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#### Supplement of

### Photochemical age of air pollutants, ozone, and secondary organic aerosol in transboundary air observed on Fukue Island, Nagasaki, Japan

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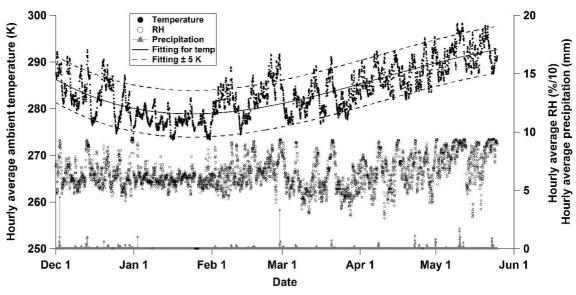


Figure S-1. Time-series plots of hourly average ambient temperature, relative humidity (RH), and precipitation during the field study.

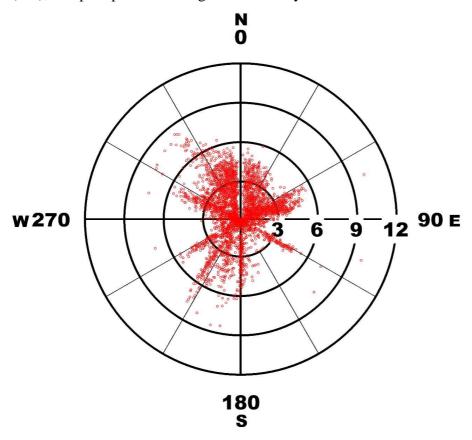


Figure S-2. Polar plot of the dependence of hourly average wind speed (radial axis in m  $s^{-1}$ ) on wind direction (degrees) observed at the Fukue monitoring station (north = 0 degrees, east = 90 degrees).

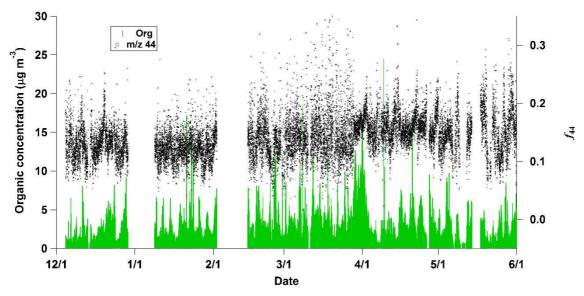


Figure S-3. Temporal variation of concentration of organic species and fraction due to m/z 44,  $f_{44}$ , in the mass spectra of organic species measured by quadrupole aerosol mass spectrometry.

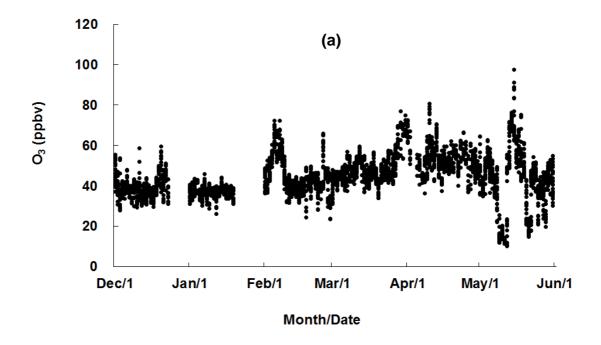
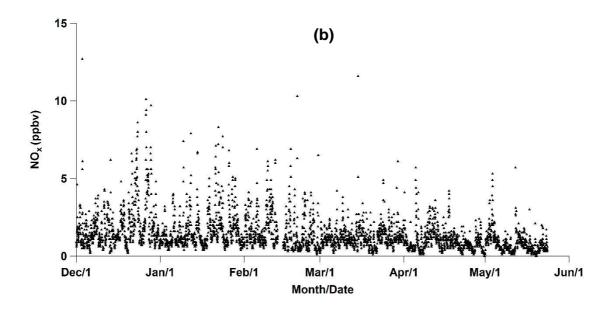


Figure S-4. Temporal variation of hourly average mixing ratios for (a)  $O_3$ , (b)  $NO_x$ , (c)  $NO_y$ , (d) CO, (e) *i*-pentane, (f) toluene, and (g) ethyne.



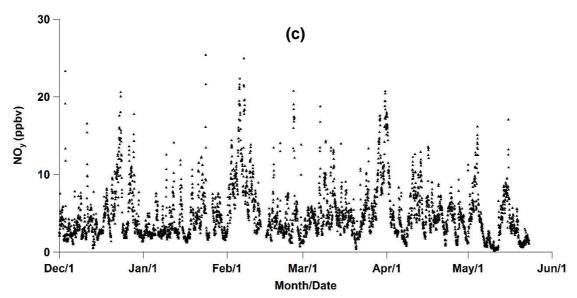
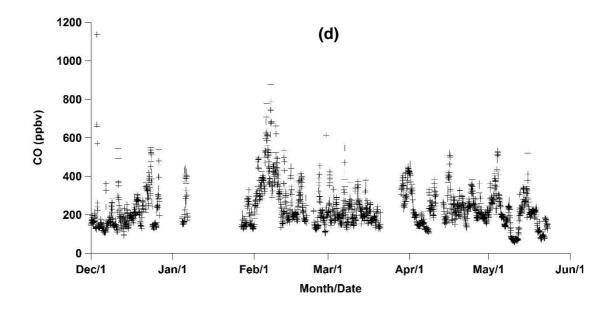


Figure S-4. Continued.



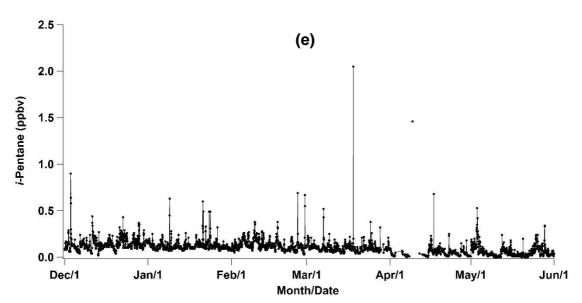


Figure S-4. Continued.

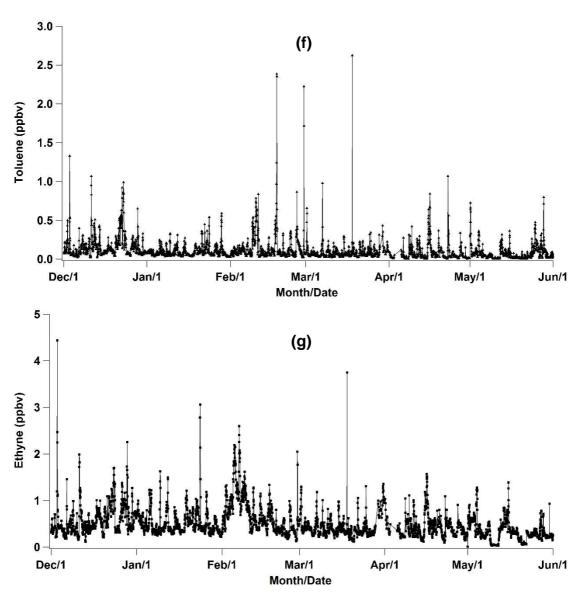


Figure S-4. Continued.

# NOAA HYSPLIT MODEL Backward trajectories ending at 2100 UTC 11 May 11 GDAS Meteorological Data

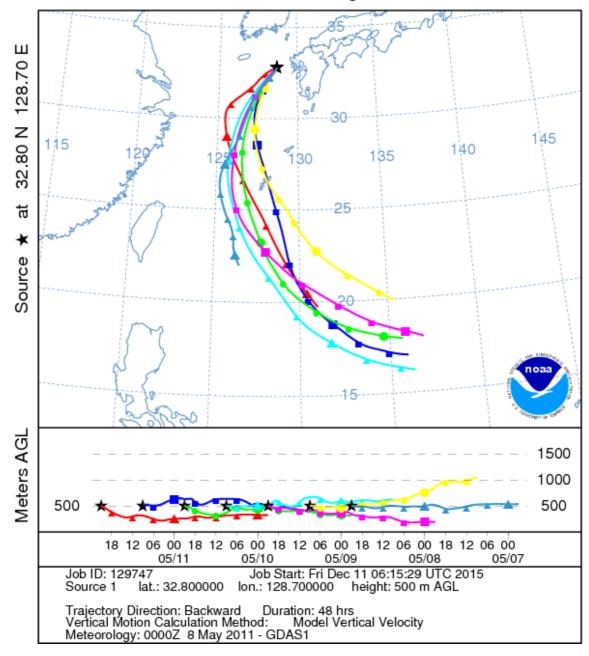


Figure S-5. Back trajectories of air masses arriving at Fukue during the low mixing ratio episode of ozone (May 9 to 11).

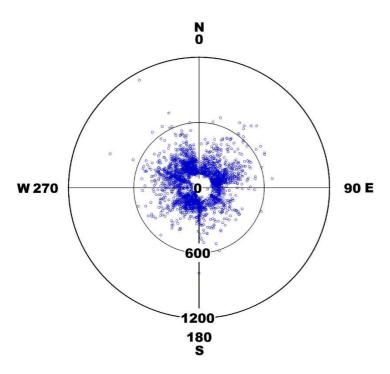


Figure S-6. Wind-sector dependence (north = 0 degrees, east = 90 degrees) of CO mixing ratio (the radial axis in ppbv).

Table S-1. Observations of chemical species concentration rise during the seven primary emission episodes indicated by CO mixing ratio<sup>a</sup>

	episode 1	episode 2	episode 3	episode 4	episode 5	episode 6	episode 7
beginning $^b$	12/21 18:00	2/1 1:00	3/28 20:00	4/15 5:00	4/24 6:00	5/1 4:00	5/12 9:00
$end^b$	12/24 7:00	2/11 10:00	4/2 13:00	4/17 1:00	4/25 16:00	5/6 4:00	5/19 17:00
particulate SO <sub>4</sub>		n/a <sup>c</sup>	<b>√</b>	✓		<b>√</b>	<b>√</b>
particulate Org		n/a <sup>c</sup>	$\checkmark$	$\checkmark$		$\checkmark$	
particulate NO <sub>3</sub>	$\checkmark$	n/a <sup>c</sup>	$\checkmark$			$\checkmark$	
particulate NH <sub>4</sub>	$\checkmark$	n/a <sup>c</sup>	$\checkmark$			$\checkmark$	$\checkmark$
$NO_x$	$\checkmark$					$\checkmark$	✓
$NO_y$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$		$\checkmark$	$\checkmark$
$O_3$	$\checkmark$	$\checkmark$	$\checkmark$			$\checkmark^d$	$\checkmark$
ethylene	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$		$\checkmark$	$\checkmark$
benzene	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$
toluene	✓		✓	$\checkmark$	✓	$\checkmark$	✓

<sup>&</sup>lt;sup>a</sup>A check mark indicates confirmation of concentration rises. <sup>b</sup>Time is in local time. <sup>c</sup>n/a indicates no data available during the episode. <sup>d</sup>Diurnal variation was observed.

### NOAA HYSPLIT MODEL Backward trajectories ending at 2200 UTC 23 Dec 10 GDAS Meteorological Data

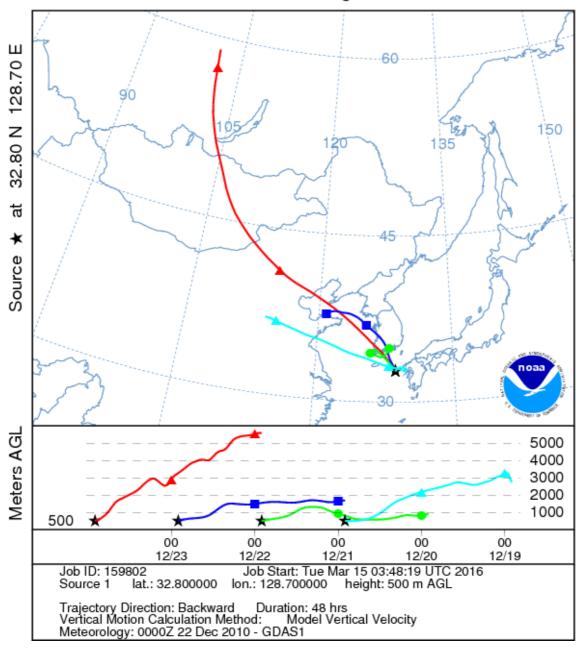


Figure S-7. 48h back trajectories of air masses during the episode 1. See Table S-1 for the detail of episode period.

#### NOAA HYSPLIT MODEL Backward trajectories ending at 0100 UTC 11 Feb 11 GDAS Meteorological Data

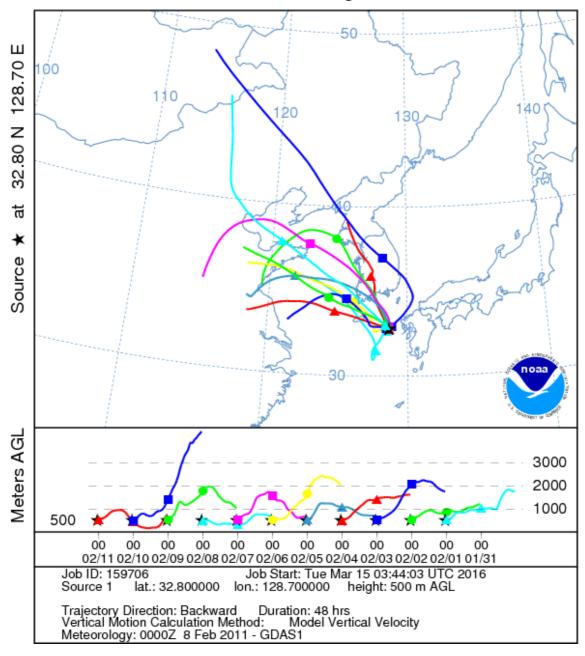


Figure S-8. 48h back trajectories of air masses during the episode 2. See Table S-1 for the detail of episode period.

#### NOAA HYSPLIT MODEL Backward trajectories ending at 0400 UTC 02 Apr 11 GDAS Meteorological Data

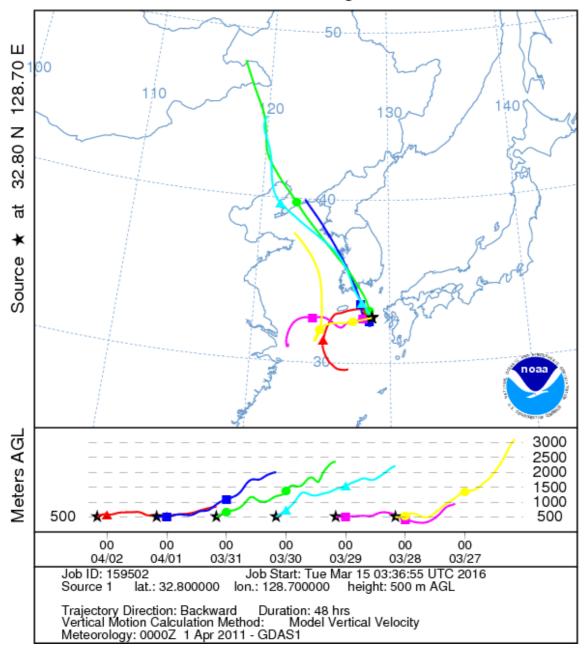


Figure S-9. 48h back trajectories of air masses during the episode 3. See Table S-1 for the detail of episode period.

## NOAA HYSPLIT MODEL Backward trajectories ending at 1600 UTC 16 Apr 11 GDAS Meteorological Data

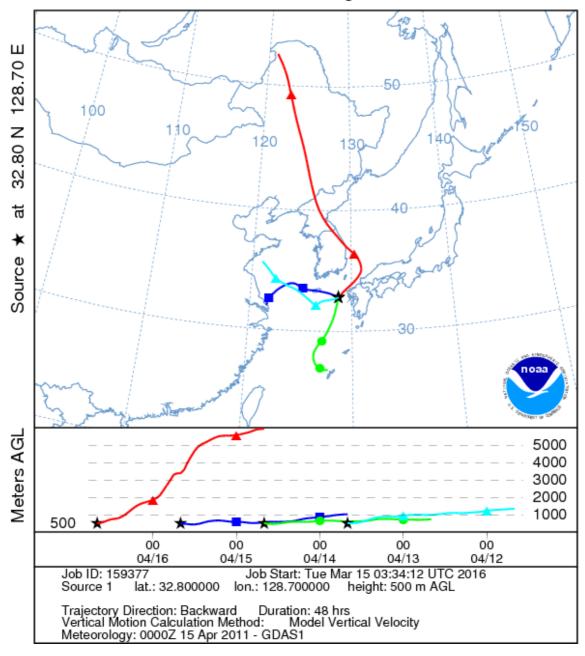


Figure S-10. 48h back trajectories of air masses during the episode 4. See Table S-1 for the detail of episode period.

#### NOAA HYSPLIT MODEL Backward trajectories ending at 0700 UTC 25 Apr 11 GDAS Meteorological Data

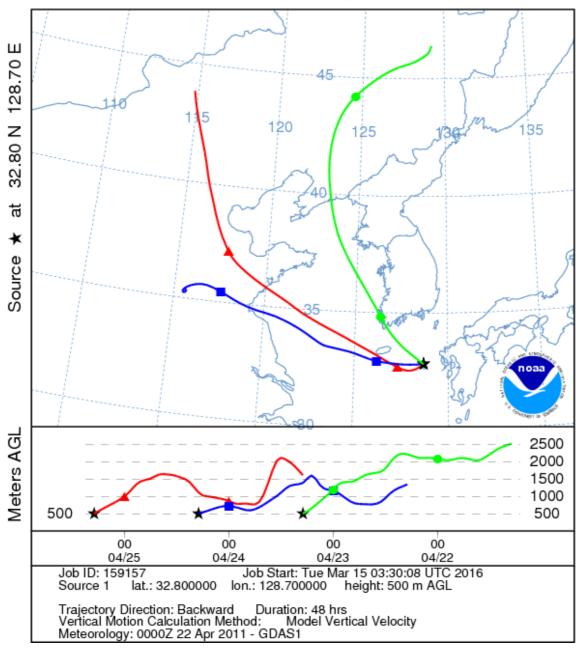


Figure S-11. 48h back trajectories of air masses during the episode 5. See Table S-1 for the detail of episode period.

#### NOAA HYSPLIT MODEL Backward trajectories ending at 1900 UTC 05 May 11 GDAS Meteorological Data

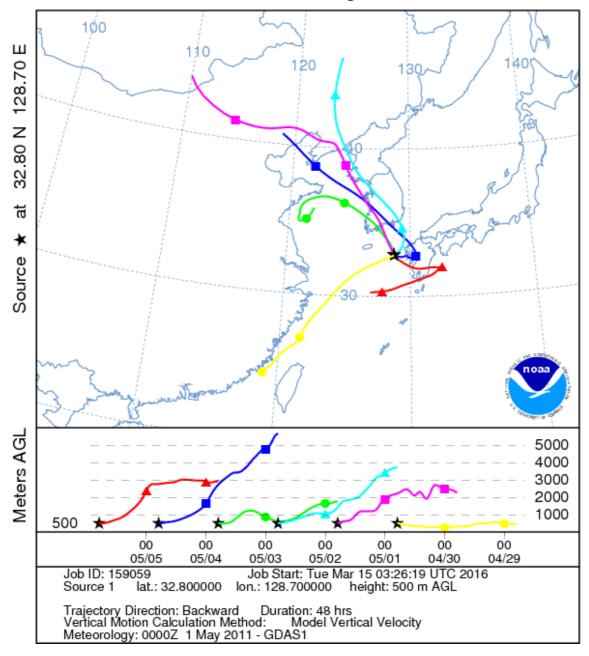


Figure S-12. 48h back trajectories of air masses during the episode 6. See Table S-1 for the detail of episode period.

# NOAA HYSPLIT MODEL Backward trajectories ending at 0600 UTC 19 May 11 GDAS Meteorological Data

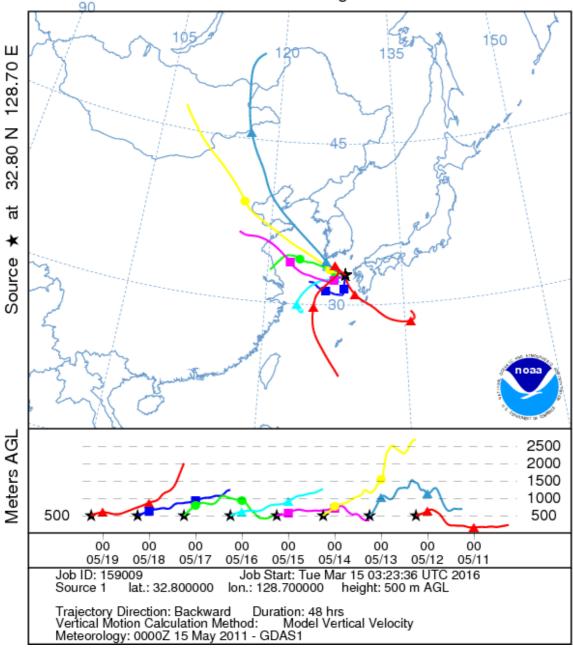


Figure S-13. 48h back trajectories of air masses during the episode 7. See Table S-1 for the detail of episode period.

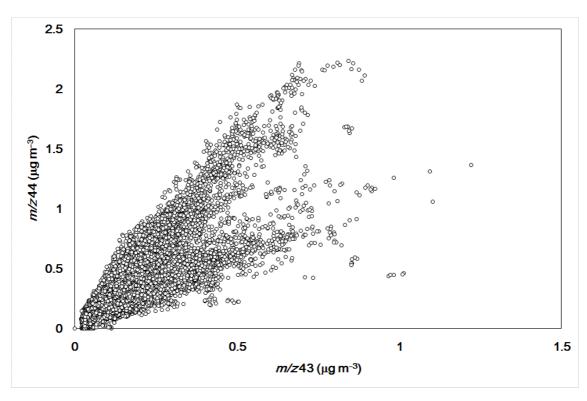


Figure S-14. Scatter plot of hourly averaged m/z 44 concentrations of organics as function of hourly averaged m/z 43 concentrations of organics.

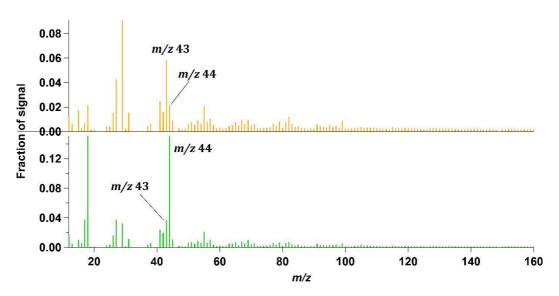


Figure S-15. Extracted mass spectra from two-factorial PMF analysis on organic aerosol mass spectra obtained from January 8 to February 2, 2011: top (orange), mass spectra identified as semi-volatile oxygenated organic aerosol (SV-OOA); bottom (green), mass spectra identified as low-volatile oxygenated organic aerosol (LV-OOA).

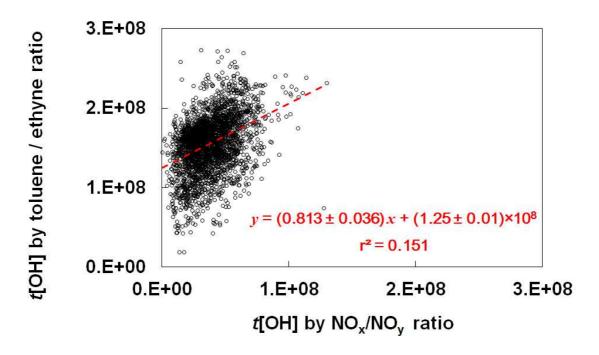


Figure S-16. Scatter plot of t[OH] estimated by  $NO_x/NO_y$  clock and toluene/ethyne clock.