

Long-Range Transport of Air Pollution under Light Gradient Wind Conditions

HIDEMI KURITA, KAZUTOSHI SASAKI AND HISAO MUROGA

Nagano Research Institute for Health and Pollution, Nagano, 380 Nagano, Japan

HIROMASA UEDA AND SHINJI WAKAMATSU

National Institute for Environmental Studies, Tsukuba, 305 Ibaraki, Japan

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ABSTRACT

The long-range transport of air pollution on clear days under light gradient wind conditions is investigated from an analysis of all days with high oxidant concentrations in 1979 at locations in central Japan that are far from pollutant sources. Surface-level wind and pressure distributions over a 300×300 km area were analyzed, together with concentration isopleths of oxidants and suspended particles produced by photochemical reactions.

It was found that the transport mechanism consists of: 1) land/sea breezes; 2) a steady onshore wind driven by the diurnal-mean land-sea temperature difference; 3) the generation of a strong thermal low in the inland mountainous region in the daytime; and 4) a subsidence inversion accompanied by a synoptic-scale high pressure system. The last three mechanisms work to combine land/sea breezes and slope and valley winds into one large-scale high-speed wind field that transports pollutants a long distance inland into the mountainous region.

1. Introduction

In the summer in central Japan, high concentrations of photochemical oxidants (OX) are frequently observed at inland stations far from industrial and populated areas. For example, the cities of Ueda and Nagano are located in the central mountainous region of Japan more than 160 km from large-scale emission sources. However, Ueda ranked seventh in Japan in 1979 for the number of days on which more than 120 ppb of OX was recorded. Furthermore, the daily maximum OX concentration in this region occurs in the late evening, mostly around 1900 JST (henceforth all times are JST) at Ueda and midnight at Nagano. Such a high nighttime concentration is the most remarkable feature of air pollution at these stations. Moreover, these high concentrations of OX occur on clear days with light gradient winds under synoptic-scale high pressure systems. These observations are indicative of the long-range transport of photochemical smog.

Similar air pollution at stations far from large emission sources has also been recorded in California. Edinger *et al.* (1972) and Carroll and Baskett (1979) discussed the mechanism in relation to an extended sea breeze and a synoptic-scale high pressure system. In central Japan the pollutants go through mountain passes and cause serious pollution within the mountainous region. The purpose of this paper is to elucidate the dynamics of this pollution process from

the meteorological point of view. All 35 days on which more than 100 ppb of OX was recorded at Ueda in 1979 are analyzed. Since this pollution is believed to be the result of long-range transport, the area analyzed is a 300×300 km region covering the central mountainous region, the Kanto district facing the Pacific Ocean, and the Hokuriku district facing the Japan Sea (Fig. 1). Distributions of the surface-level wind and pressure are investigated over this area, together with ground-level concentration measurements of OX and suspended particles (SP). For convenience, high OX concentrations observed later than 1800 are referred to as "night smog," while those observed earlier than 1800 are denoted as "daytime smog."

2. Geographical features and data analyses

The area analyzed here is the central part of Japan, the widest (300 km) region of Honshu (the main island of Japan). Figure 1 illustrates the geographical features of the region. The central mountainous region consists of mountain chains 2000–3000 m high with many basins and valleys that form a complex terrain. The average height of this region is ~ 1200 m MSL, and so it is called the "roof of Japan." The Ueda and Nagano basins investigated here are typical basins in this region and are connected by a narrow valley. Their average altitudes are 450 and 350 m MSL, respectively.

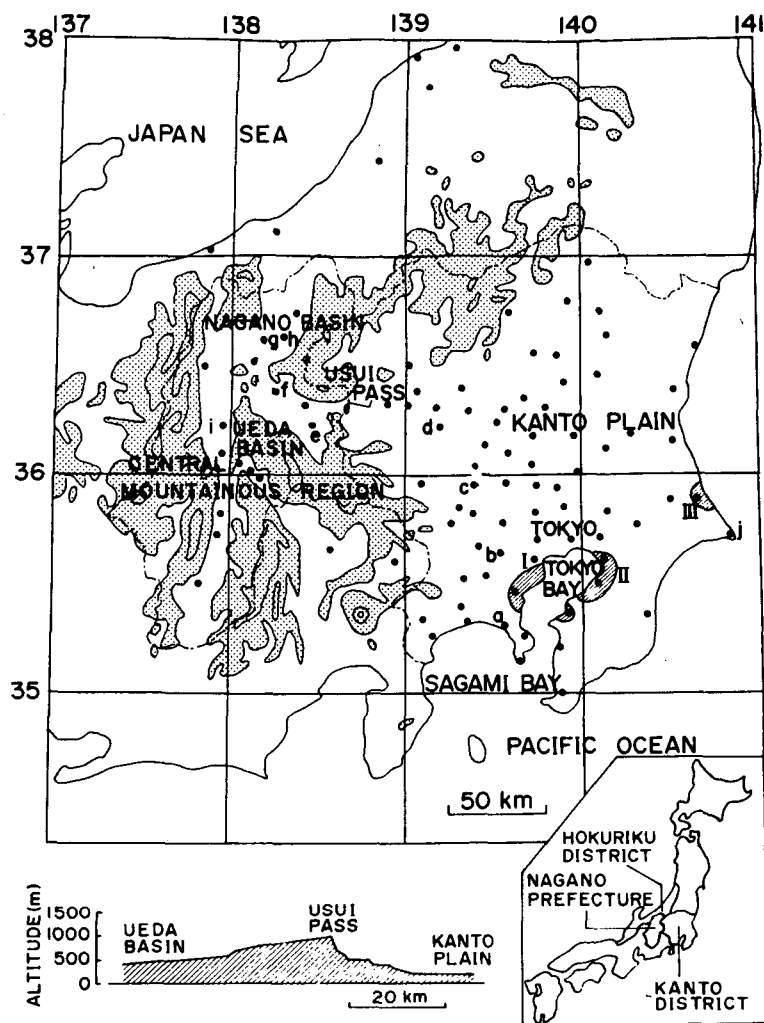


FIG. 1. Geographical features of the area analyzed, showing atmospheric monitoring stations (dots) and locations of large-scale emission sources (cross-hatched area). Industrial areas: I, Keihin; II, Keiyo; III, Kashima. Cities: a, Kamakura; b, Chofu; c, Sakado; d, Honjo; e, Saku; f, Ueda; g, Nagano; h, Suzaka; i, Matsumoto; j, Choshi. Land higher than 1000 m MSL is denoted by stippled area.

To the east of the central mountainous region, the Kanto Plain, the largest plain (15 000 km²) in Japan, extends to the Pacific Ocean. The central mountainous region and the Kanto Plain are connected by mountain passes about 1000 m high. Highly industrial and populated areas, i.e., the city of Tokyo (23 boroughs), and the Keihin and Keiyo industrial areas, are located in the coastal region around Tokyo Bay (Fig. 1). Large-scale sources of pollutant emissions are concentrated in these areas. The distances to Ueda and Nagano from these areas are 160 and 180 km, respectively.

All days (35) from May to September 1979 on which more than 100 ppb of OX were recorded at Ueda were selected for the analysis, which was performed in two steps:

1) Surface wind data from the Automated Meteorological Data Acquisition System (AMeDAS) were used to prepare a map of surface wind vectors every three hours. The surface wind data were used for this analysis because AMeDAS is a fully equipped and very dense monitoring system (one station per 450 km² on average). Also, large-scale wind fields that originated in the daytime and remained until the late evening were confirmed to be the main contributors to the long-range transport process, and these fields were rather uniform vertically up to at least 300 m altitude (Ueda and Kurita, 1983). Atmospheric pressures measured at all meteorological stations in this area were converted to their sea-level values and detailed distributions were plotted. Based on these data, the characteristics of the meteorology that cause

the high OX concentrations in the central mountainous region were examined.

2) Air pollution data at the atmospheric monitoring stations (95 stations were selected, as shown in Fig. 1) were analyzed to gain insight into the conditions leading to the high OX concentrations, and their time variations. For the analyses, the OX and SP (less than 10 μm in diameter) concentrations were measured by the neutral KI and light scattering methods, respectively.

Of the 35 days with high OX concentrations, 15 June 1979 was selected for further analysis. It was a typical night-smog day on which the highest OX concentrations were recorded both at Ueda and Nagano. Using OX and SP as tracers, their transport was investigated in detail in relation to the flow and pressure fields.

3. Surface wind fields on high concentration days

On days when the daily maximum concentration of OX exceeds 100 ppb at Ueda, two distinct large-scale wind fields can always be recognized in the daytime over the area analyzed: one is a wind field from the Pacific Ocean which covers the whole Kanto area, and the other is the extended sea breeze from the Japan Sea. Figure 2 shows such a typical daytime flow pattern at 1500, averaged over six days on which southerly sea breezes from Sagami and Tokyo bays prevailed over the Kanto area. This pattern is very different from those found in the morning and at night when there is no such large-scale wind field, with only localized light winds such as land breezes in the coastal regions and mountain winds in the mountainous region.

As shown by Fig. 2, the two wind fields form a convergence line. In Fig. 3 the convergence lines for all 35 days with high OX concentrations are depicted. From the occurrence of night smog in Ueda and/or Nagano, these days can be classified into three types:

1) The first type consists of six days where night smog occurred both at Ueda and Nagano (Case A). On these days the prevailing winds at these sites were from the Pacific Ocean from 1200 to 1800.

2) The second type was found on 19 days in which night smog was recorded at Ueda but not Nagano (Case B). On almost all of these days, the convergence lines lay between these two cities in the daytime and so Ueda was influenced by the wind from the Pacific Ocean while Nagano was within the Japan Sea wind field.

3) The third type consists of those ten days on which night smog did not occur at either Ueda or Nagano, but daytime smog occurred in both (Case C). On almost all days in Case C, convergence lines were located to the east of Ueda.

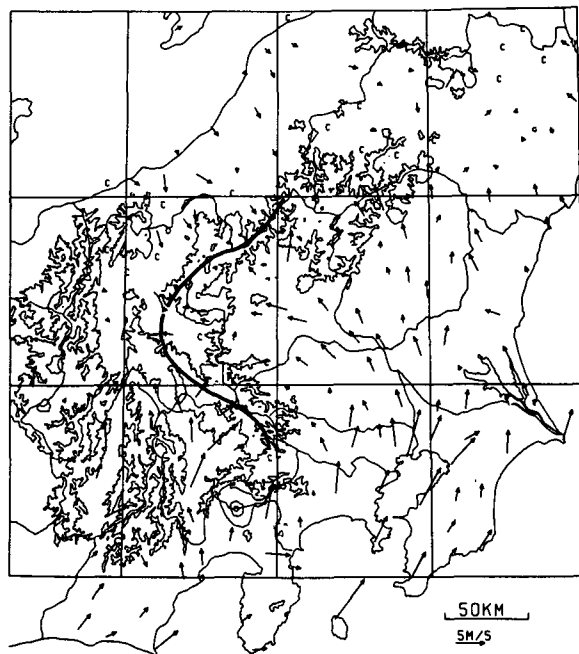


FIG. 2. A typical flow pattern at 1500 JST averaged over six days on which an extended sea breeze from Sagami and Tokyo bays prevailed over the Kanto district. The thick solid line represents a convergence line between the winds from the Pacific Ocean and the Japan Sea.

From the correspondence of the night smog with the location of the convergence line, it may be assumed that the condition necessary for night smog in this area is that the Pacific Ocean wind field prevails.

In order to elucidate the routes of the air masses that caused night smog in Ueda and/or Nagano (Cases A and B, 25 days), the surface wind field on the Pacific side was investigated in more detail. It can be classified roughly into two types. The type with the highest frequency (14 days) is shown in Fig. 2. In this situation, southerly sea breezes from Sagami and Tokyo bays prevail over the Kanto area, merging and turning to the west in the northwestern part of the area. The other type includes 11 days on which the southern part of the Kanto area was affected by sea breezes from Sagami and Tokyo bays, while the northern part was in an easterly wind field from the Sea of Kashima. With both types of wind fields, the air mass moved northward from Sagami and Tokyo bays and then turned toward Nagano Prefecture in the northwestern part of the Kanto area. Therefore, it is likely that the night smogs at Ueda or Nagano were caused by polluted air masses from the city of Tokyo, or other highly industrialized areas along Tokyo Bay, that travelled a long distance and entered Nagano Prefecture late in the evening.

On the other hand, daytime smogs (Case C) occurred when the Pacific Ocean wind did not intrude into Nagano Prefecture or when the wind in the

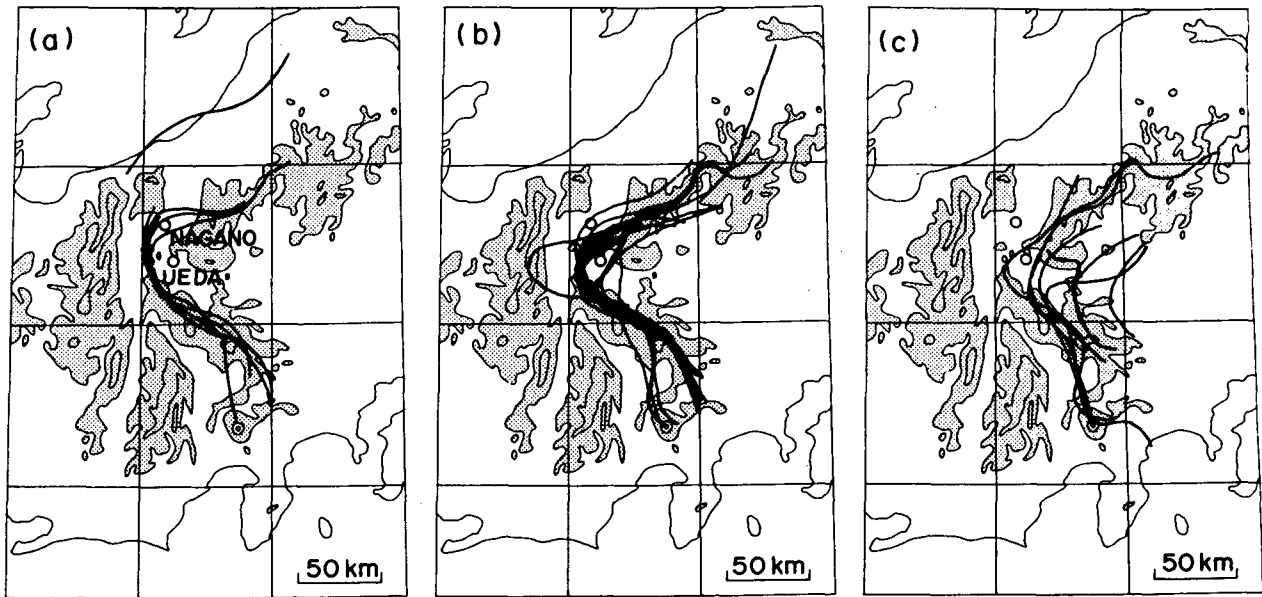


FIG. 3. Locations of convergence lines at 1800 JST for all 35 days analyzed. (a) Night smog both in Ueda and Nagano (Case A). (b) Night smog only in Ueda (Case B). (c) Daytime smog in both Ueda and Nagano (Case C).

Kanto area was northeasterly or easterly. Under these conditions, air masses entering Nagano Prefecture were not polluted; however, the daytime smogs were caused by the polluted air masses that had caused high OX concentrations one day before (on more than 70% of those days) and remained in these areas due to light surface winds averaging 1.0 m s^{-1} .

4. Generation of a thermal low

The diurnal variation of the wind field on 15 June 1979 when a typical night smog appeared at Ueda and Nagano is shown in Fig. 4. At 1000 sea breezes started to blow both in the Pacific Ocean and Japan Sea coastal regions. By that time slope and valley winds appeared in the central mountainous region and its surroundings. By 1300 the sea breeze from the Pacific Ocean side had penetrated far inland, and it had combined with the slope and valley winds to form one large-scale wind field. This wind field prevailed over the period from 1300 to 2000. Southerly winds from Sagami and Tokyo bays covered the southern part of the Kanto area, becoming easterly to the north and passing through the Usui Pass into the central mountainous region. There was clearly an airflow toward Nagano through the basins and valleys. In this way, winds from the Kanto area penetrated into Nagano Prefecture and formed a convergence line over Nagano with the wind field from the Japan Sea. The convergence line stayed in this region from 1400 to 2000. By 2100, the large-scale winds over the Kanto and Hokuriku areas were much reduced, but localized intense winds continued.

It is well known that middle latitude sea breezes penetrate less than 100 km inland when the gradient wind is weak and the land surface is flat (e.g., Atkinson, 1981; Asai and Mitsumoto, 1978). However, on days when high OX concentrations were observed in Ueda, large-scale wind fields were created both on the Pacific Ocean and Japan Sea sides about three hours after the start of the sea breeze, and continued until the late evening. In a previous paper (Ueda, 1983) it was shown that the diurnal-mean land-sea temperature difference drives a steady onshore or offshore wind, causing larger-scale seasonal changes in the climate of the coastal region. In the summertime, a steady onshore wind is superimposed on the land/sea breeze, enhancing the net onshore wind in the daytime, resulting in the so-called extended sea breeze.

In addition, a thermal low, generated and sustained in the central mountainous region in the daytime, is believed to contribute significantly to the creation of these large-scale wind fields. This is clearly seen in Fig. 5, in which a time series of the pressure distributions on a typical night-smog day (15 June 1979) is presented. By 1500, a strong low pressure area centered near Nagano had been generated, the central pressure being $\sim 6 \text{ mb}$ less than that in the Pacific coastal region. This low pressure enhanced the sea breezes and brought them to the central mountainous region.

The diurnal pressure change at the center of the thermal low (Nagano) was as follows; at ~ 0900 the pressure began to decrease, reaching a minimum at 1500. Then it recovered and reached the same level

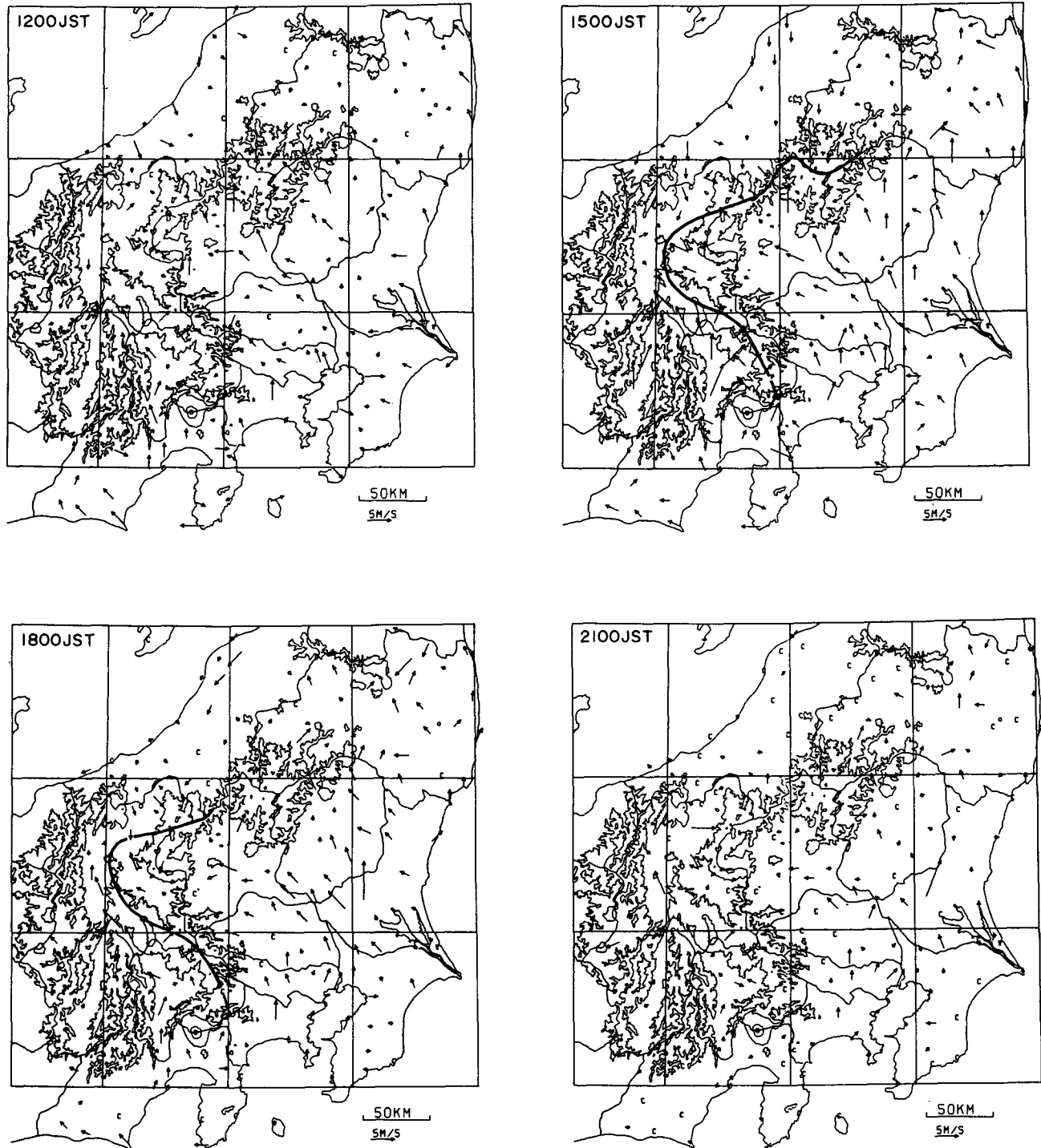


FIG. 4. Diurnal variation of the surface wind fields on 15 June 1979. The thick solid line represents a convergence line between the winds from the Pacific Ocean and the Japan Sea.

as the coastal region by 2100. The peak-to-peak value of the diurnal change was 6.2 mb, a remarkable contrast to the value of 0.7 mb observed at Choshi (Fig. 1) near the Pacific coast. Kimura (1975) presented a two-dimensional linear model of heat islands

that predicts a pressure decrease of only 1.9 mb at the center of a 300 km wide flat land surface, even when the land surface is 10 K warmer than the sea temperature. This large difference between the observed and predicted values suggests that the altitude

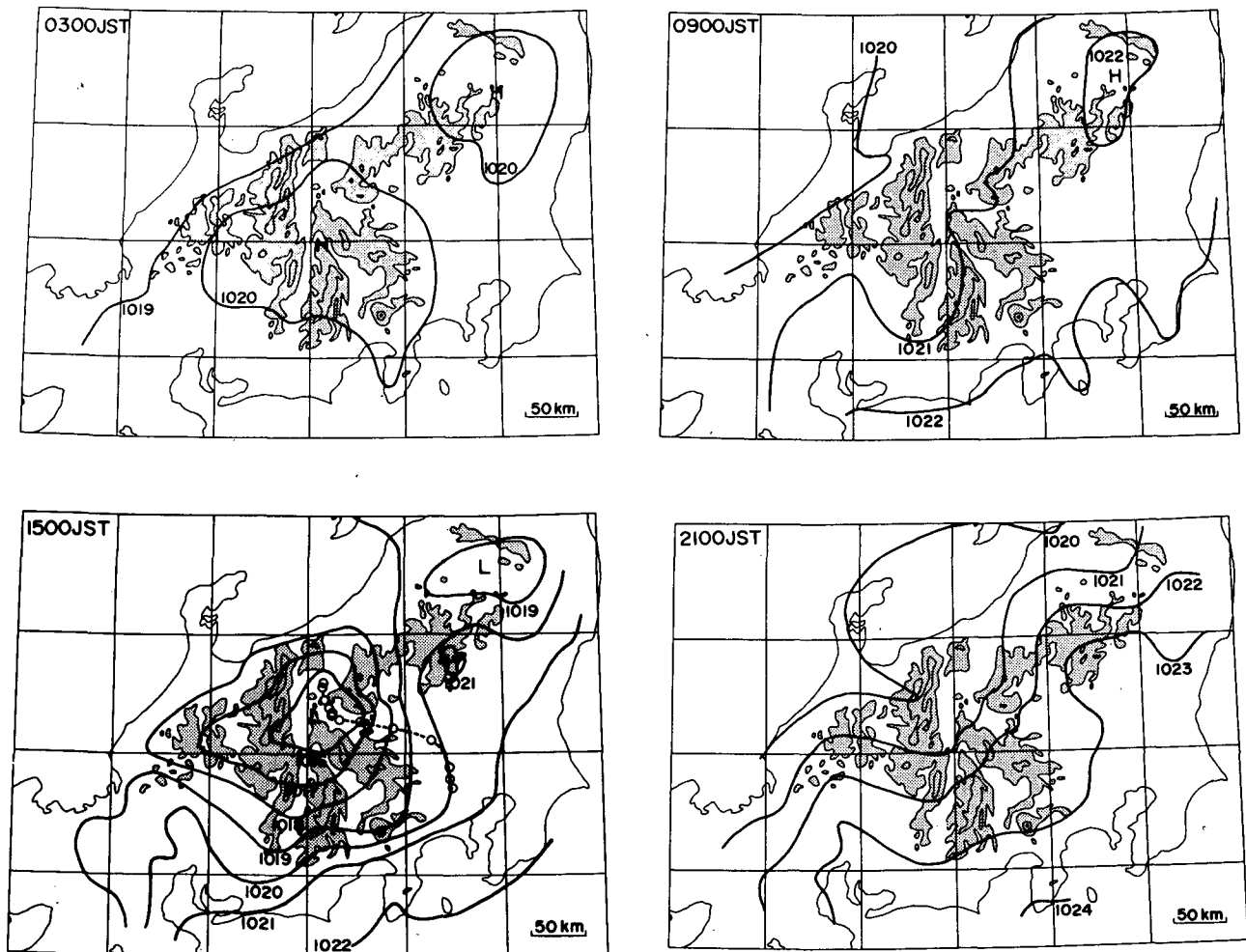


FIG. 5. Diurnal variation of the pressure distribution on 15 June 1979. Numbers indicate pressure (mb). Dashed line with open circles is trajectory of the high oxidant concentration zone.

of the land is very important. That is, differential heating between high-altitude land and the sea seems to generate a strong thermal low in the mountainous region. In contrast, the observed pressure generated in the central area due to differential cooling at night was only 1 mb higher than that in the coastal region. The asymmetry in the daytime and nighttime pressure distributions was due to a difference between the diurnal-mean land and sea temperatures. Corresponding to the generation of a strong thermal low in the daytime and a weak high pressure at night, an inflow to the central mountainous region started to blow at 1300, and easterly winds from the Kanto area into Nagano Prefecture continued to blow intensely from 1400 to 1700, and then at a reduced intensity until 2000. Moreover, the air mass that had been transported by the late evening remained in this region throughout the night.

On all 25 days when high nighttime OX concentrations were recorded at Ueda, such a thermal low

with a pressure difference of -6 to -4 mb (relative to coastal pressure) was generated and sustained in the daytime. Surprisingly, there was no exception. The center of the thermal low was located at either Nagano, Ueda or Matsumoto, in good correspondence with the tip of the convergence line between the Pacific Ocean and Japan Sea wind fields. Moreover, on those 25 days, a subsidence inversion accompanied by a synoptic-scale high pressure system was always formed at a level between 1000 and 4000 m. It is possible that this inversion confines the vertical extent of the airflow, and so confines the pollutants as well.

5. Movement of high-concentration zones

As shown in Table 1, the time of the maximum OX concentration at Ueda for the days analyzed was generally around 1900. However, the time was dependent on the location of the convergence line between the two large-scale wind fields. When the

TABLE 1. Time of the maximum oxidant concentration (OX \geq 100 ppb) at Ueda and its relation to the location of the convergence line between the winds from the Pacific Ocean and the Japan Sea.

Location of convergence line	Percentage of occurrences of maximum concentration at each time (JST, hours)											Total
	12	13	14	15	16	17	18	19	20	21	22	
North of Ueda	0	7	0	3	1	0	10	26	9	4	3	63
Vicinity of Ueda	0	0	0	3	3	8	6	0	0	0	0	20
East of Ueda	6	0	3	3	5	0	0	0	0	0	0	17
Total	6	7	3	9	9	8	16	26	9	4	3	100

convergence line lay to the north of Ueda, the maximum concentration occurred most frequently at 1900, and less frequently at 1800 and 2000. When the convergence line passed over the vicinity of Ueda, the maximum values were recorded with highest frequency at 1700, and next at 1800. In contrast, when the convergence line was located to the east of Ueda, the concentration maximum occurred either at 1200 or 1600 on most occasions, and it was not seen later than 1700.

In Nagano, however, OX concentrations attained their maximum values on average at 2100 when the convergence line stayed to the north. That is, the time of the maximum OX concentration was delayed along the route of the air mass travelling from the Kanto area; it was about two hours later than at Ueda. When Nagano was in the Japan Sea wind field, the maximum value appeared earlier than 1700.

On those days when night smog appeared at Ueda, high concentrations of OX and SP were also observed at many stations in the Kanto area. An analysis of data at these stations revealed that the time of the maximum concentration was delayed along the route of the polluted air mass. The movement of the zones of >100 ppb OX concentration and of $>100 \mu\text{g m}^{-3}$ SP is shown for a typical night-smog day, 15 June 1979, in Fig. 6. By 1400, high concentrations of OX and SP appeared 30 km inland from the coastline of Tokyo Bay. These zones had similar shape and size and moved northward at first and then turned to the west in the northwestern part of the Kanto area. At nearly 1800 they moved over the Usui Pass and arrived in the central mountainous region. The air mass was then transported through the valley between Ueda and Nagano and arrived at Nagano at 2100.

A trajectory analysis was made for the polluted air mass. First the upper winds were obtained by extrapolation of the surface data using the power-law profile with a power of $1/6$; then hourly three-dimensional wind fields were estimated by means of the variational calculus method with the continuity equation as a constraint (Kitada *et al.*, 1983). Trajectories of non-dispersive and nonbuoyant parcels emitted at the 10 m level near the center of the high concentration zone at 1400 were tracked using a technique developed by Ozoe *et al.* (1983). Particle path calculations were

made concurrently with linear interpolations of the estimated wind fields, since velocities at each grid point change with time. The particle velocity at any point was interpolated from the adjacent grid-point velocity in a second-order approximation, and the motion of the particle was computed for time steps of 30 s. The Runge-Kutta method was used to minimize the integration error. The trajectories in the mountainous region beyond the Usui Pass were determined by assuming that air parcels travel along the valley with speeds 1.5 times the surface values. The result is presented in Fig. 6c; it shows good agreement with the observed movement of the high-concentration zones.

Diurnal variations of the OX and SP concentrations at eight stations along the route of the polluted air mass are depicted in Fig. 7. The time of maximum concentration was progressively delayed along the route: 1400 at Chofu, 1700 at Honjo, 2200 at Ueda and 2400 at Nagano for both OX and SP. Assuming that the polluted air mass started at Chofu at 1400, the trajectory analysis predicts the maximum concentration times to be 1730 at Honjo, 2200 at Ueda and 2400 at Nagano, in good agreement with the observations.

Since a large amount of SP as well as OX results from photochemical reactions (Grosjean and Friedlander, 1975), the behavior of OX and SP is very similar (Figs. 6 and 7). However, a substantial difference between the behavior of OX and SP arose at night. Figure 8 presents a comparison of the OX concentration distributions at 1600 and 2200. Obviously, highly concentrated OX was widely distributed in the daytime, while at night the high-concentration zone was limited and the surrounding area had a concentration level of less than 30 ppb. As seen in Fig. 6, after the polluted zone arrived at Nagano at 2100 the zone of high OX concentration gradually decreased in size and then disappeared by 0300 the following day. However, the zone of high SP concentration remained in the Nagano and Ueda basins throughout the night. This difference may be explained by the reactions of OX with NO and other pollutants that are emitted from ground-level sources in the mid-sized cities along the route of the air mass, and/or by deposition on the ground-surface. As the

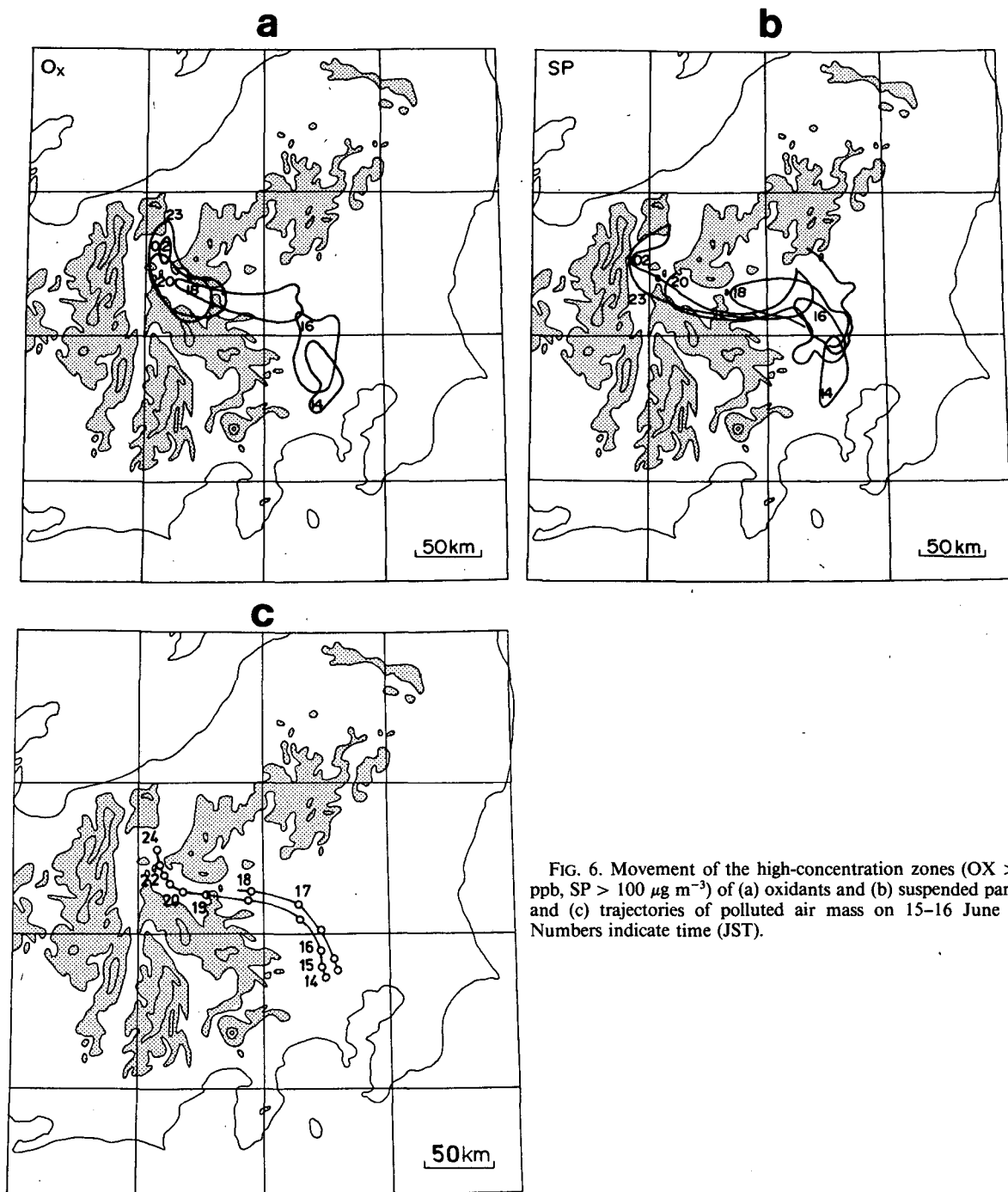


FIG. 6. Movement of the high-concentration zones (OX > 100 ppb, SP > $100 \mu\text{g m}^{-3}$) of (a) oxidants and (b) suspended particles, and (c) trajectories of polluted air mass on 15-16 June 1979. Numbers indicate time (JST).

nocturnal inversion develops near the ground surface at midnight, the turbulent diffusion of pollutants is damped (e.g., Ueda *et al.*, 1981; Fukui *et al.*, 1983). Under these conditions the loss of OX near the surface is not resupplied from above and so the ground-level concentration remains at very low levels in the early hours of the following day (Gloria *et al.*, 1974; Cox *et al.*, 1975). In contrast, the loss of SP

due to reaction and deposition may not be so large and does not influence the vertical distribution to such an extent (Husar *et al.*, 1977; Blumenthal *et al.*, 1978). This seems to be the reason why the maximum value of OX at Suzaka (Fig. 7) was less than half that in Nagano (the largest city along the route of the air mass; population 330 567), while the SP concentration remained at the same level as the other sites.

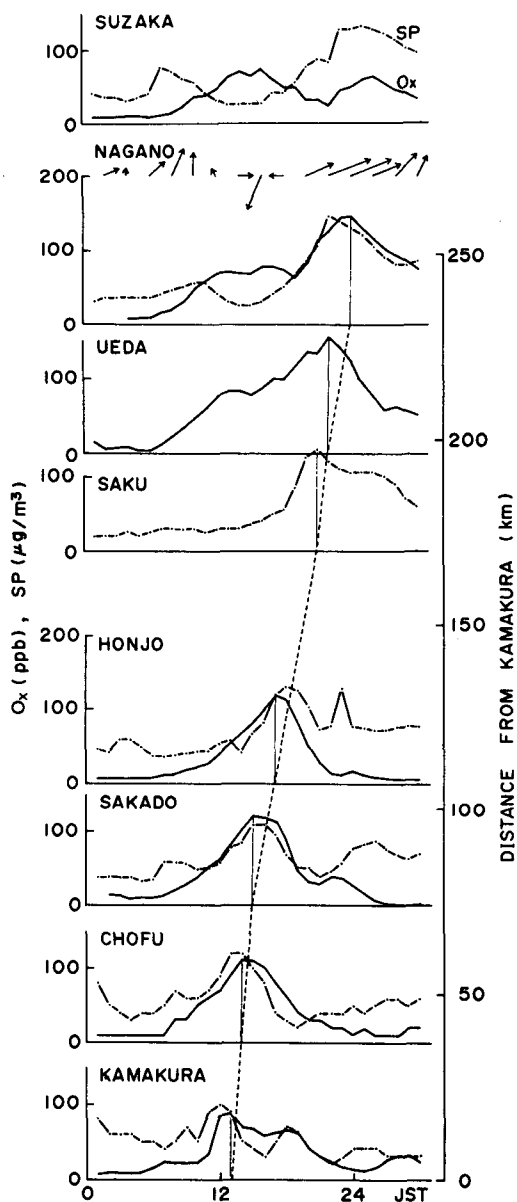


FIG. 7. Diurnal variations of the concentrations of oxidants and suspended particles at eight stations along the route of the polluted air mass on 15 June 1979.

The nighttime behavior of reactive pollutants such as those described above is very important in the study of air pollution problems, e.g., when considering the representativeness of atmospheric monitoring station data, or the forecasting of photochemical smog. The reactive pollutants contained in the atmosphere above the nocturnal inversion layer can be entrained when the mixed layer reaches that altitude the following morning; this results in a rapid increase of ground-level pollutant concentration (Blumenthal *et al.*, 1978; Wakamatsu *et al.*, 1983; Uno *et al.*, 1984). The

nighttime behavior of pollutants warrants further investigation.

6. Concluding remarks

Long-range transport of pollutants over hundreds of kilometers is well known in northern Europe and North America (e.g., Cox *et al.*, 1975), and is generally driven by gradient winds. In Japan, this type of air pollution was not believed to be serious. However, as atmospheric monitoring systems became fully equipped, seriously polluted areas were found more than 100 km inland from industrial and populated areas. A characteristic of the air pollution in these areas is that it occurs on clear summer days with a light gradient wind under a synoptic-scale high pressure system. In addition, it was found that such pollution is due to secondary pollutants, such as oxidants and suspended particles, and occurs frequently in the late evening. While this type of air pollution has not received much attention in Japan, such long-range transport is clearly a very important phenomenon in photochemical smog and acid rain problems.

In the present paper, distributions of surface wind and pressure over a 300×300 km area in the central part of Japan, together with the concentration isopleths of oxidants and suspended particles on all days with high oxidant concentrations at stations (Ueda and Nagano) far from large emission sources, were analyzed. As a result, one mechanism of long-range transport was determined.

The long-range transport discussed here occurs on clear summer days under a synoptic-scale high pressure system; the gradient wind is light but local winds such as land/sea breezes and slope and valley winds are intense. Under such conditions, the diurnal-mean land-sea temperature difference drives a steady onshore wind that is superimposed on the land/sea breezes and accentuates the net onshore wind in the daytime to form an extended sea breeze. If there is a mountainous region inland, the differential heating between the land at high altitude and the sea generates and sustains a significantly strong thermal low in this region. Moreover, a subsidence inversion accompanied by a synoptic-scale high pressure system limits the vertical extent of air flow and pollutant dispersion. These three mechanisms work to combine local winds, such as land/sea breezes and slope and valley winds, into one large-scale wind field that drives the long-range transport of pollutants. In contrast, in the nighttime, because of the diurnal-mean land-sea temperature difference, a thermal high pressure in the mountainous region does not occur or, if it does, it is very weak. Moreover, the steady onshore wind, in turn, opposes land breezes and results in weak offshore winds. This wind regime results in a situation where

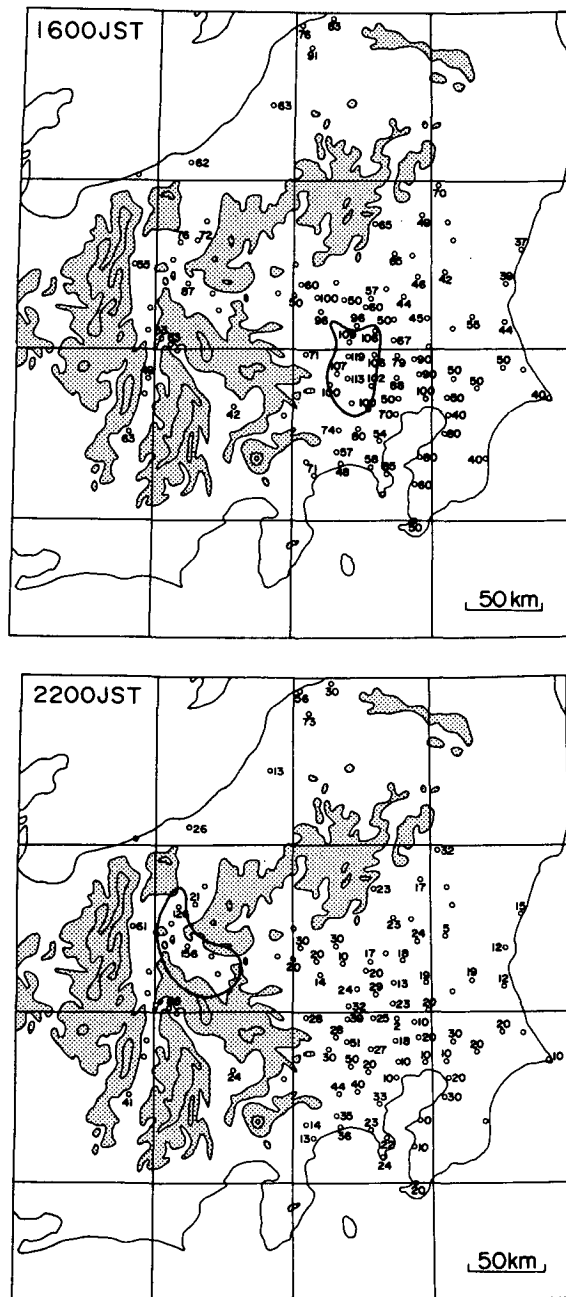


FIG. 8. Comparison of the oxidant concentration distributions between 1600 and 2200 on 15 June 1979. Numbers near solid circles indicate concentration (ppb).

an air mass from the industrial and populated areas in the coastal region is transported a long distance into the mountainous region during the daytime and is confined to that area overnight.

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