

Development of Monitoring Techniques  
for Global Background Air Pollution

by

MRI Special Research Group  
on Global Atmospheric Pollution\*

Abstract

WMO decided in 1970 [Resolution 12 (EC-XXII)(Oct 1970)] and in 1971 [Resolution 10 (Cg-VI)(May 1971)] to launch a project aiming at the establishment of a global network of baseline air pollution stations designed to document longterm changes in atmospheric compositions of particular significance to weather and climate. Following to these resolutions, JMA decided to establish a baseline air pollution station at an appropriate site, which will become operational in 1981. The research group of MRI shares the responsibility of the developing feasible monitoring systems to monitor the atmospheric level of gaseous and particulate background pollutants at a baseline station. It is a basic requirement that the monitoring methods must be sufficiently sensitive, specific and reliable for the very low background concentration and be automated for a routine work at remote places.

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\* Principal Investigator:

H. Saito (Formerly Physical Meteorological Division)

Co-Investigators:

Gaseous pollutants:

K. Kawamura, K. Fushimi (Geochemical Division)

Particulate pollutants:

T. Ito, A. Ono (Physical Meteorology Division)

Aerosol-Sonde: M. Misaki, I. Kanazawa, M. Ikegami  
(Upper Atmosphere Physics Division)

Neutron activation analysis: N. Yano, I. Yamaji,

K. Maebashi (Applied Meteorology Division)

Atmospheric turbidity: K. Murai, M. Kobayashi, R. Goto,

T. Yamauchi (Upper Atmosphere Physics Division)

During the period from 1973 to 1976, considerable efforts at MRI have been extended in the research on monitoring methods, the development of prototype monitoring system and the observations on Minamitori-shima and Chichi-jima for site selection as described in this report.

#### 1. Measurement of gaseous pollutants

Automated instruments which provide measurements of extremely low concentration of  $\text{NO}_x$  and  $\text{SO}_2$  in the air at a rate of once per 6 hours have been developed.

The procedure for the measurement of  $\text{NO}_x$  is fundamentally based on the method which uses a mixed solution of sodium hydroxide and sodium arsenite as an absorption solution. The detection limit of the instrument in terms of concentration is about  $0.05\mu\text{g}/\text{m}^3$  for  $\text{NO}_2$  and  $0.07\mu\text{g}/\text{m}^3$  for  $\text{NO}$ . A series of observation to evaluate the nitrite ion equivalence of  $\text{NO}_2$  gas absorbed in the above mentioned mixed solution has been done at Minamitori-shima. The results of observation showed that the equivalence was 0.88 on the average. As for the measurement of  $\text{SO}_2$ , the procedure is a modification of the well known West-Gaeke method and the detection limit of the instrument is about  $0.06\mu\text{g}/\text{m}^3$ .

An automatic CO analyzer has been developed. The principle of measurement is based on the reaction of CO with mercury oxide at a temperature of  $200^\circ\text{C}$ . The detection limit of the analyzer is about 3ppb, and the standard deviation computed from the results obtained in our laboratory is about 3.2% at the CO concentration of 0.11ppm.

#### 2. Development of automatic counter for aerosol concentration measurement

The MRI type automatic Pollak Counter of high reliability has been improved further in the part of photo-detector so as it can

detect small variation precisely in the very low concentration of aerosol particles, say, below 1000 particles per  $\text{cm}^3$ . The improved photo-detector includes a photo-electric current balancing circuit and a motor driven optical filter system over the photocell. The former increases the sensitivity of the measurement, while the latter serves as a reference for the percentage extinction of photo-electric current.

In order to ensure the accuracy of the concentration measurement, a new model of calibration system was built up. It consists of the MRI type Aitken Counter, exploited in 1974, and a polaroid camera with an automatically operating control device. With the instrumentation an absolute value of aerosol concentration is obtained within a few minutes.

### 3. Development of aerosol-sonde

Two kinds of radiosonde for measuring the concentration of aerosol particles have been developed. The first one was designed for the large particles, with the diameter more than  $0.3 \mu\text{m}$ , with the photo-electric sensing technique which detects and counts the pulse of light scattered by the individual particle in the sample air. The second type of sonde was for the small particles, with the diameter less than  $0.3 \mu\text{m}$ , with the conventional technique utilizing the fog chamber method.

### 4. Development of automatic aerosol samplers and analytical method

Three types of aerosol samplers were developed in order to make a sampling in unpolluted atmosphere. Two of them are the samplers which can collect the size separated samples for instrumental neutron activation analysis, and the other is the sampler for electronmicroscopic analysis.

Main special features of the samplers are; (1) Accurate flow rate measurement, (2) Check of self instrumental contamination and

(3) Automatic operation of the samplers. Principle and analytical procedure of the instrumental neutron activation analysis are described in this report.

#### 5. Measurement of atmospheric turbidity

A spectro-pyrheliometer and a spectro-pyranometer were designed for the monitoring of the atmospheric turbidity due to aerosol particles. The direct solar radiation is measured by the spectro-pyrheliometer which is set on the equatorial mounting and we get the spectral distributions of extinction coefficient due to the scattering and absorption by aerosol particles. From the spectral distributions of extinction coefficient, we can infer the size distributions of particles. By using the spectro-pyranometer, the spectral distribution of global and diffuse sky radiation are measured, and the analysis of data leads to determination of the optical properties of the aerosols.

#### 6. Data collection for site selection

The present atmospheric level of a background air pollution was measured on Minamitori-shima (24°18' N; 153°58' E) and Chichi-jima (27°05' N; 142°11' E) which are the proposed sites for establishment of a baseline air pollution station in Japan. The expeditions were made in summer and autumn in 1975, and in autumn in 1976. The main results of the observations are described as follows.

The concentrations of gaseous pollutants in the atmosphere were measured at Mt. Norikura (36°06' N; 137°33' E, elevation 2770m), Minamitori-shima and Chichi-jima.

The observations of NO<sub>2</sub>, NO, CO and O<sub>3</sub> in the air at Mt. Norikura were carried out in October, 1976, and their average values were 1.8 μg/m<sup>3</sup>, 0.8 μg/m<sup>3</sup>, 0.13 ppm and 0.039 ppm, respectively. The average values of NO<sub>2</sub>, NO, SO<sub>2</sub> and CO in the

air observed at Minamitori-shima were  $1.2 \mu\text{g}/\text{m}^3$ ,  $0.6 \mu\text{g}/\text{m}^3$ ,  $0.7 \mu\text{g}/\text{m}^3$  and  $0.15 \text{ ppm}$ , and those of  $\text{SO}_2$ ,  $\text{CO}$  and  $\text{O}_3$  in the air measured at Chichi-jima were  $0.6 \mu\text{g}/\text{m}^3$ ,  $0.08 \text{ ppm}$  and  $0.024 \text{ ppm}$ , respectively. Because of scanty data, it is not yet known about the concentration of  $\text{NO}$  in the maritime air. The concentration of  $\text{NO}$  observed at Minamitori-shima is two times larger than that assumed to exist over all other areas except land.

Our results on the atmospheric background concentrations of gaseous components, particularly  $\text{NO}_2$  and  $\text{SO}_2$  observed at Minamitori-shima and Chichi-jima agree well with those obtained by the other researchers.

The concentrations of Aitken particles and the large particles (with diameters more than  $0.3 \mu\text{m}$ ) were continuously recorded for 2 weeks in summer and 2 weeks in late fall of 1975 in Minamitori-shima.

In summer season, the concentration of Aitken particles was found to be steadily in low value of 200-300 particles/cc. In fall season the concentration was mostly low, being under the influence of maritime air mass as in summer, but occasionally enhanced up to 2000 particles/cc when the island was reached by polluted air mass which came from the main land of Japan. The same situation was also found again but more markedly in case of the observation in Chichi-jima carried out for two weeks in fall 1976.

The time variation of the large particles was rather complicated even in Minamitori-shima, sometimes being related more or less with temperature and humidity, and sometimes with the wind direction. The average concentration of the large particles was determined as 20 particles/cc, which was about two times higher than that in Minamitori-shima.

The surface distribution of the aerosol concentration was surveyed in both islands, Minamitori-shima and Chichi-jima, in 1975 and in 1976, respectively, for the purpose of the site

selection, with the particular caution for the particles produced from the anthropogenic origin and from sea spray raised at the coral reef.

Minamitori-shima has the extremely simple topography, shaped as a triangle with about 2000 m sides, having a flat surface of the highest elevation of 8 m, and surrounded by the coral reef. There is no habitant except the members of a weather station of JMA and U.S. Coast Guard. High concentration of the small particles was found only in the plumes of the effluents from the stacks of the electric power stations. On the other hand, no increase in the concentration of the large particles was detected in the effluents. The highest concentration of the large particles was found along the coast line of the windside, decaying with the distance towards the leeside. This apparently indicated that those particles were produced from sea spray at the coral reef. Even under the normal weather condition, it was found that such unfavorable situation was prevailing throughout the small, flat, island.

The topographical condition of Chichi-jima is quite complicated, mostly occupied by mountains of about 300 m elevation, partly populated in the sea shore area. The result of the survey revealed that there was no particular area polluted with the persistent high concentration of aerosols. Anthropogenic effects on the aerosol concentration was not remarkable even in the most densely populated area in the island. The particular pollutants, which were released into the atmosphere by man's activities seemed to be scattered immediately, and caused no significant enhancement of the back ground level of concentration.

Atmospheric aerosols have been sampled at Chichi-jima and Minamitori-shima. The mean concentration of total suspended particles at Chichi-jima was  $16.4 \mu\text{g}/\text{m}^3$ . Also the concentrations of the trace elements and their mass-size distributions

were determined in the samples.

Enrichment factor was defined in order to classify the main sources of elements. The sources are sea water(group 1), crust (group 2), and man-made and so on(group 3).

Na, Br, Cl, Mg, K were classified into the group 1. Mass-size distributions and their modes were  $4\ \mu\text{m}$  in diameter. But the data of Minamitori-shima was different from that of Chichi-jima. The reason was explained by giant sea spray particles.

Cs, V, Co, Mn, Cr, Sc, Fe, Al, Ce and Sm were classified into the group 2. The size distributions of the elements were similar to that of group 1 and their modes were  $5\ \mu\text{m}$  in diameter.

I, Zn, Se, Sb were classified into the group 3. The size distributions show that the most parts of the mass distributed in submicron particles except Iodine.

Spectral values of the extinction coefficient of the aerosol particles contained in vertical column of the atmosphere at Minamitori-shima were determined from spectral measurements of direct solar radiation in 1975. These values are smaller by about one order than the maximum value obtained in Tokyo. In general, the discrepancies of the values are larger in shorter wavelength region than in the longer.

By using the inversion technique, we inferred the size distributions of aerosols based on the measured extinction coefficients. Comparisons of the size distributions between Minamitori-shima and Tokyo show that the amount of particles in Minamitori-shima is smaller than in Tokyo, especially in the region of particle radius smaller than about  $1.0\ \mu\text{m}$ .

#### Conclusion on the site selection

Considering the result of the preliminary observations on the gaseous and particular pollutants in Minamitori-shima and Chichi-jima, proposed islands as the site of Japanese baseline atmospheric

pollution station (BAPS), it is concluded as follows.

(i) Minamitori-shima

Minamitori-shima was raised first as the most favorable site for BAPS because it is remotest from Japan Proper Island, and also because the island is almost free from the origin of the anthropogenic pollution. The result of the preliminary investigation, however, showed that the influence of sea spray raised along the periphery of the island could not be disregarded for the accuracy of the measurements and for the maintenance of the instruments of high precision.

(ii) Chichi-jima

According to the observational result on the surface distribution of aerosol concentration and gaseous pollutants in the island, there was no particular area where the pollution was remarkable. Although the situation seems to be favorable for the time being, the decision on the site selection must be done taking into consideration the fact that anthropogenic activities have been increasing in this island.



