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New Particle Formation of Marine Aerosols

K. Miura[1]*, H. Furutani[2], Y. Iwamoto[2], K. Nagano[3], H. Kobayashi[4], M. Mochida[5], H. Mukai[6], S. Hashimoto[6], M. Takami[6] and M. Uematsu[2]

[1]*Department of Physics, Faculty of Science, Tokyo University of Science, 1-3 Kagurazaka, Shinjuku-ku, Tokyo 162-8601, Japan*
[2]*Atmosphere and Ocean Research Institute, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8564, Japan*
[3]*Department of Liberal Arts, Faculty of Science and Technology, Tokyo University of Science, Japan*
[4]*University of Yamanashi, Japan*
[5]*Nagoya University, Japan*
[6]*National Institute for Environmental Studies, Japan*
**E-mail: miura@rs.kagu.tus.ac.jp*

Introduction

Sulfur and organic species originating from the ocean make new particles which increase the number of cloud condensation nuclei and change the properties of cloud (Charlson *et al*. 1987). However, in the planetary boundary layer, there are many sea-salt particles that provide surfaces for heterogeneous chemical reactions with sulfur or organic gases. There are very few papers concerning new particle formation observed in the boundary layer under a high-pressure system (e.g., Covert *et al*. 1996). This suggests that particles are produced in the free atmosphere.

To clarify the mechanism of their formation, the size distribution of submicron aerosols was measured on research vessels, on shores, and in the free troposphere. We describe here the measured results on the Hakuho Maru KH-04-3 expedition, at Cape Ochiishi, and at the summit of Mt. Fuji.

Methods

Size distributions from 4.4 nm to 5000 nm in diameter were measured with a scanning mobility particle sizer (SMPS) (TSI Inc., 3034, 3936N or 3936L) and an optical particle counter (OPC) (RION Co. Ltd., KC01E or KR12A). Sample air was dried to lower than 40% with a ribbon heater. The transport of sulfate, organic carbon (OC), and black carbon (BC), was estimated using the Chemical weather FORecasting System (CFORS). CFORS was developed by Prof. Itsushi Uno and Mr. Koji Ishihara of the Research Institute for Applied Mechanics (RIAM), Kyushu University, Japan. The system migrated, in June 2002, to the National Institute for Environmental Studies (NIES) with financial support by the Center for Global Environmental Research (CGER). (http://www-cfors.nies.go.jp/~cfors/research/). Outline of RIAM-CFORS (by Prof. Uno, RIAM, Kyushu Univ.) is shown on the web site (http://www-cfors.nies.go.jp/~cfors/outline.html).

Results and Discussion

KH-04-3 expedition

Measurements of size distributions had been performed during the Hakuho Maru KH-05-2, KH-06-2, KH-08-2, KH-10-2,

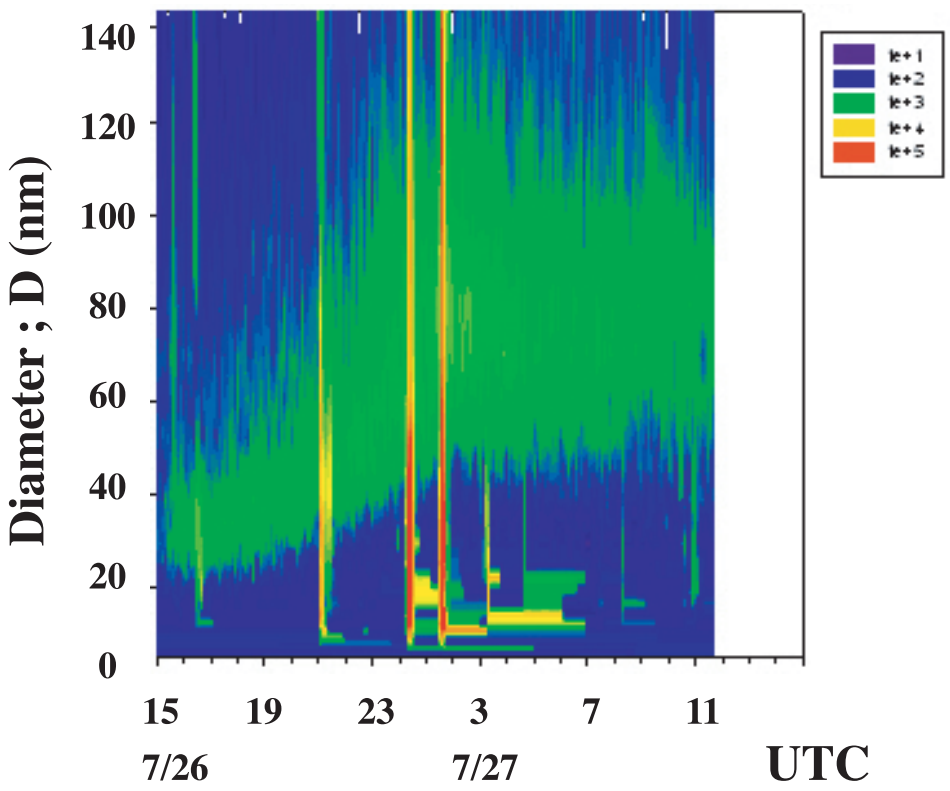

Fig. 1. Size distributions measured on R/V Hakuho Maru, on 26 and 27 July, 2004.

cruises, the Tansei Maru KT-07-7, KT-09-5, cruises, and the Mirai MR07-4, MR07-5, MR08-6, MR09-1, cruises. However, unfortunately, a clear event of a new particle formation was not observed. We describe one event of a new particle formation observed in the Hakuho Maru KH-04-3 expedition.

The Hakuho Maru KH-04-03 cruise took place during the periods from 13 July to 25 August, 2004. Size distributions from 4.4 to 5000 nm in diameter was measured on the surface continuously, with three counters through a diffusion dryer (about 20% RH) from a sampling pipe (3 m length) standing on the compass deck, which was about 15 m above the sea surface and in the direction of 30 degrees right from the front of a funnel. Measurements were taken every 5 min. The peak at 30 nm appeared at 16:00 on 26th UTC and grew by condensation during daytime (LT = UTC + 11 h) and had a broad distribution between 60 and 100 nm at 0 o'clock on 27th (Fig. 1). The exhaust of the ship sometimes caused a high concentration of aerosol particles. But this plume was independent of contamination and seems to have been produced around the station.

A year-round observation of size distribution of aerosol particles at Cape Ochiishi

Cape Ochiishi faces the North Western Pacific Ocean, where primary productivity is high. A test observation was carried out from 22 May to 18 June, 2008 and a

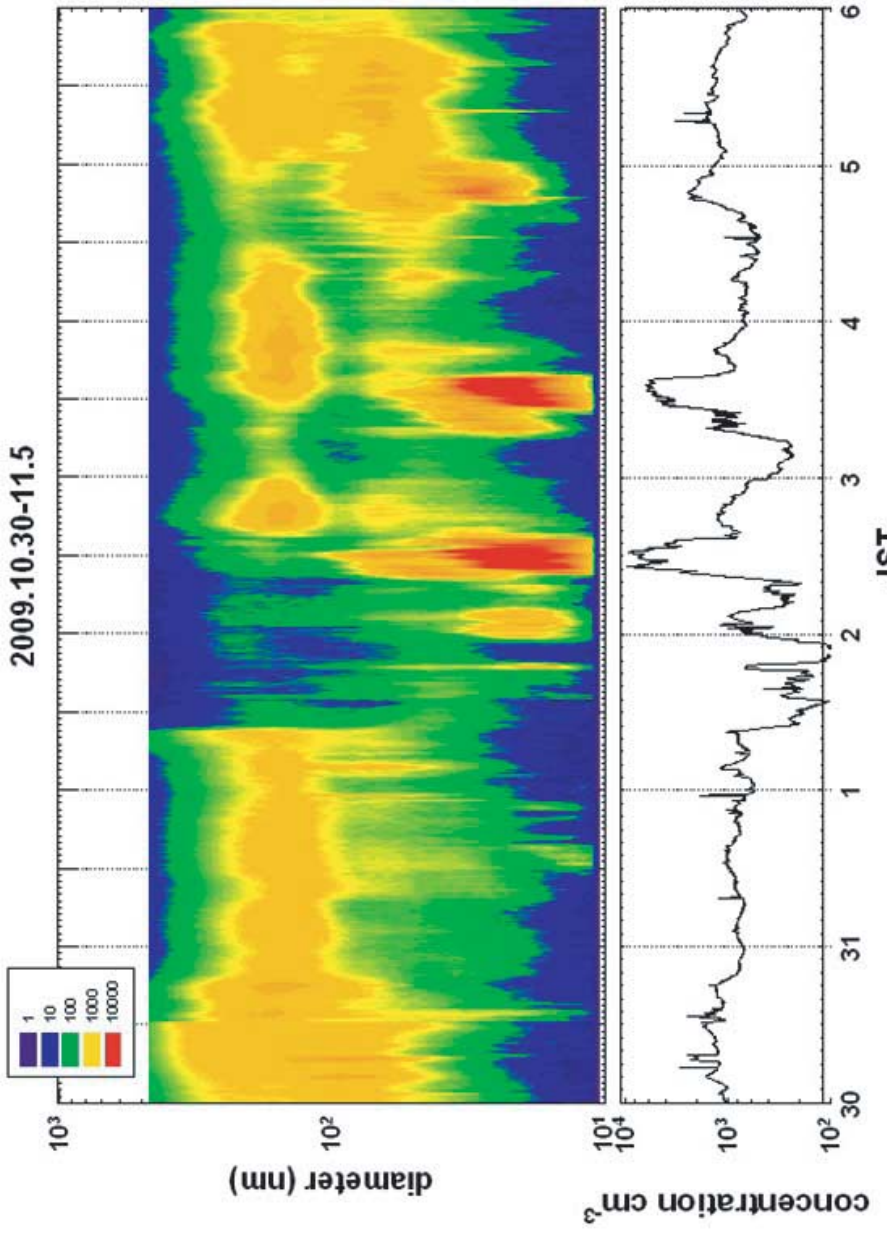

Fig. 2. Size distributions measured at Cape Ochiishi, between 30 Oct. and 5 Nov., 2009.

Table 1. Number of burst events measured at Cape Ochiishi.

Month	Events	Observation days	Fraction (%)
2008 May	1	10	10.0
2008 June	1	18	5.6
2009 October	5	16	31.2
2009 November	3	13	23.1
2009 December	5	11	45.5
2010 January	10	30	33.3
2010 February	4	26	15.4
2010 March	2	31	6.5
2010 April	0	30	0.0
2010 May	3	31	9.7
2010 June	1	27	3.7
2010 July	1	21	4.8
Sum	36	264	13.6

year-round observation was undertaken from 16 October, 2009, to 7 September, 2010. A size distribution from 10 nm to 487 nm in diameter was measured with a scanning mobility particle sizer (SMPS, TSI 3034) (Fig. 2). Sample air was dried to lower than 40%. The existence of an inversion layer was estimated with a temperature profile measured at the surface, and 10 m, 30 m, and 50 m in altitude.

A burst of particles smaller than 20 nm in diameter and continuing for longer than 3 h was observed 36 times during the observation periods (Table 1). Seven events were observed in early summer and the others were in autumn and winter. A banana shape was faintly observed 21 times. The transport of sulfate, OC, and BC was observed 11, 26, 30 times, respectively. The source of the air mass was estimated with these elements, weather map, surface wind direction and backward trajectory analysis. The air mass of 26 events was estimated to be continental. It suggests that maritime nucleation was observed during the year, however, a clear nucleation related to marine sources was not observed.

Observation of size distribution at the base and the summit of Mt. Fuji

As the summit of Mt. Fuji, Japan, is usually positioned in the free troposphere, we can measure the variation of aerosol in the free troposphere. Size distributions from 4.4 nm to 5000 nm in diameter were measured with a scanning mobility particle sizer (SMPS, TSI 3936N25 or 3936L22) and an optical particle counter (OPC, RION KR12 or KC01C), at the summit (3776 m, 35.36N, 138.73E) and the base Tarobo (1300 m) in July and August, 2006 to 2010. Sample air was dried to lower than 20% with a diffusion dryer. Radon, O_3, and CO gases were also measured as tracers.

One feature of the results is a peak measured with SMPS at the summit. This peak of 10 nm appeared at about 11.00 h on 9 and 10 August 2006, and increased in size during the daytime. Volume distributions kept an almost constant value during the increase in size. This suggests that this size increase was due to coagulation, and that this phenomenon happened over a wide area. Moreover, these events did not synchronize at Tarobo. This suggests that these peaks may indicate a new particle production by gas to particle conversion in the free troposphere around the summit. The event on 9 August occurred after the passing of a typhoon. It is expected that sulphur and organic species carried by the

typhoon produced new particles in the free troposphere.

A burst of particles smaller than 20 nm in diameter continuing longer than 3 h was observed 81 times during five summer seasons (134 days). These events were more observed at night (56) than in the daytime (25). The reason for this phenomenon is not obvious. Most of these events did not synchronize at Tarobo.

The origin of the air mass of 13 times events in 2010 was estimated with a weather map, a prediction of chemical weather map by CFORS, surface weather elements, and surface gas concentrations. The results showed that a continental, and maritime, origin was estimated 2, and 5 times, respectively. It is found that the concentration just before the maritime event showed a lower value than that just before the other events.

Conclusions

It is difficult to clarify the mechanism of new particle formation by the nucleation of gas-phase compounds emitted from marine biogenic sources by measuring size distributions on research vessels, so a year-round observation on Cape Ochiishi facing the North Western Pacific Ocean, and intensive observations at the summit of Mt. Fuji in the free troposphere, were undertaken.

(1) The Ochiishi observation showed that the maritime nucleation was observed during the year; however, a clear nucleation related to marine sources was not observed.

(2) The Mt. Fuji observation showed that the burst of particles smaller than 20 nm in diameter continuing longer than 3 h was observed 81 times during five summer seasons and these events were observed more at night than in the daytime. It is also found that the concentration just before the maritime event was lower than those just before the other events.

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