

Retrieval of hourly atmospheric radiocesium in the early period of the TEPCO Fukushima Daiichi Nuclear Power Plant accident by analyzing used filter-tapes of operational air pollution monitoring stations

*Haruo Tsuruta (Remote Sensing Technology Center of Japan, Japan), Yasuji Oura (Tokyo Metropolitan University, Japan), Mitsuru Ebihara (Waseda University, Japan), Toshimasa Ohara (National Institute for Environmental Studies, Japan), Yuichi Moriguchi (University of Tokyo, Japan), Teruyuki Nakajima (National Institute for Environmental Studies, Japan)

1. Background

The data of atmospheric radionuclides measured in the Fukushima prefecture and its surrounding area just after the TEPCO Fukushima Daiichi Nuclear Power Plant (FD1NPP) accident are expected to contribute to,

- (1) evaluation of internal exposure dose from inhalation,
- (2) re-estimation of the source term of radionuclides released to the atmosphere by the accident, and
- (3) validation of numerical simulation results by atmospheric transport and deposition models and their improvement.

They have still uncertainties, however, because there were very limited data of atmospheric radionuclides measured just after the accident which are open to the public (Fig. 1).

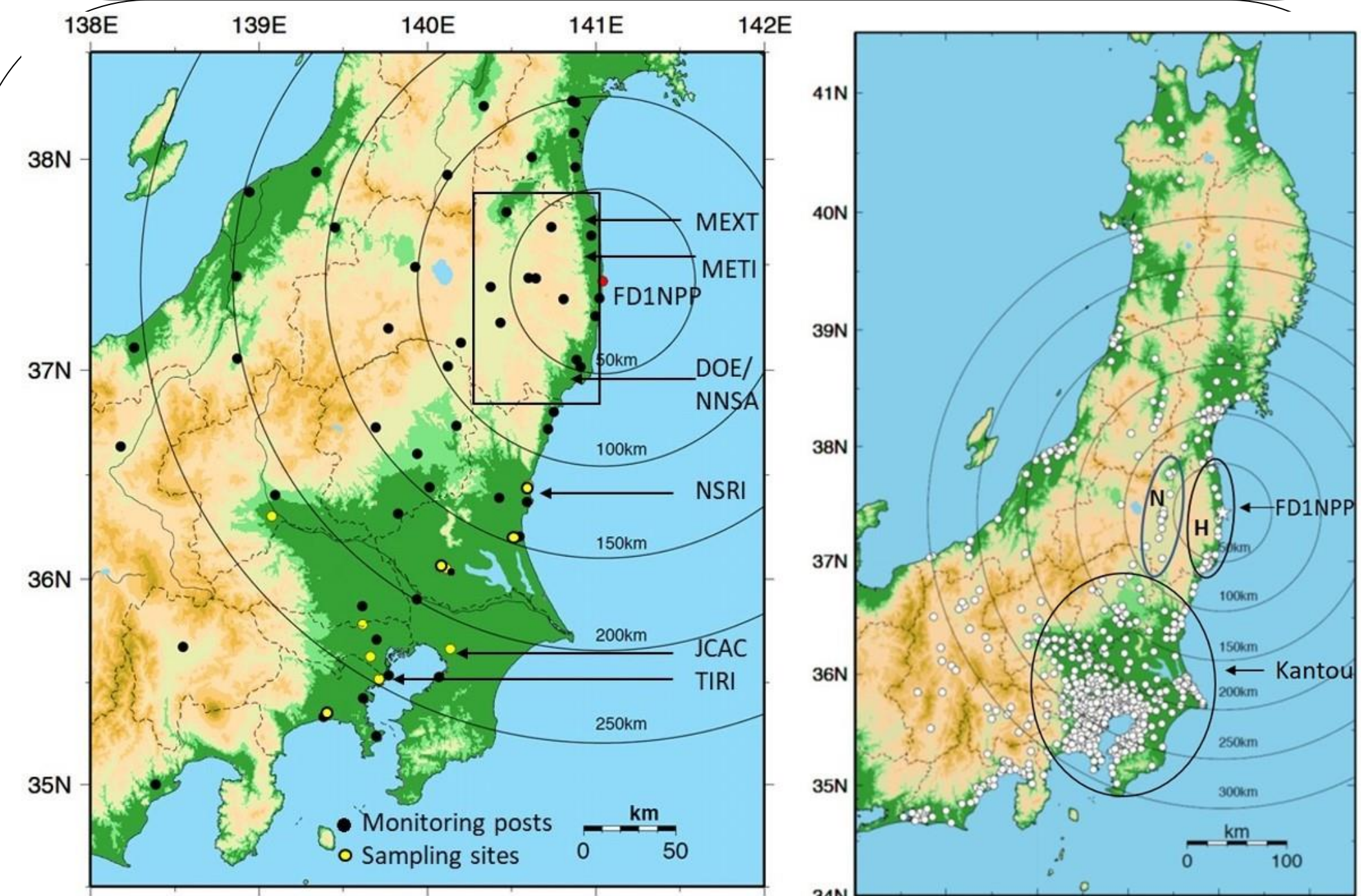


Fig. 1 (left) Sites for radiation dose and atmospheric radionuclides measured after the accident. Fig. 2 (right) SPM monitoring sites in eastern Japan before the accident.

Table 1 Days and areas of the transport of plumes/polluted air masses in the Fukushima and Kantou areas during March 12-25, 2011

Plume	No.	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	Range of plumes	Source	Ref.
																						North	South	Unit
P1	○																					○		1
P1V	●																					○		1
P1	●																					○		1
P1'	●																					○		1
P2	○																					○		1
P2	○																					○		1
P3	○																					○		1
P4	○																					○		1
P4'	○																					○		1
P5	○																					○		1
P5'	○																					○		1
P6	○																					○		1
P7	○																					○		1
P8	○																					○		1
P8'	○																					○		1
P9	○																					○		1
P9'	○																					○		1
P10	○																					○		1
P11	○																					○		1

No.: Number of major plumes transport from FD1NPP in the early period after the accident.

- : Maximum ^{137}Cs concentrations: $^{137}\text{Cs} \geq 1,000 \text{ Bq m}^{-3}$.
- : Maximum ^{137}Cs concentrations: $1,000 < ^{137}\text{Cs} \geq 100 \text{ Bq m}^{-3}$.
- : Maximum ^{137}Cs concentrations: $100 < ^{137}\text{Cs} \geq 15 \text{ Bq m}^{-3}$.
- : High ^{137}Cs concentrations continued to the following morning.
- : Precipitation was observed in the area.

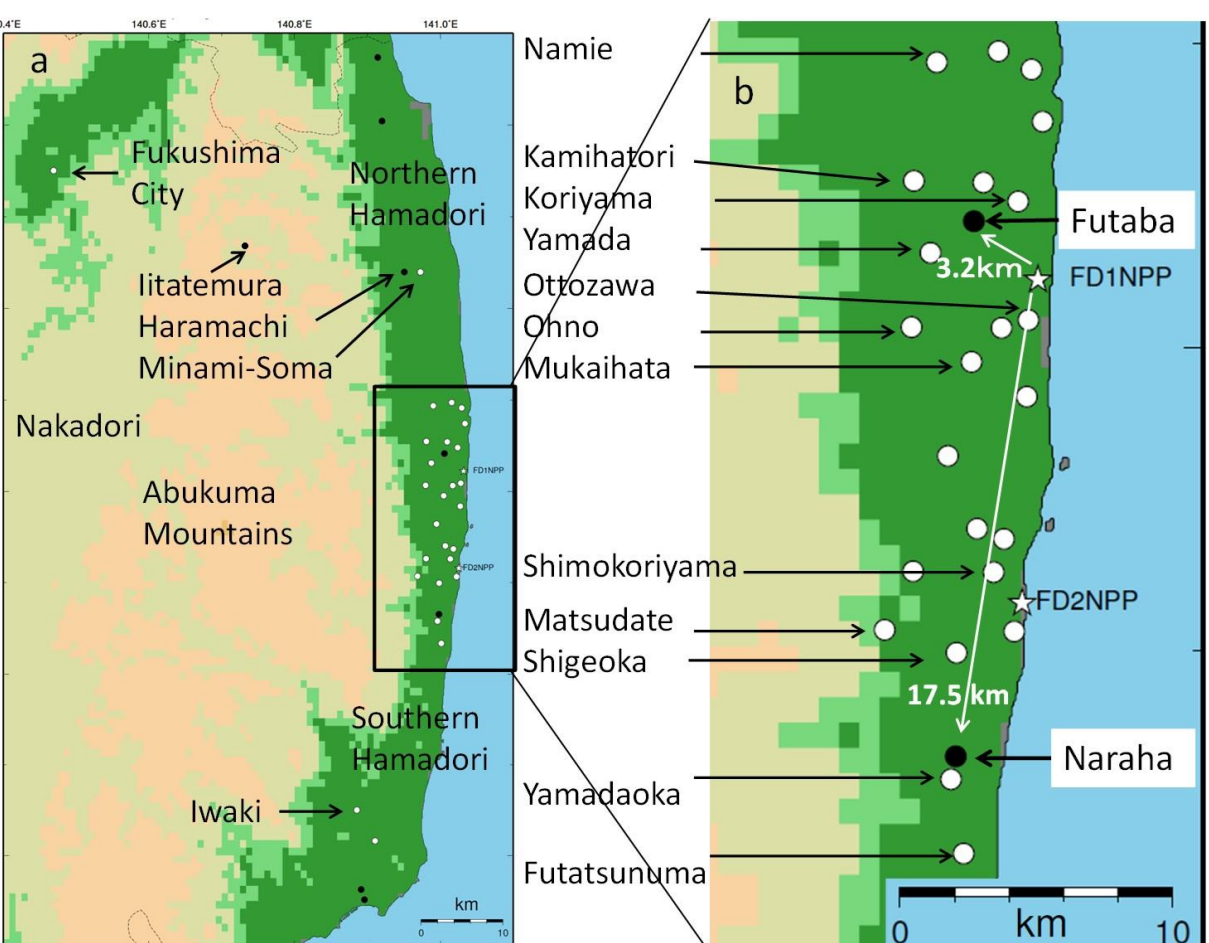


Fig. 3 Map of (a) the eastern part of Fukushima prefecture and (b) SPM stations (●) in the air pollution monitoring network and monitoring posts for environmental radioactivity (○) by Fukushima prefecture in Hamadori.

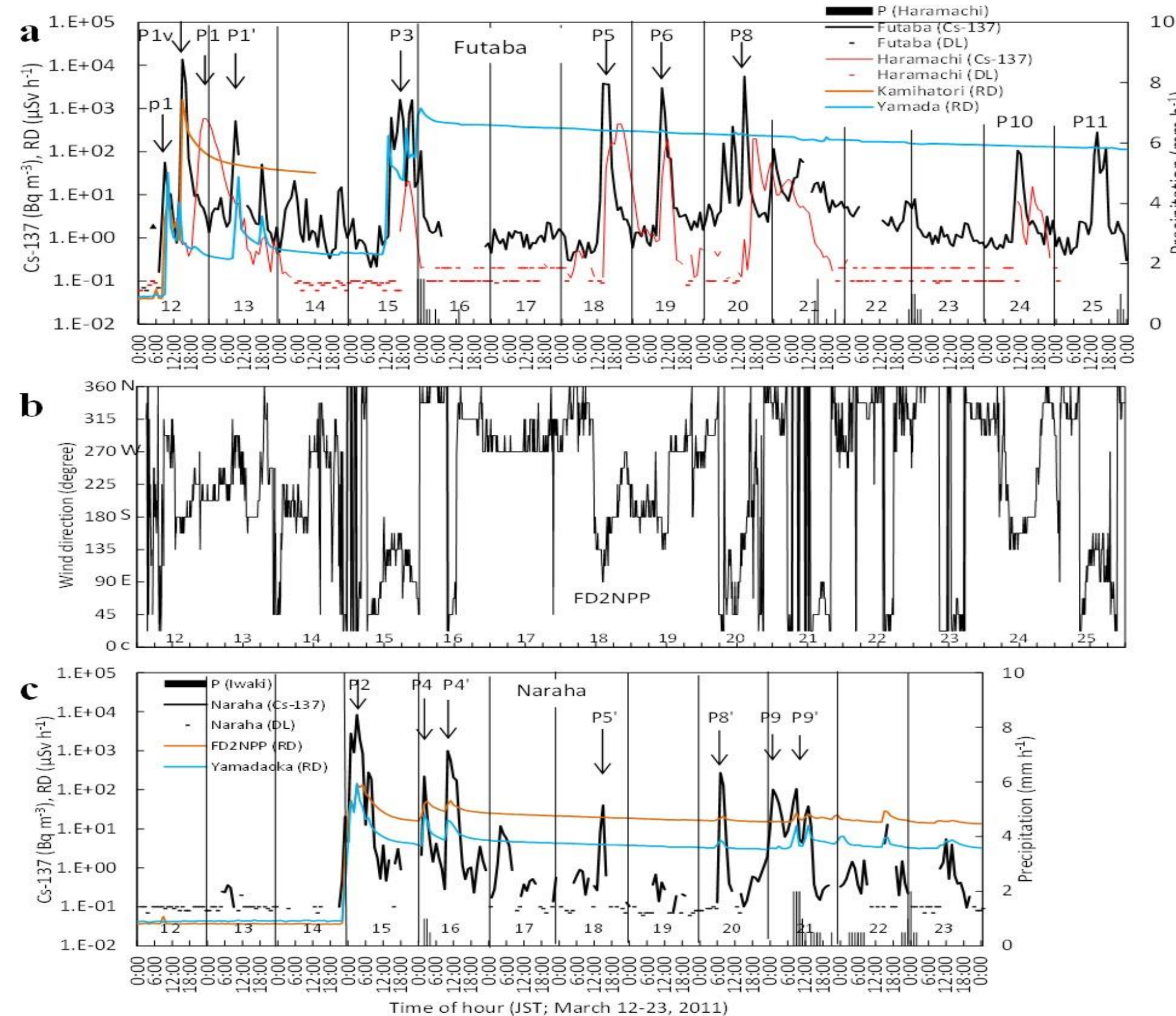


Fig. 4 Time series of (a) the hourly ^{137}Cs concentrations at Futaba and Haramachi, the radiation dose (RD) at Kamiatori and Yamada, the precipitation at Haramachi during March 12-25, 2011, (b) the wind direction at the FD2NPP, and (c) the hourly ^{137}Cs concentrations at Naraha, RD at the FD2NPP and Yamadaoka, the precipitation (P) at Iwaki during March 12-23, 2011. DL means the ^{137}Cs concentration below detection limit.

2. Objectives

1. This paper: to analyze the hourly ^{137}Cs in atmospheric aerosols just after the FD1NPP accident, by measuring radionuclides collected on filter-tapes of Suspended Particulate Matter (SPM: $d < 10 \mu\text{m}$) mass monitor with β -ray attenuation method, which are routinely operated at air pollution monitoring stations all over Japan. The used filter-tapes were kindly offered by local governments through the Ministry of the Environment, Japan.

2. Goal: to retrieve the spatio-temporal distribution of ^{137}Cs and ^{131}I in the Fukushima prefecture and surrounding area during March 12-31, 2011, by measuring radionuclides (^{137}Cs and ^{129}I) in SPM collected on the filter-tapes.

Results (1)

1. Analyzing the used filter-tapes, 19 plumes were identified in Fukushima prefecture and the Kantou area during March 12-25, 2011 (Table 1, Fig. 2).

2. Many plumes were observed in northern or southern Hamadori when the wind direction was from the south or north, respectively (Table 1, Figs. 3, and 4).

3. The highest ^{137}Cs concentration of $13,600 \text{ Bq m}^{-3}$ was measured at Futaba at 14:00-15:00 (JST), March 12, after the vent operation of Unit 1 (Figs. 3, 4 and 8).

4. Transport pathways of plumes (P1-P9) were estimated by the atmospheric transport and deposition models (Fig. 5).

5. In Nakadori, plume P3 was transported from south to north, while plume P8 was transported from north to south due to the local wind pattern (Table 1, Fig. 7).

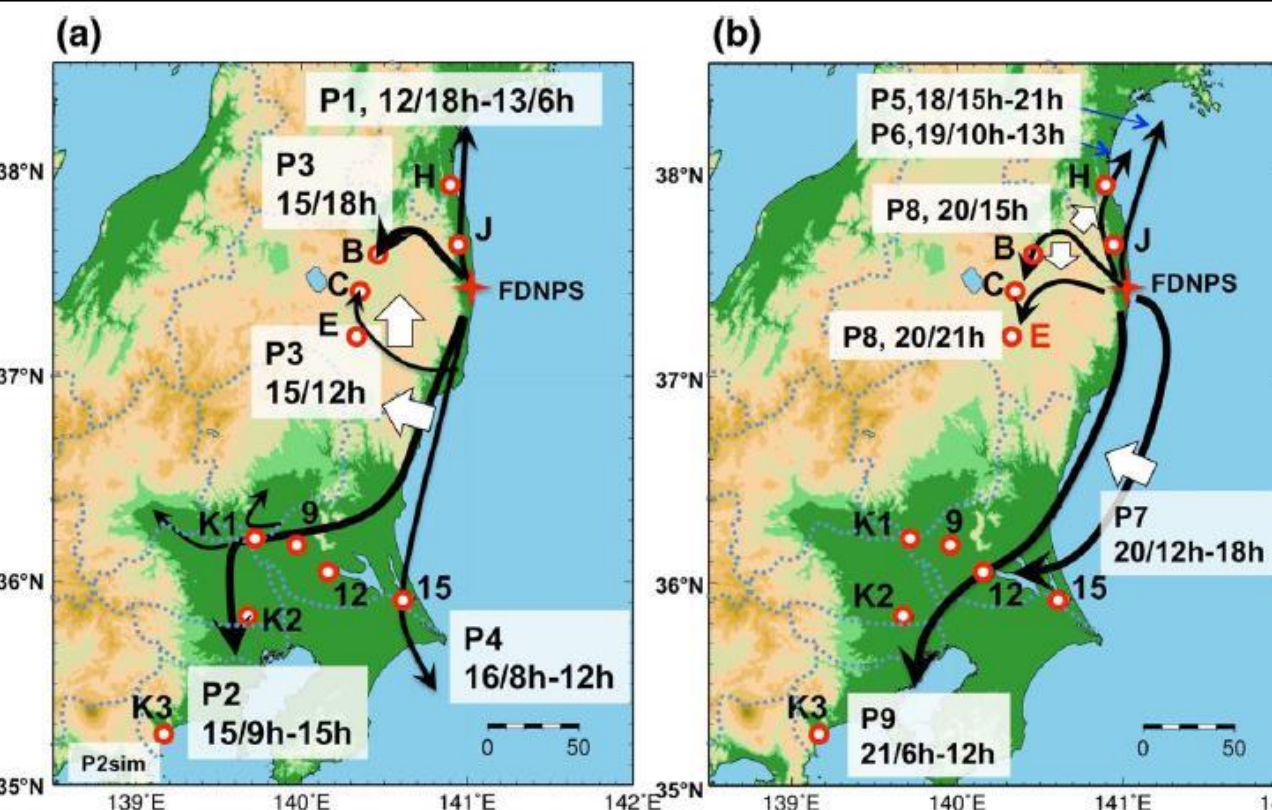


Fig. 5 Schematic diagrams of transport routes for nine plumes (P1-P9). Thick arrows indicate the general trend of the movement of each plume. The alphabetical and numerical numbers are the SPM sites. "P3, 15/18h" means Plume P3, and 18:00 (JST) on March 15, 2011. See the paper for the details.

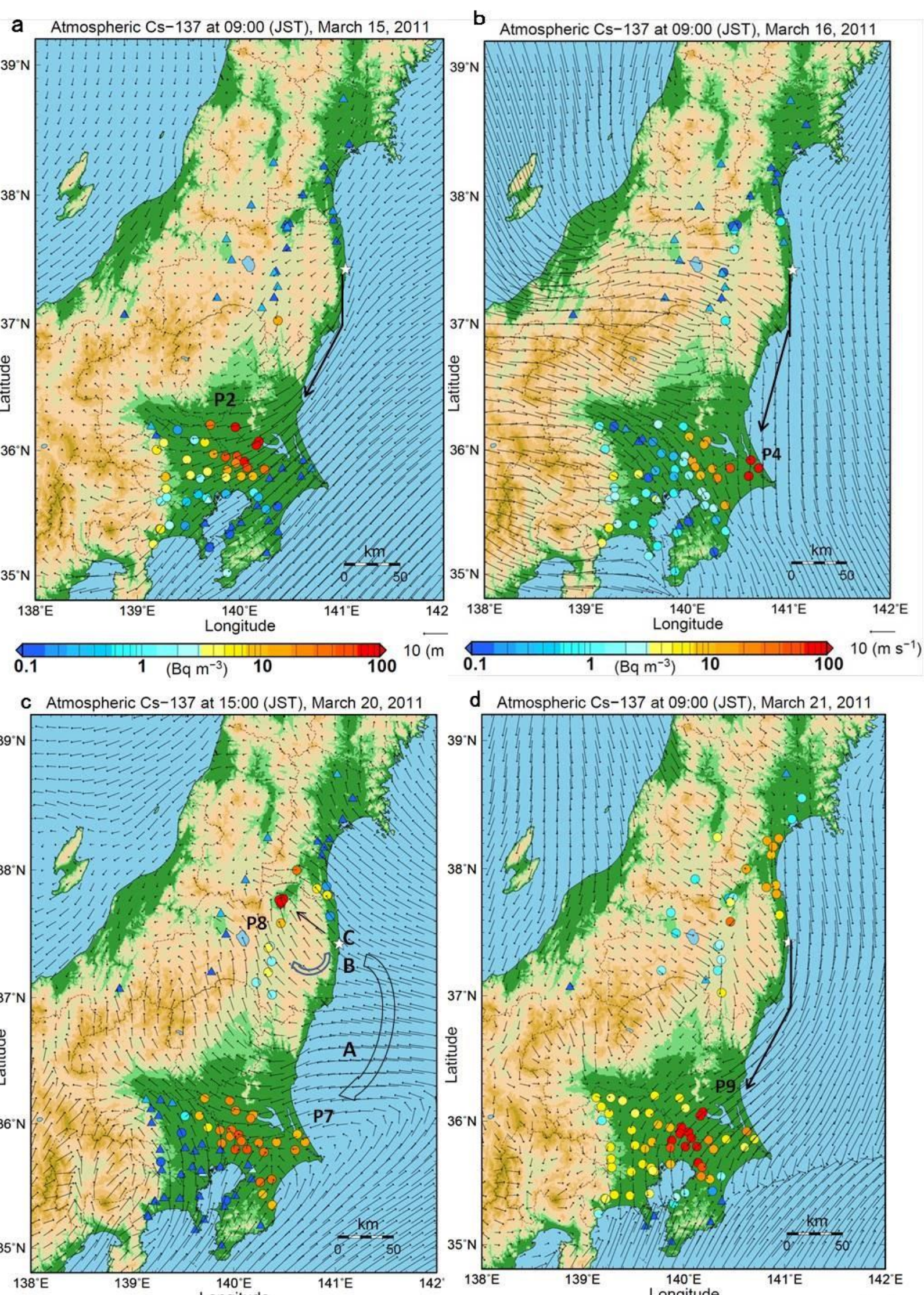


Fig. 6 Spatial distributions of atmospheric ^{137}Cs concentrations (colored dot) in major Plumes and wind vectors (black arrows) at 1000 hPa by mesoscale objective analysis (JMA) in eastern Japan. Black straight and curved lines are the schematic routes of plumes. (a) Plume P2 at 9:00 (JST), Mar. 15, 2011. (b) Plume P4 at 9:00, Mar. 16. (c) Plumes P7 and P8 at 15:00, Mar. 20. Plumes were shifted clockwise as A, B, and C. (d) Plume P9 at 9:00, Mar. 21.

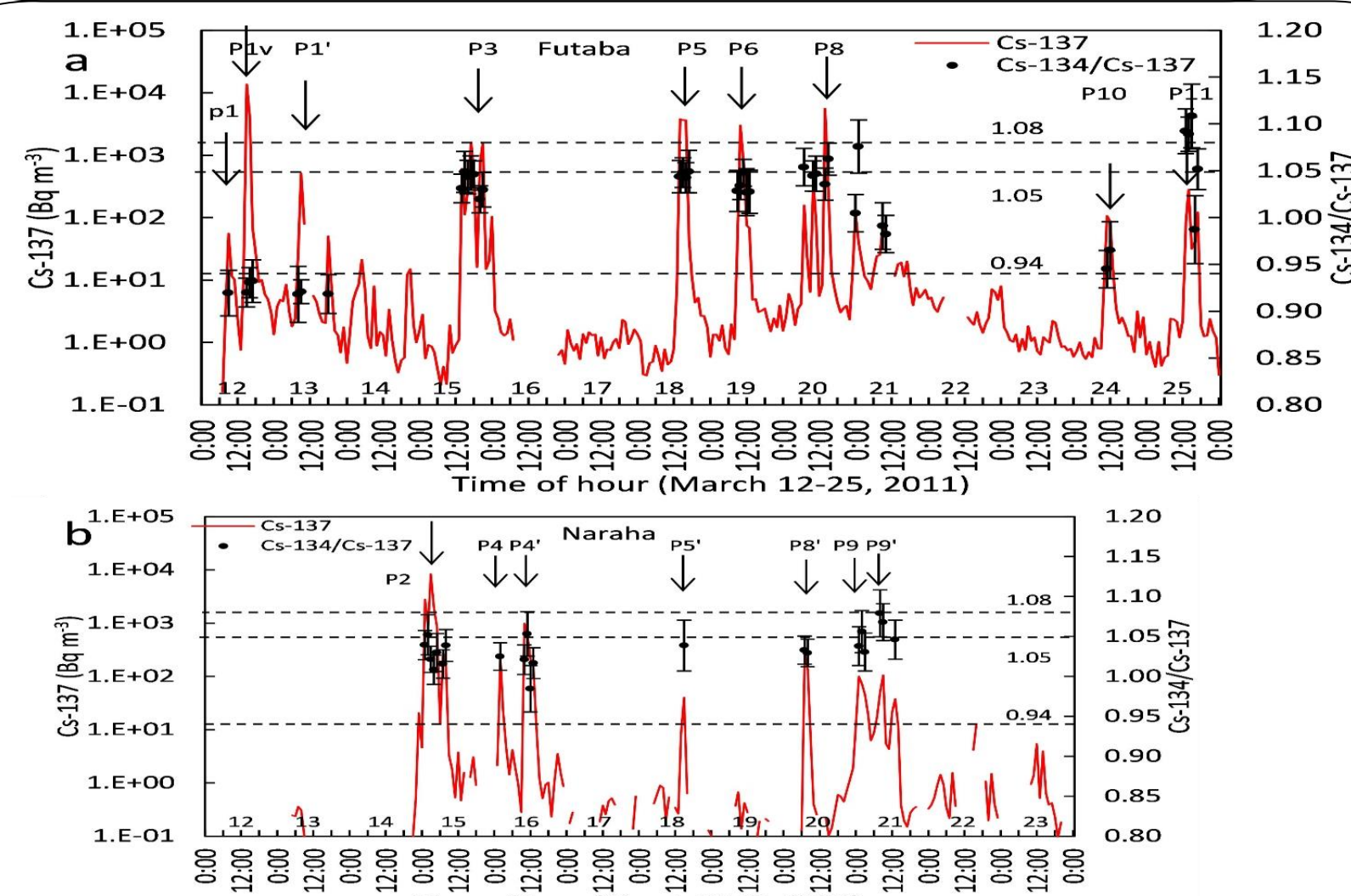


Fig. 8 Time series of the ^{137}Cs concentrations and the ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ at (a) Futaba and (b) Naraha, during March 12-25, 2011. The numerical values of 0.94, 1.05, and 1.08 are the inventory data of the ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ at Unit 1, Unit 3, and Unit 2, respectively.

3. Method

1. SPM monitoring stations: Hamadori (H in Fig. 2) and Nakadori (N) in the east-coast of the Fukushima prefecture. Kantou area including Tokyo Metropolitan Area $\geq 170 \text{ km}$ southwest of the FD1NPP.

2. Periods: March 12-25, 2011 (Excluding the dates when plumes were assumed not to be transported to the SPM sites)

3. Measurement: ^{134}Cs and ^{137}Cs by Ge detectors

Uncertainty analysis caused by re-use of the filter-tapes:

1. Cross-contamination.
2. Identification of date and hour for each spot.

Results (2)

5. 4 plumes were transported to the Kantou area when a northeasterly or easterly wind prevailed. The maximum ^{137}Cs concentrations were equal to or higher than those measured in Nakadori located about 60-70 km west of the FD1NPP (Table 1, Fig. 6).

6. The measured plumes during March 12-13 and the other days were estimated to be released from Unit 1 and Unit 2/3, respectively, by comparing the activity ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ in the plumes with those in the inventory data (Fig. 8).

7. The area of high ^{137}Cs deposition densities in the Kantou area by MEXT (2011) was consistent with that of the SPM stations with the high time-integrated ^{137}Cs concentrations on the morning of March 21 when the precipitation was observed uniformly in the Kantou area (Fig. 9).

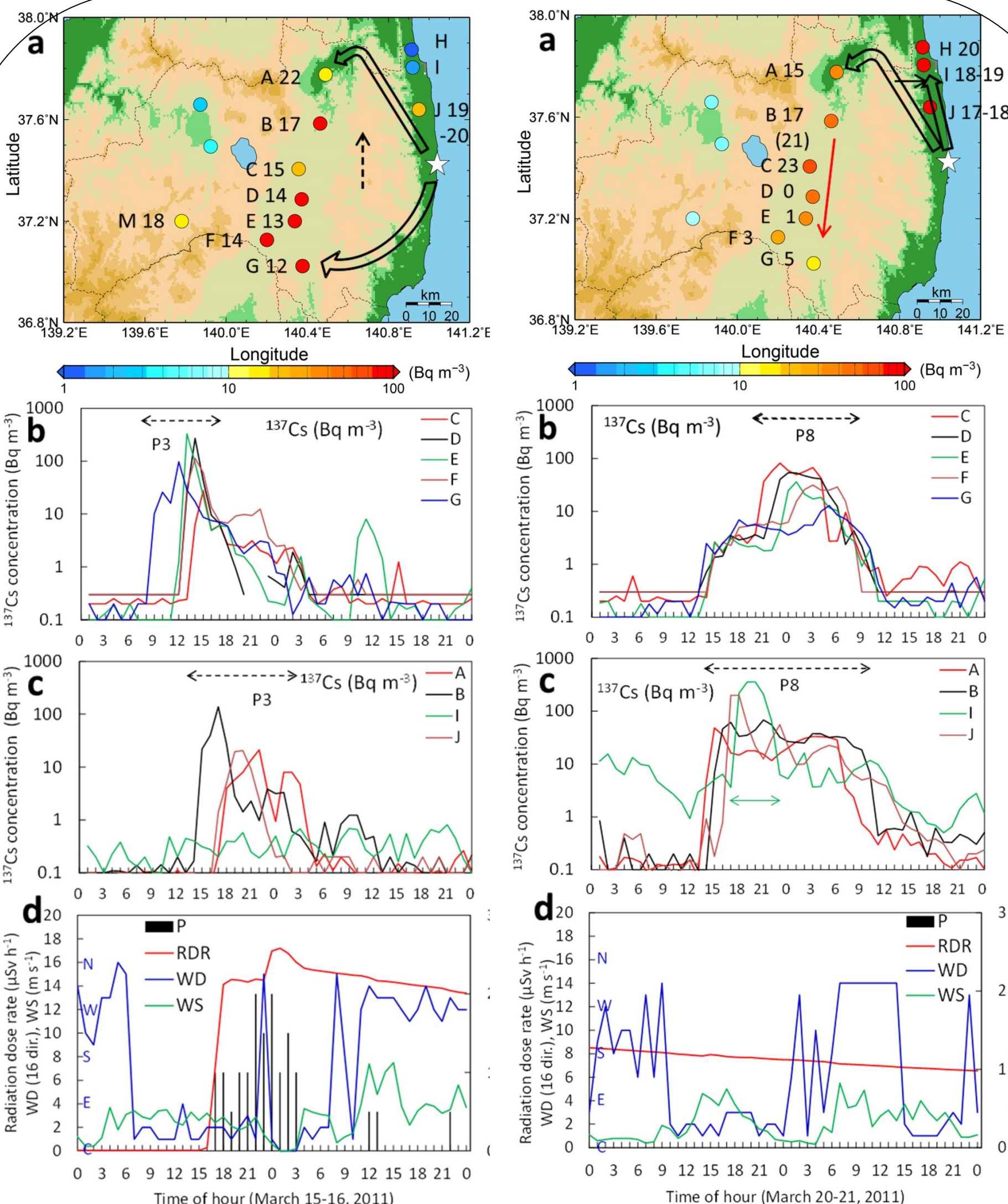


Fig. 7 (a) The maximum ^{137}Cs concentration with the colored dot. The number is the time (hour) of the maximum ^{137}Cs during the plume arrival. (b & c) Time series of ^{137}Cs concentrations at sites A-J in Fig. 7a. (d) Time series of radiation dose rate (RDR), wind direction (WD), wind speed (WS), and precipitation (P) at Fukushima AMeDAS station (JMA) located near site A.

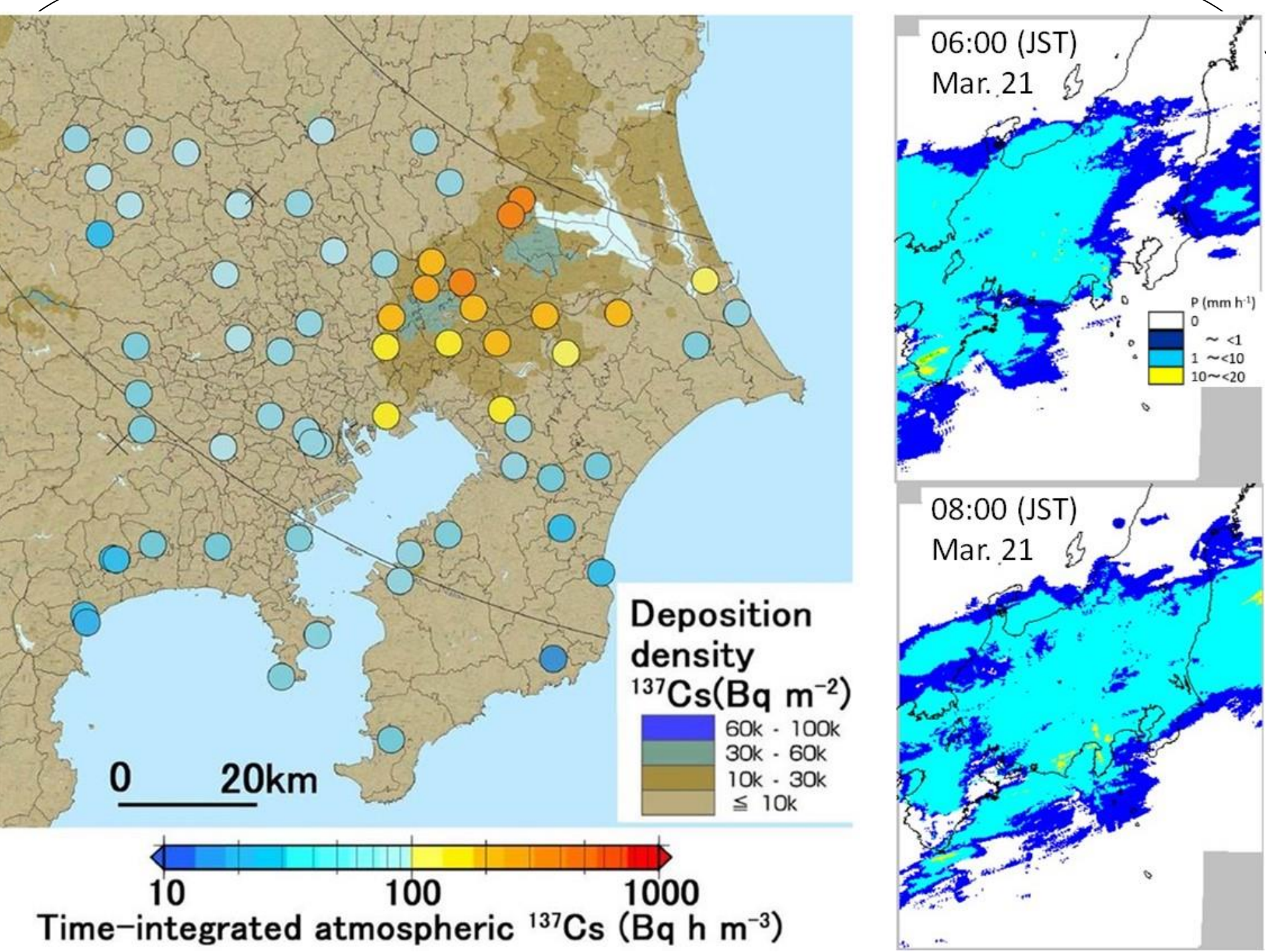


Fig. 9 (Left) Time-integrated atmospheric concentrations (Mar. 21) and deposition map (by MEXT) for ^{137}Cs in the TMA. (Right) Precipitation maps at 06:00 and 08:00, March 21, by Radar-AMeDAS (JMA).

References:

1. Tsuruta et al., Sci. Rep., 4, 6717 (2014).
2. Tsuruta et al., Earozoru Kenkyu, 32, 244-257 (2017) (in Japanese).
3. Tsuruta et al., Geochemical J., 52, 103-121 (2018).
4. Nakajima et al., PEPS, 4(2) (2017).
5. Nishihara et al., JAEA-DATA/Code 2012-018 (2012).

Acknowledgements:

We greatly thank to all the local governments who kindly offered the used SPM filter-tapes. This study was partly supported by JSPS KAKENHI (grant numbers are 24110002, 24110008, 24110009) and by the Environment Research and Technology Development Fund (JPMEERF20155001) of the Environmental Restoration and Conservation Agency of Japan.