area, space heaters are the main emission sources. The average emission density per unit area in urban areas is on the order of $10^{-1}$-$10^{-2}$, compared with those in industrial areas. Stack height is about 20-30 m in built-up areas such as in downtown Tokyo and about 10-20 m in other urban areas. The estimated vertical distribution of emissions (Air Quality Bureau Environment Agency, 1977), which was calculated from effective stack height indicates that maximum emission occurs at 20-
60 m in downtown Tokyo and at 20-40 m in other urban areas.

Results and Discussions

(a) Seasonal variation of elevated SO$_2$ concentrations

The number of hours in which 1-hour SO$_2$ concentration exceeded 0.1 ppm during a 5-year period in the Southern Kanto region was 2504. Fig. 4 shows the annual variation of these hours. The annual variation has a distinct yearly cycle, showing a
maximum in winter. The number of hours during winter (December-February) is 1273 which is 50.8% of the total hours. This annual variation corresponds reasonably well with SO₂ emissions from space heaters showing a maximum in winter and a minimum in summer, while large point sources in industrial areas such as power and chemical plant emit a reasonably steady amount from month to month.

The air flow during winter in the Southern Kanto region is normally dominated by a northwesterly component which brings fresh air from mountains. On the other hand, the sea breeze from Tokyo Bay prevails during the daytime during summer, which transports the pollutants from coastal industrial areas inland. Nevertheless, elevated SO₂ concentration tends to occur in winter. It is, therefore, assumed that elevated SO₂ values are mainly due to small emissions from space heaters in urban areas which have a large annual variation.

(b) Geographical distribution of elevated SO₂ concentrations

Fig. 5 shows the geographical distribution of elevated SO₂ values exceeding 0.1 ppm during five winter seasons (December-February) from 1974 to 1979. From Fig. 5,
it is seen that there are significant differences in the duration of elevated SO$_2$ values over the studied region, with the largest values being observed in downtown Tokyo.

This distribution of elevated SO$_2$ concentrations is not coincident with the distribution of emissions from tall stacks in industrial areas (cf. Fig. 3). In addition, taking into account the air flow during winter in which the strong northwesterly monsoon prevails over this region, it is concluded that the contribution from numerous small emissions from space heaters in the urban area is important in the elevated SO$_2$ values in the Southern Kanto region.

(c) Diurnal variation of elevated SO$_2$ concentrations

As shown in Fig. 6, elevated SO$_2$ values exceeding 0.1 ppm were observed during the daytime with a maximum value of 229 at 1000 JST. The number of hours of elevated concentration is more than 100 from 0900 to 1200 JST, and the sum total from 0900 to 1200 JST accounts for 51.5% of the elevated concentrations. This is more than 50 hours from 0900 to 1900 JST, and the total accounts for 88.7%, while at night and early morning the value is less than 10.

SO$_2$ concentration have in general two diurnal peaks in an urban area, one in the early morning and another in the evening, usually after sunset. The double daily peaks are primarily associated with stagnation of pollutants in a stable surface layer. In addition, the morning peak is probably caused by a corresponding maximum in the emission rate and may sometimes be caused by fumigation. The double daily peaks are reported in many cities (e.g. Bringfelt 1971). In the studied region, daily maximum SO$_2$ concentration tend to occur in the morning or in the evening in suburban areas (Misawa 1982). However, elevated SO$_2$ values, which mainly appear in downtown Tokyo (cf. Fig. 4), occur during the daytime with a peak in late morning when there is a thermally unstable condition due to surface solar heating. It, therefore, seems that extreme elevated SO$_2$ values exceeding 0.1 ppm have an intimate relationship with
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(d) Meteorological conditions associated with elevated SO$_2$ concentrations

Various motions of the surface boundary layer determine plume transport and diffusion. In particular, wind speed and stability are important elements. In an attempt to identify meteorological conditions associated with elevated SO$_2$ concentrations, all days during which 1-hour SO$_2$ concentration exceeds 0.1 ppm at one or more stations were selected from the five winter seasons and the relationship to those two elements was examined. The stability of the surface boundary layer is closely related to the vertical temperature profile. Unfortunately, there is no continuous measurement of vertical temperature profile in the studied area. Here, the diurnal range of air temperature was used as representing the stability of the surface boundary layer in the daytime in which elevated SO$_2$ values were mainly observed.

All 451 days during the five winter season were divided into the same groupings used by daily maximum wind speed and diurnal range of temperature at the Tokyo Meteorological Station, and the percentage of the days in which elevated SO$_2$ concentration was observed in each class was calculated. Daily maximum wind speed classes used were light (less than 5), moderate (5 to 9.9) and strong (10 ms$^{-1}$ or greater). Diurnal range of temperature classes used were small (less than 5), moderate (5 to 9.9) and large (10$^\circ$C or greater). Analysis was carried out separately for days when SO$_2$ concentration exceeded 0.1 ppm at one or more stations and days when it exceeded 0.1 ppm at six or more stations. The number of such days during five winter seasons were 191 and 31 days respectively.

Fig. 7(a) shows the result when SO$_2$ concentration exceeded 0.1 ppm at one or more stations. Higher percentage values are found with large diurnal range of temperature classes. It is, therefore, assumed that elevated SO$_2$ concentration tends to occur under thermally unstable surface boundary layer conditions during the daytime.

Plume characteristics widely vary due in large part to the stability of the surface boundary layer (and thus due to the intensity of vertical turbulence). Under thermally unstable conditions which are indicative of great turbulence, looping plumes are formed. Temperature gradients less than adiabatic near the surface while greater than adiabatic above is caused by surface heating during morning hours following stable nighttime conditions. Under such a condition a fumigating plume is formed. These two plume types cause elevated ground-level SO$_2$ values by vertical transport of pollutants. In particular, fumigating plumes are more critical for ground-level concentrations. A thermally stable layer above suppresses upward diffusion while it enhances downward transport by developing an unstable layer below the plume. Lyons and Cole (1973) showed that fumigating and looping plumes associated with an
unstable surface layer were important to higher ground-level SO₂ values along the shores of Lake Michigan. Davis and Newstein (1968) also reported that ground-level peak values of SO₂ were associated with fumigation in Philadelphia. van Dop et al. (1980) measured SO₂ concentrations at three levels (3,100 and 200 m) at Cabauw Tower, Netherlands, and showed that elevated ground-level SO₂ concentrations were caused by vertically turbulent transport of pollutants above the surface level during the daytime.

Higher concentrations caused by fumigating or looping plumes are associated with unstable and light wind conditions in the surface boundary layer. However, higher percentage values in Fig. 7(a) are also found with moderate/strong winds and a large diurnal range of temperature.

Air flow in an urban area is strongly influenced by buildings. Under strong winds, there are higher levels of vertical turbulence on the downwind side of buildings. Furthermore, plume rise is strongly depended on wind speed, and the height of plumes decrease with increased wind speed. This was made clear both by studies of plume height measurements (e.g. Bringfelt 1968; Hamilton 1967) and by theoretical studies of plume rise (e.g. Brummage 1968). As results of enhanced vertical turbulence and lower plume rise under strong winds, plume impinges on the surface near emission sources and thus higher concentrations are also observed at ground level. Higher ground-level concentrations associated with strong winds were reported by, for example, Lott (1982), Misawa (1977), Moore (1967) and Zanetti et al. (1977).

Fig. 7(b) shows the result when SO₂ values exceed 0.1 ppm at six or more stations.
The daily maximum wind speed/diurnal range of temperature classes with a high percentage is similar to that in Fig. 7(a). It is, therefore, assumed that elevated SO\textsubscript{2} values are caused at many stations with enhanced vertical transport of pollutants, and not with pollutant stagnation over the area.

It is concluded that elevated ground-level SO\textsubscript{2} concentrations are primarily determined by downward transport of pollutants at above levels. It is believed that those pollution phenomena associated with vertical transport of pollutants are important factors to higher ground-level SO\textsubscript{2} concentrations around high-level stacks. Observations of vertical profile of SO\textsubscript{2} concentration around high-level sources show that there are in general highly polluted air at elevated levels (Georgii 1978; Gillani et al. 1978; Smith et al. 1978; van Dop et al. 1980; van Egmond et al. 1978), though levels vary. Those high concentrations at above levels will cause high ground-level concentrations due to enhanced vertical turbulence (Husar et al. 1978; Lyons and Cole 1978). In an urban area, on the other hand, stagnation of pollutants near the surface has been considered as an important factor, because there are low-level emission sources. Unfortunately, there is little information for probability vertical profile of SO\textsubscript{2} concentration in urban areas. However, it is noted that emission sources in urban areas are low-level, and not surface-level. In particular, there are relatively high-level sources in downtown, and, thus, there are probably higher concentrations at above levels, and not at the surface.

Conclusions

Elevated ground-level SO\textsubscript{2} concentrations exceeding the Japan 1-hour criterion of 0.1 ppm are mainly observed during winter in the Southern Kanto region which covers the largest industrial area and the most densely populated area in Japan. The geographical distribution of elevated SO\textsubscript{2} values during winter is not coincident with the distribution of emissions characterized by emissions from tall stacks in industrial areas. The elevated SO\textsubscript{2} concentrations are mainly measured during the day in downtown Tokyo, and are caused by small low-level stacks (ca. 20-30 m) for space heating.

The elevated ground-level SO\textsubscript{2} concentration occurs on a day when the plume is transported downward by a thermally unstable condition of the surface boundary layer (and thus by enhanced vertical turbulence). The elevated concentration also occurs on a day when the plume is transported downward by greater turbulence on the downwind side of buildings, or when the plume impinges on the surface of nearby emission sources under strong winds. Stagnation of pollutants with light winds and stable conditions is of less importance to elevated ground-level SO\textsubscript{2} concentration. In other words, elevated ground-level concentrations of SO\textsubscript{2} are primarily determined by the intensity of vertically turbulent transport.
On the basis of these results, a serious question is raised: those pollution phenomena are seen at only nearby emission sources, thus does the monitoring site truly represent air pollution in urban areas? However, the elevated ground-level SO₂ concentration associated with unstable conditions or strong winds that appeared in downtown Tokyo is probably typical in urban areas, because a large number of small stacks are located at many places, and thus there are emissions at numerous sites in urban areas.

The types of elevated pollution described here, that result from fumigating or looping plumes and from downwash are not new phenomena around high-level stacks in industrial areas. However, it is important to emphasize that these phenomena are intimately concerned with extremely elevated SO₂ values in urban areas. In urban areas with a large number of low-level emission sources, stagnation of pollutants near the surface has been considered an important factor in elevated concentrations of SO₂. In fact, an air pollution advisory is issued on a day when stagnation is likely to occur. But, as noted above, extremely elevated SO₂ pollution occurs on a day when there is unstable or strong wind conditions at the surface layer and thus on a day when stagnation of pollutant is least likely to occur.

Acknowledgements

The author would like to thank Prof. H. Sitara, Tohoku University for the guidance and for valuable discussions on this study. The author also wishes to acknowledge Prof. J.C. Kimura California State University, Long Beach for his helpful advice.

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