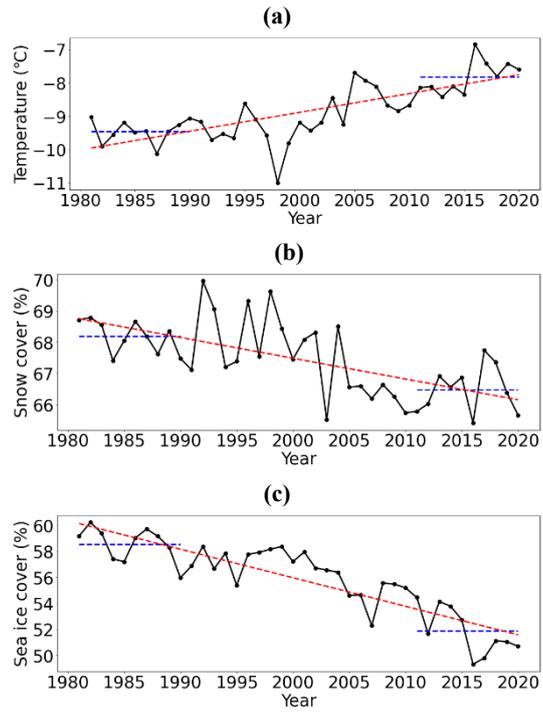
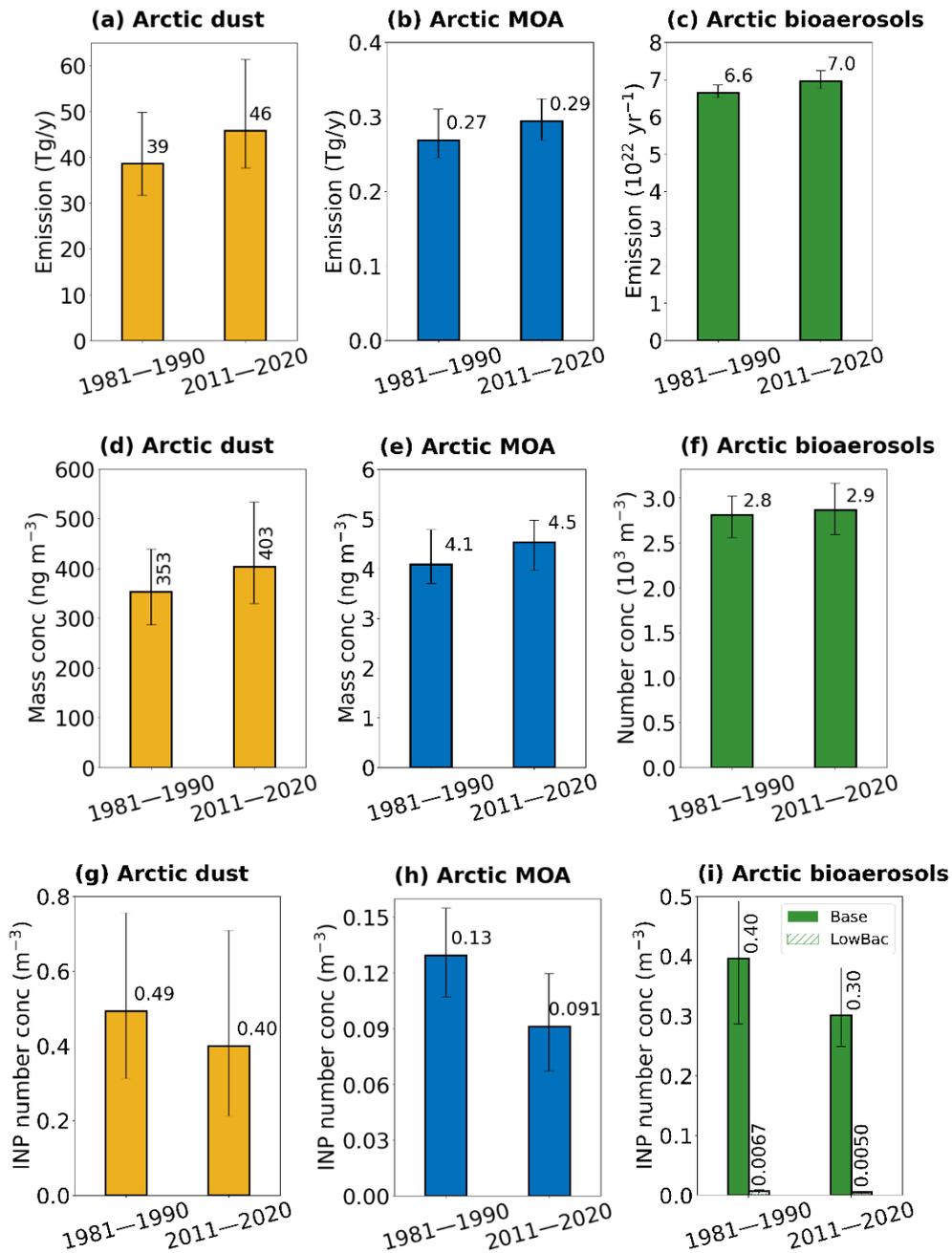


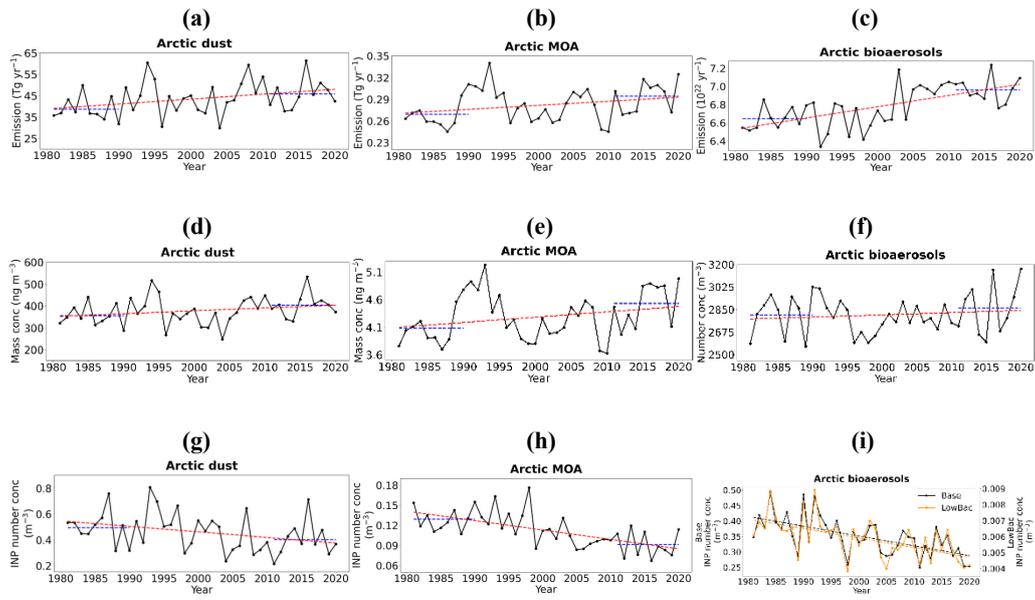
**Figure S1:** Location of observational sites, including Northern Norway (69.93°N, 22.81°E, 905 m a.s.l.), Alert, Canada (82.45°N, 62.51°W, 200 m a.s.l.), Ny-Ålesund, Svalbard (78.91°N, 11.88°W, 475 m a.s.l.), Utqiagvik, Alaska (71.30°N, 156.77°W, 11 m a.s.l.), and Northern Greenland (81.60°N, 16.67°W, 24 m a.s.l.).



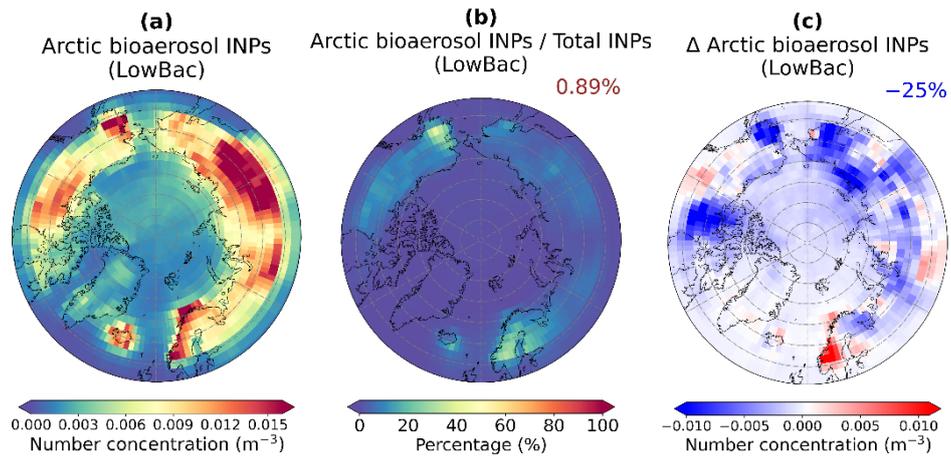
**Figure S2:** Year-to-year variations of (a) surface temperature, (b) snow cover, and (c) sea ice cover from 1981 to 2020 in the Arctic. The dashed blue lines represent the average values for the periods 1981–1990 and 2011–2020. The dashed red lines indicate the temporal trends from 1981 to 2020.



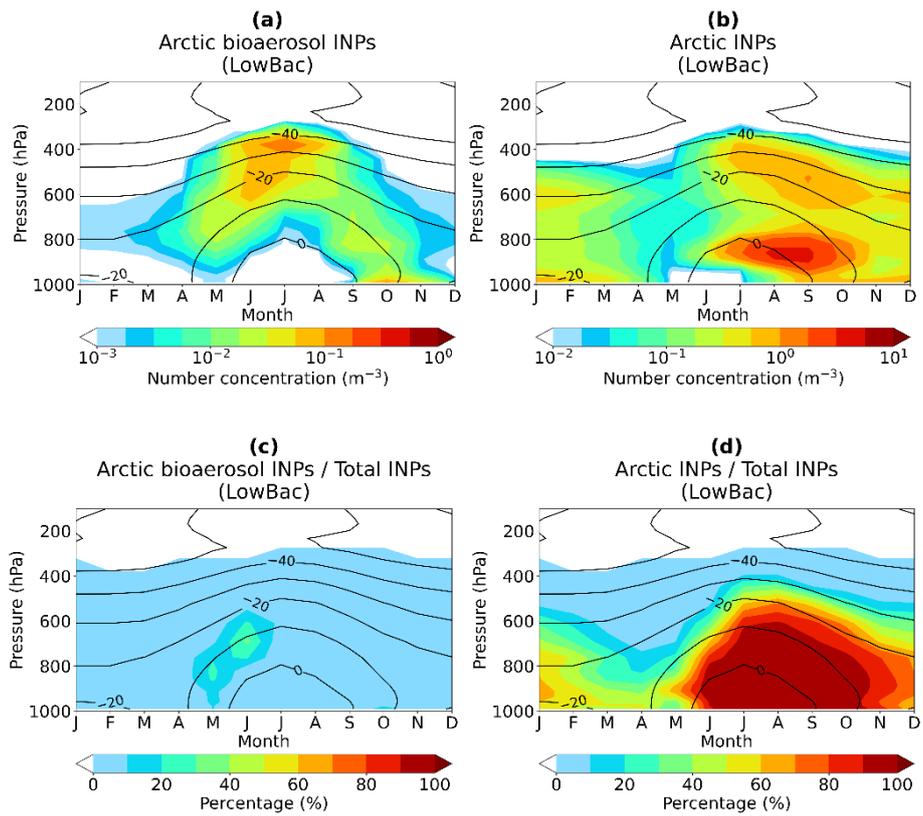
**Figure S3:** (a–c) Emissions, (d–f) atmospheric concentrations, and (g–i) INP number concentrations of Arctic dust, Arctic MOA, and Arctic bioaerosols in the Arctic lower troposphere (> 730 hPa) during 1981–1990 and 2011–2020. In panel (i), solid bars represent the Base simulation results, and hatched bars indicate LowBac results. Error bars indicate interannual variability (minimum–maximum ranges) for each period.



**Figure S4:** Year-to-year variations of Arctic dust, Arctic MOA, and Arctic bioaerosols for (a–c) emissions, (d–f) atmospheric concentrations, and (g–i) INP number concentrations in the Arctic lower troposphere (> 730 hPa), from 1981 to 2020. The dashed blue lines represent the mean values for the periods 1981–1990 and 2011–2020. The dashed red lines (or black and orange in the last panel) indicate the temporal trends from 1981 to 2020.



**Figure S5.** Spatial distributions of (a) INP number concentrations of Arctic bioaerosols and (b) their contributions to total INPs during 1981–2020, as well as (c) the changes in Arctic bioaerosol INPs from 1981–1990 to 2011–2020 in the Arctic lower troposphere ( $> 730$  hPa), for the LowBac simulation.



**Figure S6.** Vertical distributions and temporal variations of (a–b) Arctic bioaerosols INPs and Arctic INPs (sum of INPs from Arctic dust, Arctic MOA, and Arctic bioaerosols), as well as (c–d) their contributions to total INPs in the Arctic during 1981–2020 for the LowBac simulation. Black lines indicate monthly mean Arctic temperatures.