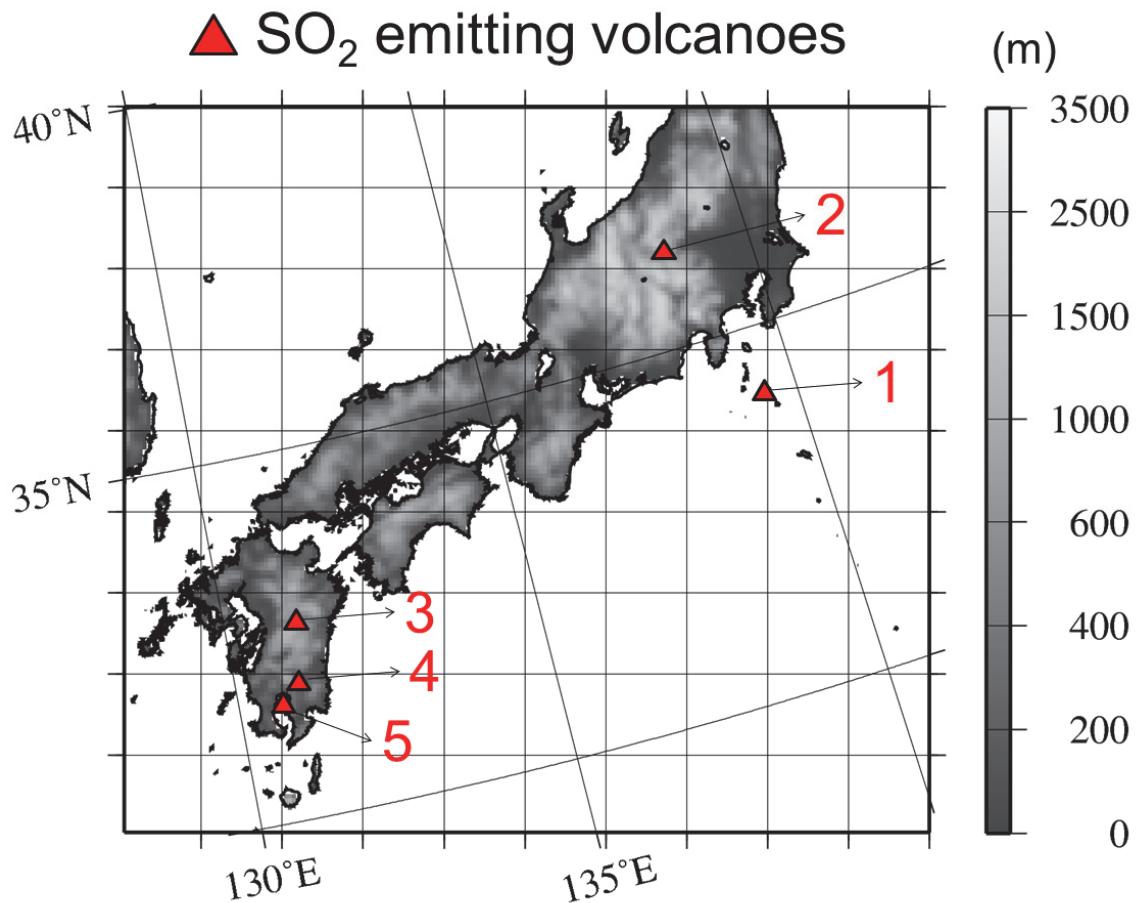


## Supplement 1

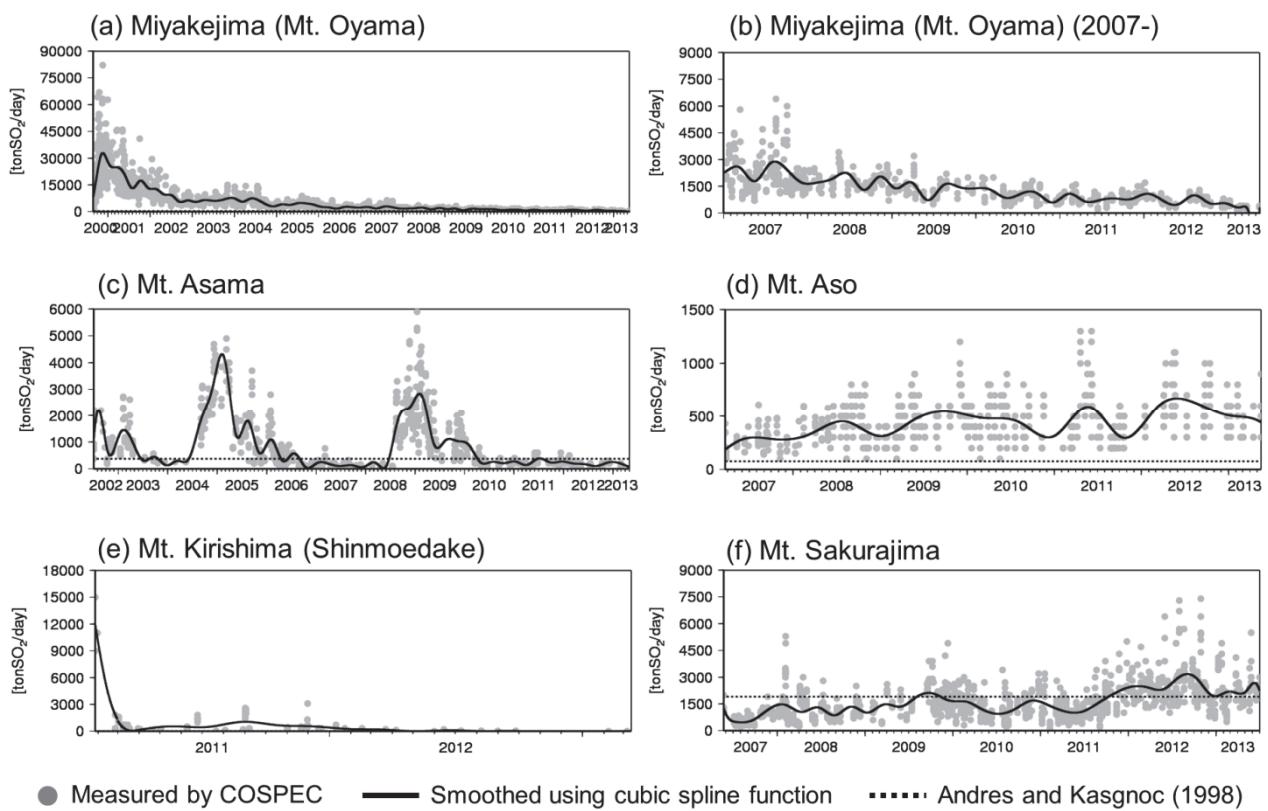
Supplement 1 includes the locations of the five major SO<sub>2</sub>-emitting volcanoes and the time series of temporally smoothed and raw SO<sub>2</sub> emission flux data measured by the JMA and used in the NHM-Chem model.



1. Miyakejima (Mt. Oyama), 2. Mt. Asama,
3. Mt. Aso, 4. Mt. Kirishima, 5. Mt. Sakurajima

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Figure S1-1: Locations of the five SO<sub>2</sub>-emitting volcanoes in Japan monitored by the JMA.

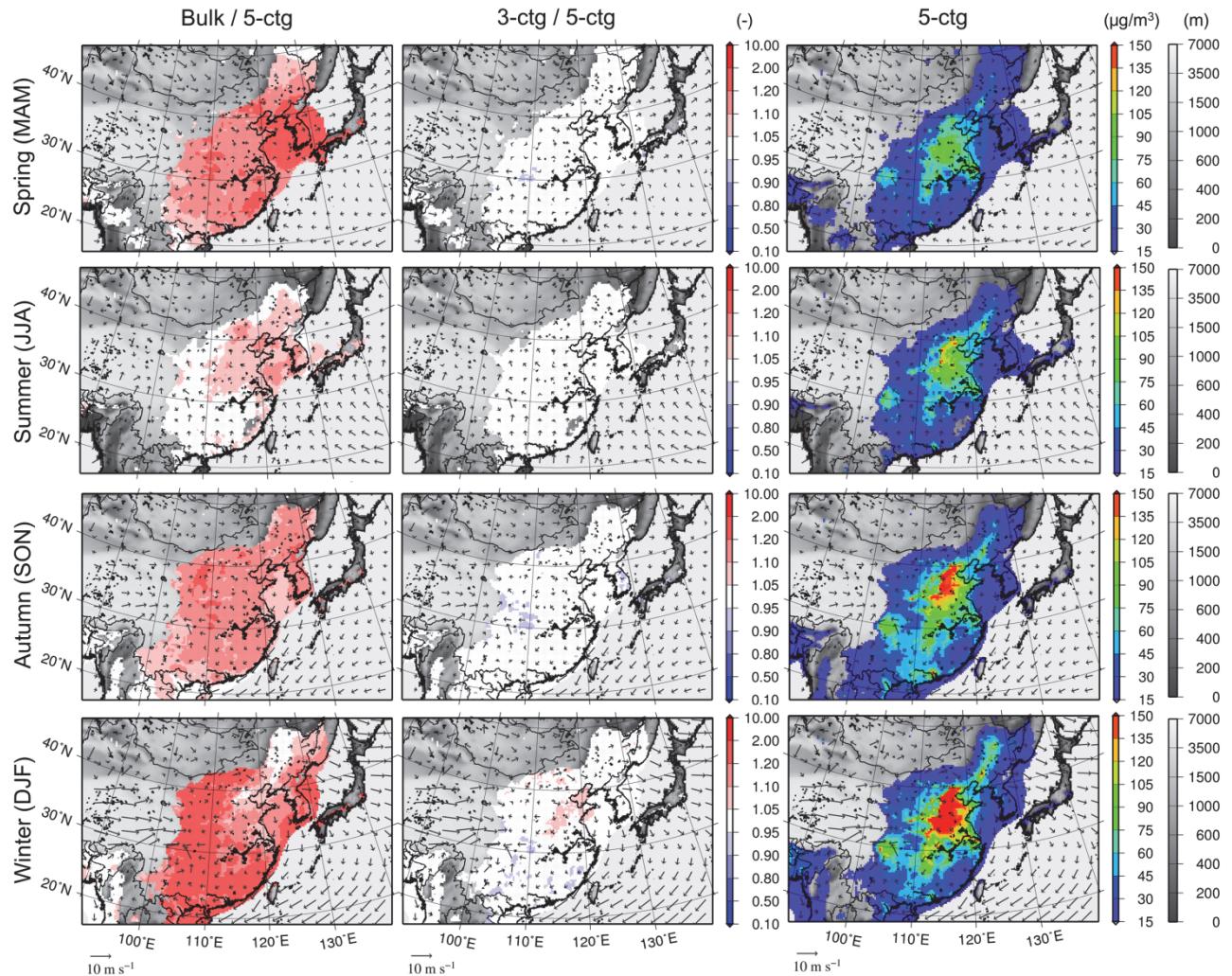


**Figure S1-2:** The time series of temporally smoothed data (solid line), together with the raw data (gray dots) and estimations of Andres and Kasgnoc (1998) (dotted line), of the  $\text{SO}_2$  emission flux, measured by the JMA and used in the NHM-Chem model.

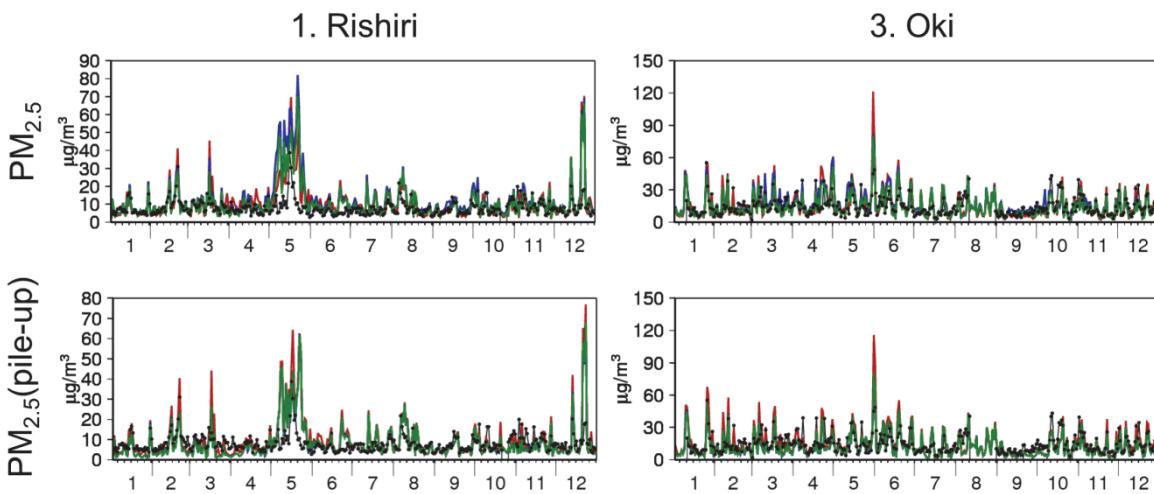
## Supplement 2

**Supplement 2** shows the seasonal mean surface air concentration of PM<sub>2.5</sub> (pile-up), the simulated and observed daily mean surface air concentrations of PM<sub>2.5</sub> and PM<sub>2.5</sub> (pile-up) at Rishiri and Oki, and the statistical analysis used to compare the observed and simulated PM<sub>2.5</sub> and PM<sub>2.5</sub> (pile-up) for the 5-category, 3-category, and bulk methods.

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**Figure S2-1** Same as **Fig. 4** but for PM<sub>2.5</sub> (pile-up).



**Figure S2-2** The simulated (red: bulk, blue: 3-category, green: 5-category) and observed (black) daily mean surface concentrations of (top to bottom) PM<sub>2.5</sub> and PM<sub>2.5</sub> (pile-up) at the Japanese EANET stations of (left to right) Rishiri and Oki in the year 2006.

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**Table S2** Statistical analysis used to compare the observed and simulated PM<sub>2.5</sub> for the 5-category, 3-category, and bulk methods.

Variable	N <sup>a</sup>	Obs. <sup>b</sup>	Med. <sup>b</sup>	Unit	Sim:Obs <sup>c</sup>			R <sup>d</sup>			RMSE <sup>e</sup>			Fa2 <sup>f</sup>		
					5-ctg	3-ctg	blk	5-ctg	3-ctg	blk	5-ctg	3-ctg	blk	5-ctg	3-ctg	blk
PM <sub>2.5</sub> <sup>g</sup>	695	8.83	µg m <sup>-3</sup>		1.06	1.21	1.06	0.59	0.57	0.64	8.69	10.1	9.28	0.81	0.81	0.77
PM <sub>2.5</sub> (pile-up) <sup>g</sup>	695	8.83	µg m <sup>-3</sup>		0.79	0.76	0.88	0.59	0.58	0.63	8.48	8.45	9.66	0.67	0.67	0.74

<sup>a</sup>Number of available data

10 <sup>b</sup>Median of observation data

<sup>c</sup>Simulation to observation median ratio

<sup>d</sup>Correlation coefficient

<sup>e</sup>Root mean square error

<sup>f</sup>Fraction of simulated values within a factor of two of the observed values

15 <sup>g</sup>Daily mean surface air concentration