Measurements of Atmospheric Concentration of NOx and SO2 in the Sea Areas of Osaka Bay and Seto-Inlands Sea*

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Measurements of atmospheric concentration of NOx and SO2 were carried out by using the chemiluminescence method of NOx and the ultraviolet-fluorescence method of SO2, and by using the training ship Fukae-maru of Kobe University of Mercantile Marine in the sea areas of Osaka Bay and Seto-inlands Sea. Measurement results make clear the followings. (1) The exhausts from ships have obvious impacts on NOx and SO2 concentration in the narrow water areas and/or in the ports where marine traffic is congested. (2) The atmosphere of the north-east area of Osaka Bay is highly polluted in some weather conditions such that the wind due to large scale pressure systems is weak and the land-and-sea breeze is prevailing.

1. Introduction

Recently there have been published the reports1-3) which have investigated the impacts of pollutants' discharge from ships on the air pollution of some coastal regions surrounded by industrialized zone and/or ur-banized hinterland such as Tokyo Bay where marine traffic is highly congested. The authors have also estimated the discharge amount of NOx and SO2 from ships in the Osaka Bay area, and pointed out that those discharge is counted to be so large amount to be a match for the amount discharged from the land area around Osaka bay that some control measures should be con-sidered because the atmospheric environment over Osaka Bay area is still now deteriorating. In order to consider the control measure for the whole area of Osaka Bay including both land and sea areas, it is needed to know the actual situation of air pollution not only of land region but also of sea region. There could have been found, however, very little data of air pollution in the sea zone so far. So the authors carried out the measurements of atmospheric concentration of NOx and SO2 in the sea region of Osaka Bay, and also in the regions of Seto-inlands Sea and around Shikoku island by using the training ship Fukae-maru of Kobe Univer-sity of Mercantile Marine. The measurements results are reported here in this paper.

2. Measurement method

In Japan, the officially designated method, which are used at the air environment monitoring stations of governmental organization, are the Saltzman reagent method for NOx and the electric conductivity method for SO2. NOx is here defined by the sum of NO and NO2. These methods require many minutes for measure-ment, ordinarily 60 minutes for each measurement. They are not appropriate, therefore, for the on-board measurement in the sea area. So the chemiluminescence method, the type of APNE-350E made by the Horiba Instrument Company is used here for NOx and the ultraviolet-fluorescence method, the type of APSA-360 made by the same company mentioned above for SO2. By the use of these methods, the measurement time work can be finished within a few minutes for each measuring point.

In order to confirm the correctness of the method used here, the test measurements for the chemiluminescence method were carried out at an official monitoring station near the Kobe University of Mercantile Marine, and their results were compared with those obtained officially by the Saltzman reagent method at the same station. Fig. 1 shows the comparison of data of hourly average value obtained by continuous measurement of four days. As can be seen in the figure, the results agree well with each other, though, as for NO, there exists some difference.

The inlet of air sampling tube was fitted at the superstructure on the bow deck of the training ship "Fukae-maru" in order to avoid the exhaust gas flow discharged from her own stack.

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3. Measurement results at sea areas in the Seto-Inlands Sea and around Shikoku island

3.1 Results in open sea areas

Fig. 2 shows the measurement results on the sea route around Shikoku island starting from the authors' university at Kobe, going down south through Kii Channel, and finally going back through Seto-inlands Sea to Kobe. The bottom figure shows the NOx and SO2 data measured on the sea route going westward off the southside of Shikoku island, and the upper shows those data on the route going through Seto-inlands Sea from west to east. Fig. 3 shows the same data of Fig. 2 denoted by the circle symbols.

In Seto-inlands Sea, there are narrow channels such as Akashi-, Bisanseto-, Kurushima-Channel, where the marine traffic intensities are high due to their narrow waterways. Looking at the figures, both NOx and SO2 concentrations increase to relatively higher level in those narrow channels compared to open sea areas. It is considered therefore that the NOx and SO2 pollutants discharged from the ships going through those narrow channels give some effects on the atmospheric pollution. In open sea areas outside of those narrow channels, air pollution level is not so high, and the NOx and SO2 concentrations are in the level lower than 20-30 ppb and 5-10 ppb, respectively. As seen clearly in Fig. 3, the air pollution level decreases gradually as the route goes down southward from Kii Channel, and very low concentration was measured in the open sea southside Shikoku island, especially in the region between Muroto- and Ashizuri-Promontory. There can be found a little increase of concentration in the region near Ashizuri-Promontory, which may be due to gathering of ships in a narrow route. In Bungo Channel, the pollutants concentration increased again as the route goes up to Seto-inlands Sea.

According to the measurement results on the route around Shikoku island mentioned above, it could be said that the ship traffic, especially in narrow channels, has some effect on the air pollution. The pollution level in Seto-inlands Sea area is higher compared to that in the open sea southside Shikoku island. The pollution of Seto-inlands Sea is considered, however, to be induced not only by marine traffic but also by the pollutants discharge from the industrialized and urbanized coastal zone.

3.2 Results in harbor zones

Fig. 4 shows the results measured in some harbors during being at anchor. The data shown in the figure are the averaged values of every 30 minutes. The weather when the measurement had carried out was strongly windy at Fukae harbor, and was clear and calm at wind velocity of 0-4 m/s both at Takamatsu and Beppu.
The circumferential conditions concerning air pollution were as follows for each harbor. Fukae harbor has a light traffic, but is situated in highly industrialized zone and neighboring to two large highway's having very heavy automobile traffic, so that the atmosphere is polluted severely. Beppu harbor has a moderate traffic and is situated in a unindustrialized city of medium size. Among three harbors, marine traffic is heaviest in Takamatsu harbor where a large number of ferries and high speed crafts are coming and going continuously for 24 hours, and land traffic of heavy trucks and cars is also heavy. As can be seen in the middle of Fig. 4, both of NOx and SO2 concentration are very high for all day long. In Takamatsu, NO concentration, which is the difference between NOx and NO2 concentration in the figure, is relatively high compared to NO2. This high ratio of NO concentration indicates that NOx discharged from ships contributes directly to the air pollution of Takamatsu harbor zone. Anyhow, the pollution level of Takamatsu is so high that it would be better to investigate how the pollutant discharge is diffusing over the neighboring city area. Contrary to Takamatsu, the concentration ratio of NO2 to NO is high in Beppu and Fukae, so that the impact of marine traffic can be considered to be not so significant as Takamatsu.

4. NOx and SO2 concentration in Osaka bay

4.1 Measurement results in Osaka Bay

The data of NOx concentration in Osaka Bay measured by the training ship Fukae-maru are shown in Fig. 5, where the wind vectors measured simultaneously are shown together. In addition, the NOx data in land region measured by the official monitoring stations are also plotted. The land data are hourly averaged concentration, and sea data are momentarily measured concentration at each point in the figure. The arrow lines indicate the cruising route of Fukae-maru, and the number attached to each point shows the measurement time. As for the land data, the concentration at the time of 8:00, 9:00, 10:00 are plotted in the upper, middle, bottom figure, respectively.

The followings can be read out from Fig. 5. That is,

(1) In the morning until nine o'clock or so, the concentra-


(1) The concentration in the northern sea area was so high as same level with that of coastal zone. The coastal zone of the depth, that is, northeast region of Osaka Bay is well known as one of terribly polluted areas in Japan. So the high concentration data measured in the northern sea area should be noticed seriously.

(2) In the afternoon, however, the concentration went down to lower level compared to the morning.

(3) The concentration in the southern sea region was at rather low level at any time in the morning or afternoon.

(4) As for the wind condition, the wind in the northern sea area was calm and/or north wind in the mom-ing before 9 o'clock. After the time, however, the wind changed to sea breeze towards the land.

(5) In coastal zone, the sea breeze has already developed almost all around the bay at 8 o'clock, and the sea breeze grew further as the time went by 9, 10 o'clock and so on.

Fig. 6 shows the measurement results of NOx and SO2 concentration on a circulating route in Osaka Bay. The upper figures show the results when the measurements were carried out at November 22nd 1995 in the anti-clockwise route, and the lower at December 14th 1995 in the clockwise route. The data in the land region measured by official monitoring at 10 o'clock have been also plotted in the figures. It could be read out generally same characteristics from Fig. 6 with those from Fig. 5 mentioned above. That is, in the morning, NOx and also SO2 concentration in northern sea area were very high at the almost same level with those of coastal zone where the atmosphere is polluted severely as already pointed out. But, in the afternoon, those concentration had went down to the low level.

The measurement results mentioned above suggest that the wind system formed by sea- and landbreeze gives a great influence on the characteristics of spatial and temporal distribution of atmospheric pollution in Osaka Bay area. Comparing the wind data of Fig. 6 to that of Fig. 5, the sea breeze in winter season of Fig. 6 does not fully develop still at 10 o'clock, while it has developed fully already at 8 o'clock in June of Fig. 5. That is, the change from land- to sea-breeze occurs later in winter compared to summer. Further the strongness of breeze is weak in winter compared to summer.

4.2 Concentration characteristics of Akashi Channel and marine traffic

There are not situated any noticeable pollutant sources in the coastal zone of Akashi Channel, though rather high concentration of NOx and SO2 was observed.

Fig. 5  Comparison of NOx measurement result in Osaka Bay with NOx Concentration at monitoring stations in land area
in the zone as shown in Fig. 5. How is the influence of marine traffic on the coastal air pollution? Fig. 7 shows the NOx and SO2 discharge amounts from the marine traffic passing Akashi Channel which have been calculated by the same manner with the estimation procedure4) and by the use of traffic data6) which were measured in the days from 15th to 18th October 1990. The distribution of discharge amounts shown in Fig. 7 is roughly corresponding to the traffic distribution. The traffic entering Osaka Bay towards east has a peak in the early morning, and, on the other hand, the traffic going out Osaka Bay towards west is more intensive during the time from afternoon to night. The estimation results indicate that the annual discharge density per unit area can be accounted to be about 60 t/km$^2$.y for NOx and 30 t/km$^2$.y for SO2. According to the investigation carried out by the Hyogo prefectural officials7), the discharge density in the coastal zone of Akashi Channel has been estimated to be less than 10 t/km$^2$.y for NOx and further less for SO2. The pollutant discharge density from marine traffic is much greater than that from artificial activities in the land area. So the effect of marine traffic is considered not to be able to neglect for the air pollution in the coastal zone of Akashi Channel.

Fig. 8 shows an typical example of the relation between pollutants concentration and wind vector variation. The data shown in the figure are the data of Akashi monitoring station in a week from first to seventh June 1994, when the weather is calm and the wind system is
mainly dominated by sea- and land-breeze. Looking at Fig. 8, the concentrations both of NOX and SO2, and also wind vector are varying periodically with a day cycle. It can be read out further that the concentration increases synchronously with the change of wind direction from land to sea breeze. This means that the discharge from marine traffic gives some effects on the air pollution in the coastal zone of Akashi Channel.

4.3 Present situation of air pollution in Osaka Bay area

The measurement results of NOX and SO2 in Osaka Bay have been shown and discussed above considering the relation with the data of monitoring stations in the land area around Osaka Bay. The following points could be pointed out.

1. In the morning, the atmosphere in the northeastern sea area of Osaka Bay bordered by a line linking Akashi Channel and Izumiotsu is significantly polluted to the almost same level with that of the coastal region, where is one of terribly polluted regions in Japan.

2. The concentration level in the above sea area, however, changes greatly with time, and, in the afternoon, decreases to a lower level than that of coastal region. This change is greatly influenced by the cyclic wind system of sea- and land-breeze formed in Osaka Bay area.

3. In the northeastern area of Osaka Bay, it is known that pollutants discharge is highly intensive from...
coastal regions, though the discharge from the marine traffic is also intensive. Considering the air pollution characteristics shown in Fig. 8, it could be considered the air pollution in the northeastern sea area of Osaka Bay is influenced to some extent by the discharge from marine traffic.

(4) But it could not be said simply that the discharge from marine traffic is a dominant cause of the significant pollution in the northeastern sea area of Osaka bay. Because there is a possibility that the pollutants discharged from the land region flows into the atmosphere over Osaka Bay and is accumulated there due to the land breeze during nighttime.

(5) In Osaka Bay area, there often occurs the weather condition that the wind due to large scale pressure system is weak and the wind system is mainly determined by the cyclic wind system formed by sea- and land-breeze. It has been known that the air pollution in land area grows to serious level in such weather condition. It has been revealed here, however, that not only the land area but also the sea area is polluted significantly in such weather condition as shown in Figs. 5 and 6. It would be a very important task, therefore, to analyze the pollution mechanism taking pollutants discharge both from land and sea areas into consideration when the weather condition mentioned above is formed in order to investigate the effect of marine traffic on the air pollution in Osaka Bay area.

5. Conclusions

Measurement results of NOx and SO2 concentration in the atmosphere over the sea areas in Seto-inlands Sea and Osaka Bay, and around Shikoku island have been reported here. What have been revealed can be summarized as follows.

(1) The effect of discharge from marine traffic on the atmospheric pollution was observed clearly in some water channels where marine traffic is congested.

(2) The atmospheric pollution in Seto-inlands Sea area has been proceeded to a certain worse level in comparison with that in the southside sea area of Shikoku island.

(3) The pollutants concentration in harbor zone was at a high level when ship traffic was intensive such as Takamatsu harbor.

(4) As for the atmospheric pollution in Osaka Bay, it is noticeable that a serious polluted zone formed over the northeastern sea area when the weather is calm and the wind system is dominated by the sea-and land-breeze.

(5) In this sea area, the pollutants discharge from marine traffic is very intensive. As well known, however, the coastal region of the northeastern Osaka Bay has been highly industrialized and urbanized, so that the pollutants discharge from the land region is also very intensive. On the other hand, the characteristics of atmospheric pollution are greatly dependent on weather condition, especially on the cyclic wind system of sea- and land-breeze in Osaka Bay area. In order to investigate the effect of pollutants discharge from marine traffic in Osaka Bay, therefore, it would be an important task to develop a new simulation method which can make possible to analyze the all effects of pollutants discharge both from sea and land areas, and weather conditions in a integrated manner.

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Discussion

Prof. TAMAKI (SHINSHUU UNIVERSITY, FAC-ULTY of ENGINEERING)

(1) The Ship Research Institute has used the laser-doppler and radar-transmitting method (LR method) for measuring the NOx concentration in sea area. Could you tell me the difference between it and the Saltzman method used here by the authors?

(2) How many points were measured in sea area?

(3) How was the measurement height in sea area?

Author’s reply

(1) I am sorry, I have little knowledge about the measurement principle, of LR method. I think, however that the SRI has used the LR method for measuring the diffusion process of a exhaust gas plume from a stack of ship. So its measurement object was quite different from that of authors.

(2) The measurement instruments can measure the concentration within a few minutes, so the data were obtained here almost continuously along a route.

(3) The height was about five meters.
References

2) MESJ, Report on the investigation of control techniques against air pollutants discharge from ships, issued at 1993 and 1994.
3) Japan Association of Shipwreck Prevention, Report on research on air pollution in Japanese coastal zone due to marine traffic, issue at 1993.