



## **Chemistry–Climate Interactions of Stratospheric and Mesospheric Ozone in EMAC Long-Term Simulations with Different Boundary Conditions for CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, and ODS**

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To evaluate future climate change in the middle atmosphere and the chemistry–climate interaction of stratospheric ozone, we performed a long-term simulation from 1960 to 2050 with boundary conditions from the IPCC A1B greenhouse gas scenario (IPCC, 2007) and the WMO Ab halogen scenario (WMO, 2007) using the chemistry–climate model ECHAM5/MESy Atmospheric Chemistry (EMAC; Joeckel et al., 2006). In addition to this standard simulation we performed five sensitivity simulations from 2000 to 2050 using the rerun files of the simulation mentioned above. For these sensitivity simulations we used the same model setup as in the standard simulation but changed the boundary conditions for carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), and ozone-depleting substances (ODS). In the first sensitivity simulation we fixed the mixing ratios of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O in the boundary conditions to the amounts for 2000. In each of the four other sensitivity simulations we fixed the boundary conditions of only one of CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, or ODS to the year 2000.

In our model simulations the future evolution of greenhouse gases leads to significant cooling in the stratosphere and mesosphere. Increasing CO<sub>2</sub> mixing ratios make the largest contributions to this radiative cooling, followed by increasing stratospheric CH<sub>4</sub>, which also forms additional H<sub>2</sub>O in the upper stratosphere and mesosphere. Increasing N<sub>2</sub>O mixing ratios makes the smallest contributions to the cooling. The simulated ozone recovery leads to warming of the middle atmosphere.

In EMAC the future development of ozone is influenced by several factors. 1) Cooler temperatures lead to an increase in ozone in the upper stratosphere. The strongest contribution to this ozone production is cooling due to increasing CO<sub>2</sub> mixing ratios, followed by increasing CH<sub>4</sub>. 2) Decreasing ODS mixing ratios lead to ozone recovery, but the contribution to the total ozone increase in the upper stratosphere is only slightly higher than the contribution of the cooling by greenhouse gases. In the polar lower stratosphere a decrease in ODS is mainly responsible for ozone recovery. 3) Higher NO<sub>x</sub> and HO<sub>x</sub> mixing ratios due to increased N<sub>2</sub>O and CH<sub>4</sub> lead to intensified ozone destruction, primarily in the middle and upper stratosphere, from additional NO<sub>x</sub>; in the mesosphere the intensified ozone destruction is caused by additional HO<sub>x</sub>. In comparison to the increase in ozone due to decreasing ODS, ozone destruction caused by increased NO<sub>x</sub> is of similar importance in some regions, especially in the middle stratosphere. 4) In the stratosphere the enhancement of the Brewer-Dobson circulation leads to a change in ozone transport. In the polar stratosphere increased down welling leads to additional ozone in the future, especially at high northern latitudes. The dynamical impact on ozone development is higher at some altitudes in the polar stratosphere than the ozone increase due to cooler temperatures. In the tropical lower stratosphere increased residual vertical upward transport leads to a decrease in ozone (Kirner et al., 2014).