

Figure S1: The relative differences in global seasonally averaged surface SO<sub>2</sub> concentrations between the Improved case and the Original case in 2015 after the incorporation of detailed in-cloud aqueous-phase chemical mechanisms (%). DJF, MAM, JJA and SON represent December-January-February, March-April-May, June-July-August and September-October-November, respectively, the same below.



Figure S2: The global seasonal average ratio of net chemical loss rates of surface SO<sub>2</sub> between the Improved case and the Original case in 2015 after the incorporation of detailed in-cloud aqueous-phase chemical mechanisms.



Figure S3: Scatter plots for monthly average SO<sub>2</sub> concentrations in EU, US, CN and JK in 2015. The black and red points represent the Original and Improved simulations, respectively. Five blue dashed lines indicate 4:1, 2:1, 1:1, 1:2 and 1:4, respectively. "Ori : Obs" and "Imp : Obs" represent the linear regression slopes of "Original case-Observation" and "Improved case-Observation", respectively. The corresponding monitoring networks are (a) EMEP (unit: µg S m<sup>-3</sup>), (b) EPA (unit: ppbv), (c) CNEMC (unit: µg m<sup>-3</sup>) and (d) EANET (unit: ppbv).



Figure S4: Regional monthly-averaged concentration of SO<sub>2</sub> in EU, US, CN and JK in 2015. The black, red and green lines represent the Observed, Original-simulated and Improved-simulated concentrations, respectively. The blue columns represent the relative differences (%) between Original and Improved simulations. The corresponding monitoring networks are (a) EMEP (unit: µg S m<sup>-3</sup>), (b) EPA (unit: ppbv), (c) CNEMC (unit: µg m<sup>-3</sup>) and (d) EANET (unit: ppbv).



Figure S5: The relative differences (%) in global seasonally averaged surface SO<sub>2</sub> concentrations (ppbv) in 2015 with the incorporation of HO<sub>x</sub>-chemistry, Fe-chemistry, N-chemistry and carbonate chemistry individually from top to bottom, respectively.



Figure S6: The differences in global seasonally averaged surface SO<sub>2</sub> concentrations (ppbv) in 2015 after changing the soluble [Fe<sup>3+</sup>] concentration levels from 5  $\mu$ M to 0.1 (top), 1, 20 and 100  $\mu$ M (bottom) with the detailed aqueous-phase chemical mechanisms.



Figure S7: The differences in global seasonally averaged surface SO<sub>2</sub> concentrations (ppbv) in 2015 at different pH values on the basis of the Improved case with the overall detailed in-cloud aqueous-phase chemical mechanisms. The pH values from top to bottom are 3, 4, 5 and 6. [Fe<sup>3+</sup>] is set to 5  $\mu$ M.



Figure S8: The differences in global seasonally averaged surface SO<sub>2</sub> concentrations (ppbv) in 2015 at different pH values after the incorporation of N-chemistry individually (in Section 4). The pH values from top to bottom are 3, 4, 5 and 6, respectively.



Figure S9: The differences in global seasonally averaged surface SO<sub>2</sub> concentrations (ppbv) in 2015 at different pH values after the incorporation of HO<sub>x</sub>-chemistry individually (in Section 4). The pH values from top to bottom are 3, 4, 5 and 6, respectively.



Figure S10: The differences in global seasonally averaged surface SO<sub>2</sub> concentrations (ppbv) in 2015 at different pH values after the incorporation of Fe-chemistry individually (in Section 4). The pH values from top to bottom are 3, 4, 5 and 6, respectively.  $[Fe^{3+}]$  is set to 5  $\mu$ M.