Modeling of gravitational separation using global atmospheric tracer transport model

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The process of atmospheric molecules separation depending on their molar masses due to gravity is 'gravitational separation' (GS). Recently, the existence of gravitational separation of the major atmospheric components in the upper troposphere and lower stratosphere was confirmed both experimentally using a precise cryogenic sampler and theoretically by 2-dimensional numerical simulations with SOCRATES model. Here we extend this work by performing a more quantitative analysis using the 3-dimensional model NIES TM with the molecular diffusion parameterization. For that purpose, we consider δ value derived from ${}^{12}C^{16}O_2$ and ${}^{13}C^{16}O_2$ distribution. The modeled δ value is compared to observations and the zonal mean distributions from 2-dimensional model SOCRATES. In comparison with SOCRATES simulation, the NIES model has a number of significant advantages: a three-dimensional transport simulation driven by global JCDAS reanalysis, vertical coordinate at isotropic levels, and use of optimized CO₂ fluxes provided realistic tracer spatial distribution and seasonality. The comparison with measurements shows that the model with the used parameterization is able to reproduce the mean value and the number of small-scale fluctuations that are recorded by balloon-borne observations in the lower stratosphere. The factors affecting the GS are investigated by estimation terms in the molecular diffusion equation. Age of air also estimated in the work is consistent with model inter-comparison from TransCom Age of Air experiment and the age spectrum analysis. We found a strong relationship between age of air and δ value for main climatic zones. Thus modeled GS characteristics complement age of air information for a more comprehensive study of the upper troposphere and lower stratosphere circulation changes and provide useful insights for further model development and tuning.

Keywords: the lower stratosphere, gravitational separation, molecular diffusion, age of air