Table S1. Definitions for statistical metrics used in this study for model evaluations.

Descriptions	Metrics	**Definition***
Mean observation	$\bar{O}$	$(1/\mathrm{N})\sum_{i=1}^{N} O_i$
Mean model	$\bar{M}$	$(1/\mathrm{N})\sum_{i=1}^{N} M_i$
Correlation Coefficient	R	$\frac{\sum_{i=1}^{N}(M_i-\bar{M})(O_i-\bar{O})}{\sqrt{\sum_{i=1}^{N}(M_i-\bar{M})^2}\sqrt{\sum_{i=1}^{N}(O_i-\bar{O})^2}}$
Index Of Agreement	IOA	$1-\frac{\sum_{i=1}^{N}(M_i-O_i)^2}{\sum_{i=1}^{N}(\lvert M_i-\bar{O}\rvert+\lvert O_i-\bar{O}\rvert)^2}$
Root Mean Squared Error	RMSE	$\sqrt{(1/N)\sum_{i=1}^{N}(M_i-O_i)^2}$
Mean Bias	MB	$(1/N)\sum_{i=1}^{N}(M_i-O_i)$
Normalized Mean Bias	NMB	$(1/N)\sum_{i=1}^{N}(M_i-O_i)/\bar{O}\times 100$

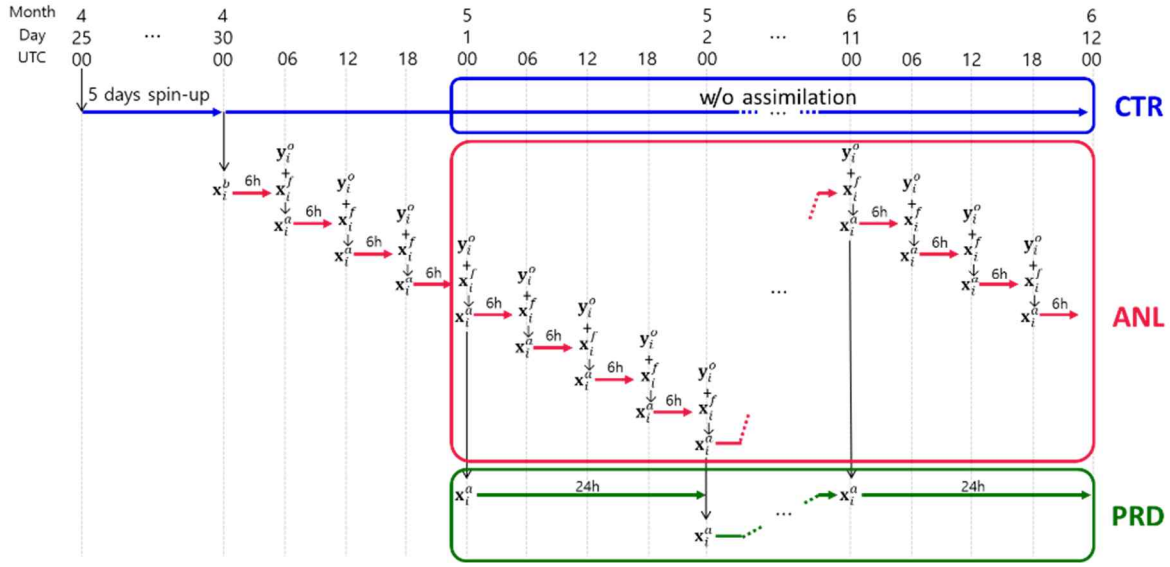


Figure S1. Simulation strategy for data assimilation cycles and 1-day predictions. The case names of CTR, ANL, and PRD indicate the control, reanalysis, and prediction run, respectively.

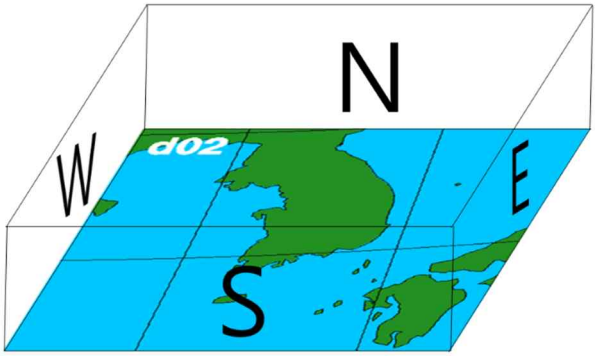
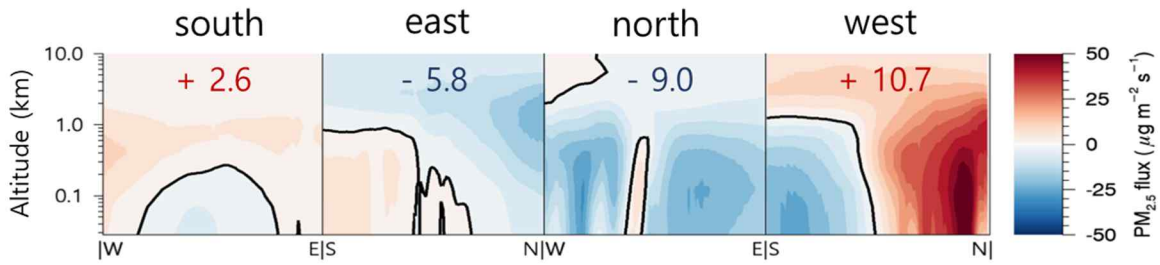
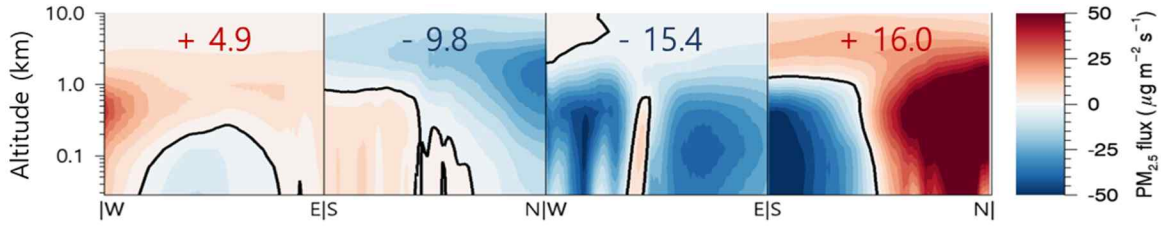


Figure S2. Three-dimensional view of the four lateral boundaries of domain 2 (D2). N, S, E, and W indicate north, south, east, and west planes, respectively.

(a) DA_ic



(b) DA_icbc



(c) Difference (DA_icbc – DA_ic)

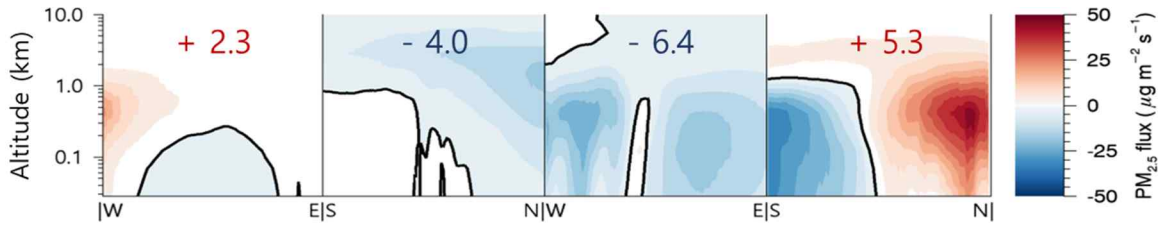


Figure S3. Averaged $PM_{2.5}$ fluxes in the four lateral boundary planes (south, east, north, and west from left to right, refer Fig. S2) for simulation periods along the four perimeters of the fine domain (D2). The thick black lines indicate zero flux. The positive and negative values on each boundary present the plain averaged fluxes. (a) DA_ic, and (b) DA_icbc, are the averaged $PM_{2.5}$ fluxes without, and with, the EnKF data assimilation in China (D1), respectively. The increments in DA_icbc experiment are also presented at the bottom (c). Note that the y-axis for the height is log-scale, to show the results below the planetary boundary layer in detail.