

**Supplementary Figure 1.** Horizontal distributions of the differences in the 30-year annual mean aerosol optical depths (AODs) between the (**a**) piClim-aer, (**b**) piClim-BC, (**c**) piClim-OC, and (**d**) piClim-SO2 experiments and the piClim-control experiment. Note that the color scales are different in each panel.



**Supplementary Figure 2.** Horizontal distributions of the 30-year annual mean (**a**–**c**) midlevel (440–680 hPa) cloud cover (%), (**d**–**f**) low-level (below 680-hPa level) cloud cover (%), (**g**–**i**) liquid water path (g m<sup>-2</sup>), and (**j**–**l**) column-integrated number concentration of cloud droplets ( $10^9 \text{ m}^{-2}$ ) in the piClim-control experiment (left), the piClim-aer experiment (middle), and their differences (right). Note that the color scales are different in each panel.



**Supplementary Figure 3.** Horizontal distributions of the 30-year annual mean (**a**–**c**) high-level (above 440-hPa level) cloud cover (%), (**d**–**f**) ice water path (g m<sup>-2</sup>), and (**g**–**i**) column-integrated number concentration of ice crystals (10<sup>9</sup> m<sup>-2</sup>) in the piClim-control experiment (left), the piClim-aer experiment (middle), and their differences (right). Note that the color scales are different in each panel.



**Supplementary Figure 4.** Vertical distributions of the 30-year annual zonal mean (**a**–**c**) cloud fraction (%), (**d**–**f**) in-cloud ice water content (mg kg<sup>-1</sup>), and (**g**–**i**) in-cloud number concentration of ice crystals ( $L^{-1}$ ) in the piClim-control experiment (left), the piClim-aer experiment (middle), and their differences (right). The solid black lines (left and middle panels) denote temperatures of 0°C and –38°C. The solid black lines (right panels) denote the values shown by the color coding in the piClim-control experiment (left panels). Note that the color scales are different in each panel.



**Supplementary Figure 5.** Horizontal distributions of the differences in the 30-year annual mean number concentrations of the aerosols averaged within the range of 150–420 hPa between the (**a**) piClim-aer, (**b**) piClim-BC, (**c**) piClim-OC, and (**d**) piClim-SO2 experiments and the piClim-control experiment (cm<sup>-3</sup>). The thick black lines denote 30-year annual mean high-level cloud cover of 70% for the corresponding experiments. Note that the color scales are different in each panel.



**Supplementary Figure 6.** Same as Supplementary Figure 5a but for boreal (**a**) winter, (**b**) spring, (**c**) summer, and (**d**) fall (cm<sup>-3</sup>).



**Supplementary Figure 7.** Same as Figure 7b but for boreal (a) winter, (b) spring, (c) summer, and (d) fall  $(10^9 \text{ m}^{-2})$ .



**Supplementary Figure 8.** Vertical distributions of the differences in the 30-year annual global mean (**a**) temperature (K) and (**b**) upward mass flux of cumulus convection ( $g s^{-1} m^{-2}$ ) between the piClim-aer (black), piClim-BC (red), piClim-OC (green), and piClim-SO2 (blue) experiments and the piClim-control experiment.