Atmospheric Turbid Conditions due to Fine Particles in Recent Years at Nagasaki, Japan

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Abstract: Atmospheric turbid conditions caused by fine particles, which are defined as the particles in the size range between 0.3 and 1.0 μ m in diameter, are occasionally significant in recent years over the Nagasaki area in Japan. These conditions make the horizontal visibility very low as 4–5 km despite of fair weather. We studied two significantly turbid cases rich with fine particles, which took place during 25–27 March 2003 and on 23 May 2005, from the viewpoint of a detailed understanding of their influences to visibility and the properties of fine aerosols. As a result of this study, the noticeably low visibility conditions due to fine particles are closely connected with the high concentration of sulfur which transported from the Asian continent. Fine particles sometimes make very turbid conditions in spring without the influence of yellow sand dust particles. This peculiarity should be paid further attention from the viewpoint of air quality conservation over East Asia.

Key words: atmospheric turbid condition, low visibility, fine particles, sulfate aerosols

1. Introduction

The problems of atmospheric aerosol environment and its related issues are very important on the viewpoint of the global warming due to the radiative forcing of atmospheric constituents. One of the most interesting and also serious subjects in these problems is the modification of air quality caused by atmospheric aerosols such as anthropogenic aerosols (sulfate aerosols, nitrate aerosols and so on) and mineral dust (yellow sand dust ; Kosa). We have made an investigation into the physical and chemical properties of atmospheric aerosols in Western Kyushu, Japan for recent four years 2002–2005 (Han et al., 2004;

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Received 6 June 2006 Accepted 14 September 2006 Shimizu et. al., 2004). In the process of this study, we sometimes found significantly turbid conditions showing high concentration of fine particles and diminishing the horizontal visibility so markedly. The fine particles described in this paper is defined as particles whose sizes are in the range of $0.3 - 1.0\mu$ m in diameter. These fine particles are mainly anthropogenic aerosols, but not the component of yellow sand particles, because yellow sand dust are mostly larger than 1.0µm in diameter (e.g., Arao et al., 2006).

Our main interest in this paper is in the properties of such small particles. A general feature of the aerosol properties in a Kosa event over East Asia has been reported by Uematsu et al. (2002), in which fine particles in the size range of $0.5-1.0\mu$ m in diameter corresponded to the number concentration of anthropogenic aerosols such as sulfur-rich particles. Takami et al. (2005) also showed the results of detailed analysis on the composition of fine particulate matters in a spring season at this region in Japan. Along the viewpoint mentioned above, we have collaborated with an observational study on the problem of atmospheric environment concerning the properties of fine particles and anthropogenic aerosols.

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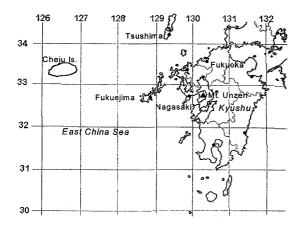


Fig. 1. Map of Kyushu district with our observational sites

2. Meteorological instruments and observations

Our investigation about the physical and chemical properties of atmospheric aerosols has been carried out using several instruments such as optical particle counter (OPC), Aerodyne aerosol mass spectrometer (AMS), mini step sampler, β -ray mass densitometer (SPM) and some basic equipments. Together with our original observational data, we utilized several kinds of meteorological data provided by Japan Meteorological Agency (JMA) and other public organizations.

The most basic data in this study were those obtained by OPC (Rion KC-01) of Research Institute for Humanity and Nature (RIHN) and Nagasaki University. The OPC used in this study measures the number concentrations of aerosol particles in every 10 minutes dividing them into five size ranges, whose size criteria are 0.3, 0.5, 1.0, 2.0 and 5.0µm in diameter. Hereafter the particle size is denoted in diameter throughout this paper. We shall concentrate our investigation into the physical and optical properties of fine particles ($0.3 - 1.0\mu$ m). The particles greater than 1µm will be contrarily denoted as large particles or coarse particles.

Fig. 1 shows the geographical map of our observational sites. One of our OPC was at Fukuejima (Fukue Island; 80m msl) where is about 100 km west from Nagasaki City. Another two were set up at Nagasaki University (32.78° N, 129.87° E; 20m) and Mt. Unzen Ropeway Summit Station (1290m), respectively. Mt. Unzen is located at a distance of 40km east from Nagasaki City. The location of Nagasaki Marine Observatory (NMO) belonging to Japan Meteorological Agency (JMA) is

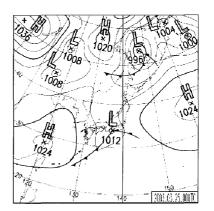


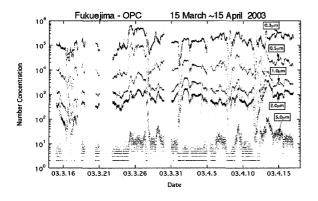
Fig. 2. Surface weather map at 09JST on 25 March 2003 (JMA & JMBMC, 2003)

only 5 km south of Nagasaki University. In Fig. 1, Cheju Island in Korea can be seen at its left hand side. The distance between Cheju Island and Nagasaki is about 350km, and the distance from Nagasaki to the nearest coast of the Asian continent (China) is about 750km.

Condition rich with fine particles during 25– 27 March 2003

Fig.2 shows the surface weather chart at 09JST on 25 March 2003. A high pressure system can be seen over the coastal region of China. The center of this anticyclone moved eastwards slowly with 600 km/day at the point of 09JST 26 March, while it moved faster with more than 1000 km/day at 09JST 27. During 25—27 May, a large number of JMA meteorological stations reported yellow sand dust events (Kosa; Asian dust), probably because of the reduction of horizontal visibility.

Fig. 3 shows the OPC time series at Fukuejima during 15 March to 15 April 2003. A marked peak of the total number concentration of aerosol particles, which defined as all the particles larger than 0.3μ m (the highest curve among five ones), appeared on 25-27 March. It can be seen from Fig. 3 that the situation of high concentrations of total particles during 25-27 March was very significant compared with the other peaks in the period shown in this figure. One of important features of this figure is that the total concentration of $6-7\times10^5$ L⁻¹ (L:liter) is almost one order larger than usual atmospheric concentrations. The logarithmic presentation of number concentrations in



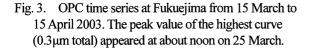


Fig.3 and the next figures also indicates that the concentration of total particles is ordinarily almost equal to that of fine particles $(0.3 - 1.0 \mu m)$, because the number concentration of fine particles occupies nearly 99% of the total concentration in usual excepting yellow dust events.

Fig. 4 and Fig. 5 also show the OPC time series during 25-27 March 2003 at Nagasaki University and at Mt. Unzen, respectively. From these figures, we can see that the maximum number concentration of total aerosol particles were $6-7 \times 10^5$ L⁻¹, almost the same level as in Fig.3. These high concentrations are so unusual that we would like to call simply this feature "atmospheric condition rich with fine particles". This of fine particle-rich condition kind can be contradistinguished from the condition rich with coarse particles (>1um), which is occasionally observed in an Asian dust event in spring. As seen from Fig. 4 and Fig. 5, the atmospheric condition markedly rich with fine particles continued for more than 48 hours and the coarse particles were rather low concentration during this period except at midnight on 27 March. Careful comparison between these two figures and Fig. 3 indicates that there are very similar trends in both fine particles and coarse particles during 25-27 March between the three observational sites, with their temporal coincidence of the peak of fine particles at about noon on 25 March. In addition, a remarkable decrease in the concentrations of fine particles can be seen in the afternoon on 27 March, because of rainy weather associated with passage of a cold front.

In relation to these OPC quantities at Nagasaki University (Fig. 4), the suspended particulate matter (SPM) measured by a β -ray mass densitometer of

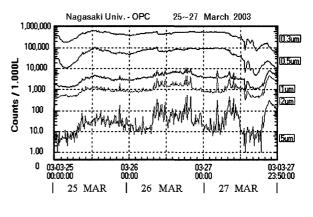


Fig. 4. OPC time series at Nagasaki University during 25–27 March 2003.

National Institute for Environmental Studies (NIES) settled at Nagasaki University, and showed its maximum density of $113\mu gm^{-3}$ at 12JST on 25 March, and the second place $105\mu gm^{-3}$ at 14JST 25, the third $104\mu gm^{-3}$ at 15JST 26. These high densities in SPM were also rare compared with usual atmospheric conditions ($30\mu gm^{-3}$) excepting large-scale Kosa events.

Fig. 6 shows the relationship between the OPC number concentrations at Nagasaki University and the horizontal visibility at Nagasaki Marine Observatory during 25-27 March 2003. The horizontal axis shows the concentration of coarse particles larger than 5µm, and the vertical one indicates that of fine particles in a size range between 0.3µm and 0.5µm. The horizontal visibility has been routinely observed in every three hours except for the midnight time (00JST) at NMO, so that the OPC number concentrations in this figure were picked up from those measured at the same time as the visibility observation. Since the concentrations of fine particles were as high as $4 \times 10^5 L^{-1}$ or more as described above, all the visibility data scatter in the upper horizontal frames of the figure. On the contrary, the concentrations of coarse particles varied in their wide range from the lowest level of $5L^{-1}$ to the highest of 347 L^{-1} . We can see from this figure that the low visibilities less than 10km had taken place in the conditions rich with fine particles, whose concentrations were larger than $5 \times 10^{5} \text{L}^{-1}$. It should be then noticed that the low visibility can be observed even in the conditions of lacking in coarse particles, provided that fine particles exist abundantly. Of course, the most serious reduction of visibility will occur under the condition in which both fine and coarse particles come out at the same

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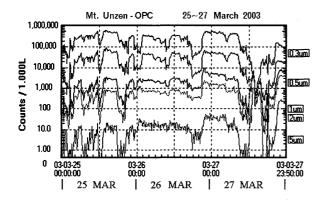


Fig. 5. OPC time series at Mt. Unzen (1290m) during 25 -27 March 2003.

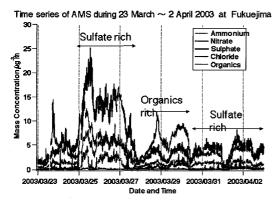


Fig. 7. AMS time series at Fukuejima during 23-31 March 2003.

time. During the three days in Fig. 6, NMO recorded the same daily minimum visibility of 4.0km at 12JST on 25, 12JST 26, and 03JST 27, when the concentration of fine particles was $6.6 \times 10^5 \text{L}^{-1}$, $4.7 \times 10^5 \text{L}^{-1}$ and $5.9 \times 10^5 \text{L}^{-1}$, respectively. These circumstances are evidently indicating extremely turbid atmospheric conditions which were filled with fine particles.

With respect to the related issues on the optics of aerosols over the Nagasaki region, Shimizu et al. (2004) and Sugimoto et al. (2005) have reported the general characteristics of Asian dust particles, from which we can obtain the more extended knowledge concerning the lidar and sky radiometer observations. Furthermore, Aoki (2003) and Sano et al. (2003) have presented the aerosol optical properties at Fukuejima on 26 March 2003 (almost fair during the daytime) and showed that the aerosol optical thickness at a wavelength of 0.5 μ m was 0.6–0.7, and Ångström exponent was 1.0–1.1. These values can be regarded

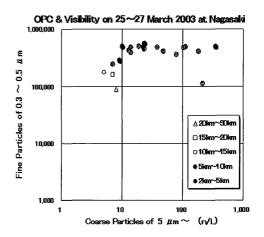


Fig. 6. Relationship between the OPC number concentrations and horizontal visibility at Nagasaki during 25–27 March 2003.

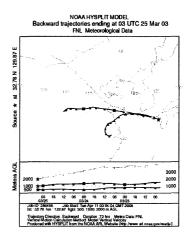


Fig. 8. NOAA HYSPLIT backward trajectories at the location of Nagasaki University ending at 12JST on 25 March 2003 for three elevations of 500m, 1000m and 2000m.

as typical values in turbid conditions rich with fine particles.

Fig. 7 indicates the time series of Aerodyne aerosol mass spectrometer (AMS) at Fukuejima during 23-31 March 2003. This AMS instrument, which was belonging to NIES, primarily measures the fine particle mode (0.1–1µm) and is not able to detect coarse particles. Fig. 7 shows that the mass concentration of sulfate during 25–27 March is significantly higher (15µgm⁻³ or more) than those of the other days. In addition, it should be noticed that the pattern of change in sulfate on 25–27 May is very similar to the time series of the OPC fine particles in Fig. 3. In other words, we can likely state that the amount of sulfate is fairly

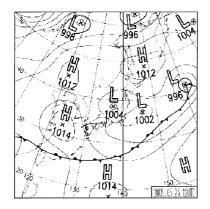


Fig. 9. Surface weather map at 09JST on 23 May 2005 (JMA & JMBMC, 2005)

proportional to the concentration of the OPC fine particles. This linkage between the temporal variations of sulfate and fine particles is very interesting, as will be also described in the following sections. The details of the AMS measurements at Fukuejima are already reported by Takami et al. (2005).

Fig. 8 indicates an example for NOAA HYSPLIT backward trajectories at the location of Nagasaki University ending at 12JST on 25 March for three elevations of 500, 1000 and 2000m and for the duration of 72hrs (NOAA, 2003). Fukuejima and Nagasaki both had high concentrations of fine particles at 12JST 25 May, as be seen from the previous figures. These backward trajectories clearly show that the atmosphere over the Chinese industrial region influenced to the Western Kyushu area in Japan. Especially, the air parcels at the two lower levels of 500m and 1000m were brought from China almost keeping the same elevations. The main source of fine particles can then be pointed out the Asian continent. In connection with Fig. 8, a HYSPLIT backward trajectory at Fukuejima ending at 14JST 25 March for the elevation of 500m was presented by Takami et al. (2005), from which we can also confirm a stream of polluted air from the continent.

4. Condition rich with fine particles on 23 May 2005

Fig. 9 shows the surface weather chart at 09JST on 23 May 2005. A high pressure region was located over the East China Sea, so that the situation of high pressure system was quite similar to the previous case (Fig. 2).

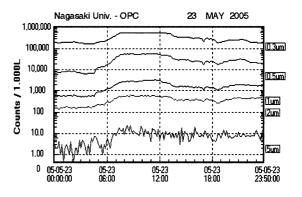


Fig. 10. OPC time series at Nagasaki University on 23 May 2005.

Fig. 10 and Fig. 11 show the OPC time series on 23 May 2005 at Nagasaki University and at Mt. Unzen, respectively. From these figures, we can easily see that the maximum number concentration of total aerosol particles were $5 - 6 \times 10^5 \text{L}^{-1}$ with rather low concentrations of large particles. This fine particle-rich condition both at Nagasaki and Mt. Unzen lasted for about half a day, while the fine particles at Mt. Unzen at the early morning was considerably lower than at Nagasaki University.

Fig. 12 shows the time series of the suspended particulate matter (SPM) obtained by a β -ray mass densitometer of NIES at Nagasaki University on 23 May 2005. The SPM mass concentration has recorded its maximum of 131µgm⁻³ at 11JST, and the second 113µgm⁻³ at 10JST. This maximum value of 131µgm⁻³ had been the highest value in the spring season in 2005. Since the quantities of coarse particles were rather small, as clearly seen from Fig. 10, it can be inferred that the SPM mass density should be primarily composed of the masses of fine particles.

Fig. 13 shows the relationship between the OPC number concentration at Nagasaki University and the horizontal visibilities at Nagasaki Marine Observatory and Nagasaki Airport (15km north from Nagasaki University) on 23 May 2005. Table 1 also presents the horizontal visibilities observed at four meteorological sites; Tsushima Airport and Fukue Airport are added to the two in Fig. 13. In this figure and table, the visibility ranges equal to 10km and farther than 10km are denoted as 10km in Fig. 13, and as "T" in Table 1, respectively. The reason for these expressions is that the airport stations usually registered only the visibility ranges less than 10km. Moreover, the blank space in

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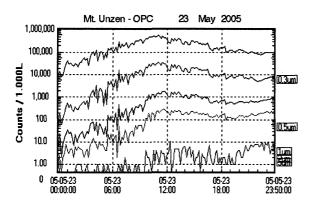


Fig. 11. OPC time series at Mt. Unzen (1290m) on 23 May 2005.

Table 1 implies no-observation. There is an inverse correlation between the concentration of the OPC fine particles and horizontal visibility. The lowest visibility 5km in Table 1 was observed at three airports at 8-10JST, when the OPC fine particles and SPM at Nagasaki University showed the highest concentrations, as seen from Fig. 10 and Fig. 12.

The analyzed data of sky radiometer at Nagasaki University on 23 May 2005 can be found in the SKYNET Sky radiometer Archives (Aoki, 2005), and showed that the maximum values of aerosol optical thickness at a wavelength of 0.5µm and Ångström exponent were 1.3 and 1.1, respectively. The optical thickness is somewhat larger than the case of 26 March 2003, while the Ångström exponent is almost the same. Since the turbidity parameters are derived from direct solar radiation and sky diffuse radiation, these quantities depend primarily on the aerosols in the total air column, so that they are not always proportional to the indices obtained from surface observations.

Fig. 14 shows the relationship between the number concentration of fine particles $(0.3 - 1\mu m)$ on 23 May 2005 and the mass concentration of elemental sulfur (S), which was detected from the Proton Induced X-ray Emission (PIXE) analysis. The PIXE sampling using a mini step sampler of Tohoku University was carried out at Nagasaki University with a time step of two hours in spring 2005. The details of the techniques of mini step sampler and PIXE analysis have been described in the paper of Matsuyama et al. (2003). For the convenience of comparison, the OPC concentration of fine particles is illustrated in a linear scale in Fig. 14. The most interesting feature of this figure is that the temporal change of sulfur density in every two hours is directly proportional to the number concentration of fine

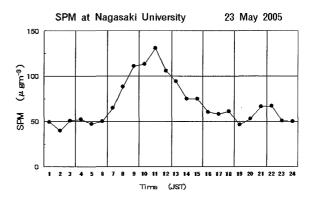


Fig. 12. Time series of the suspended particulate matter (SPM) at Nagasaki University on 23 May 2005.

particles; the quantity of S is increased with a rate of $0.2\mu \text{gm}^{-3}/(10^5 \text{ particles})$, suggesting that the amount of S is exactly depend on the fine particle concentration. This pattern agrees very well with the relationship between fine particles and AMS sulfate at Fukuejima, as already pointed out in Fig. 3 and Fig. 7. Elemental zinc(Zn) also increased in proportion to the concentration of fine particles, but this density was smaller by one order than the density of S, so that Zn was not the main component of fine aerosols. The other principal elements such as Fe, Ca and Si, which are probably main components of coarse particles, were less in density than S, although they had larger density than Zn. The density of Fe, ranked on the second place next to S, was less by half than that of S. These circumstances really suggest that S was the most prevailing element. In addition to this, the same correlations as Fig. 14 have been found on 4 April and 12 May when the OPC fine particles were also rich like on 23 May, although we do not present figures in this paper. As a result, it should be noticed that fine particle-rich conditions on 23 May 2005 were mainly consisted of sulfate aerosols.

Fig. 15 indicates the NOAA HYSPLIT backward trajectories at Nagasaki University ending at 12JST 23 May 2005 for three elevations of 500, 1000 and 2000m and for the duration of 72 hrs (NOAA, 2005). These backward trajectories also show the influence from the Chinese industrial region like those in Fig. 8, and furthermore, this case is characterized by air parcels at the two lower levels of 500m and 1000m taking mostly the same route with a low velocity from the Chinese coast to Nagasaki.

A large amount of evidence that the fine particles consist mainly of sulfate aerosols has been obtained

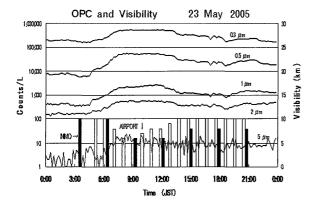


Fig. 13. Relationship between the OPC number concentration at Nagasaki University and horizontal visibility at Nagasaki Marine Observatory (NMO) and Nagasaki Airport.

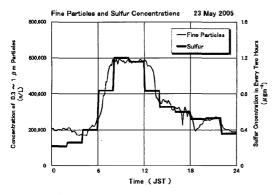


Fig. 14. Relationship of the OPC fine particle number concentration and the mass density of elemental sulfur both obtained at Nagasaki University on 23 May 2005.

both from AMS analysis in the 2003 case and PIXE analysis in the 2005 case. In addition, we have also verified the preceding paper written by Uematsu et al. (2002), in which they reported that the S-rich stage was detected at the former period of the Kosa event at Nagasaki on 17 April 1996. Since all the cases mentioned above are characterized by high proportionality between fine particles and sulfur substances, the atmospheric conditions rich with fine particles are practically ascribed to sulfate aerosols.

5. Summary and discussion

(1) Atmospheric conditions rich with fine particles, which are defined here as the particles in the size range between 0.3 and 1.0 μ m in diameter, are occasionally significant in recent years over the Nagasaki area in Japan. These conditions make the horizontal visibility very low like yellow sand (Kosa) events, although the

TIME (JST)	อ	6	7	8	9	10	11	12	19	14	1 5	16	17	18	19	20	21
NMO		т			6			7			8			т			т
Nasasaki Ainport	т	т	8	6	5	7	8	8	9	Т	т	т	т	т	т	T	т
Tsushina Airport				5	5	6	7	8	т	Т	т	т	т	T	т		
Fukue Airport				5	5	5	т	т	т	Т	т	Т	T	Т	т		

Table 1. Horizontal visibility at four observational sites in Nagasaki Prefecture on 23 May 2005.

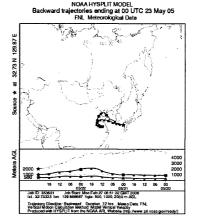


Fig. 15. NOAA HYSPLIT backward trajectories at the location of Nagasaki University ending at 12JST on 23 May 2005 for three elevations of 500m, 1000m and 2000m.

yellow sand events are caused by coarse particles larger than 1µm.

(2) Two cases of turbid conditions rich with fine particles, which took place during 25-27 March 2003, and on 23 May 2005 were investigated from the viewpoint of a detailed understanding of their influences to visibility and the properties of fine aerosols. As a summary of this study, we can point out that noticeably low visibility conditions are closely connected with the high concentration of sulfate aerosols which transported from the Asian continent.

(3) Atmospheric turbid conditions due to fine particles occasionally occur under the situation with no existence of yellow sand dust particles even in spring. This peculiarity seems to be gradually growing in recent years, and should be paid further attention from the viewpoint of air quality conservation over East Asia. The worst visibility will be then come into existence at the complex condition in which both yellow sand particles and fine particles are so rich simultaneously. If we encountered such a worse condition, we would have an extremely low visibility in which certain kinds of obstructions to traffic will be taken place.

(4) One of interesting features obtained in this study is that the concentrations of fine particles at Mt. Unzen in Figs. 5 and 11 showed almost the level as those of Nagasaki University, in spite of their considerable difference in height. The altitude of the mountain site (1290m) is nearly the upper limit of the atmospheric boundary layer. Features of turbid condition at the mountain are serious in foresight of maintaining future air quality. More careful and detailed investigations should be also performed as an environmental problem of vertical distributions of atmospheric aerosols.

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