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Anthropogenic effects on the distribution of minor chemical constituents in the mesosphere/lower thermosphere – A model study

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Abstract

We investigate the influence that rising concentrations of methane, nitrous oxide and carbon dioxide have had upon the chemistry of the mesosphere since 1961. Calculations were performed using our global 3D-model LIMA (Leibniz-Institute Middle Atmosphere), designed for the investigation of the MLT-region (Mesosphere-Lower Thermosphere) and particularly for the extended mesopause region. LIMA utilizes observed tropospheric and lower stratospheric temperature and horizontal wind data up to 35 km altitude by assimilating ECMWF/ERA-40 and ECMWF operational data. Real Lyman-α flux values are employed to determine the variable water vapor dissociation rate. Three different calculations were carried out and analyzed: (1) use of the same annual variation of the model dynamics in the chemical transport model (CTM) for all years according to the dynamics of the solar minimum year 1964 and employment of a realistic growth of the anthropogenic gases; (2) use of constant concentrations of the anthropogenic constituents at the lower border, but employment of the varying model dynamics; (3) the so-called realistic case, which considers both the long-term increase in the anthropogenic minor constituents and the varying dynamics according to LIMA calculations. The analysis of these three cases shows that the dynamics are able to counteract the impact of anthropogenic growth of minor constituents in the upper mesosphere-mesopause-lower thermosphere region in middle to high latitudes in summer. The water vapor mixing ratio increases due to rising methane concentration. The reason for this lies in a positive feedback process of autocatalytic water vapor production. The change in concentration of the minor constituents impacts both the cooling rate and the chemical heating rate. We present the relative and absolute deviations between the solar activity minimum years 1964 and 2008 for the most important minor constituents. We discuss the long-term behavior, particularly of water vapor, with regard to the impact on the NLC-region.

Keywords: Anthropogenic changes; Atmosphere; MLT; CTM

1. Introduction

Measurements of water vapor at high latitudes (Hartogh et al., 2010) have shown that the mesospheric/upper stratospheric water vapor concentration has a pronounced year-to-year variability which does not mirror the increase in anthropogenic methane. Even a decrease in the stratospheric and mesospheric water vapor mixing ratio (at 50–80 km) was observed at ALOMAR (69.29°N, 16.03°E), Norway after 2001 during 11 years of measurements (from 1996 to 2006). As Randel et al. (2006) and Scherer et al. (2008) reported, the Brewer–Dobson circulation in the tropics changed abruptly after 2001. This sudden change also influenced the water vapor distribution in the lower stratosphere. Bittner (2000), Bittner et al. (2002) and Höppner and Bittner (2007) detected a decrease in the planetary wave activity extending into mid-latitudes, and Hoffmann et al. (2011) detected a positive trend for gravity waves (GW) in summer mesopause. On the other hand, the anthropogenic increase in methane concentration in the troposphere gave rise to the conclusion that the water

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vapor concentration in the middle atmosphere had also increased (World Meteorological Organization, 1999; Khalil et al., 1993; Dlugokencky et al., 2003). It was supposed that even the occurrence rate of NLCs (noctilucent clouds) could be influenced by the rising methane concentration (Thomas et al., 1989; Thomas and Olivero, 2001). The height of the NLCs has not noticeably changed since the time of the first observation (von Zahn and Berger, 2003). The term "equithermal submesopause" was introduced in 1996 and reflects the constancy of the mean mesopause temperature during the last 40 years (Lübken et al., 1996). Some years ago, Lübken (2000, 2001) found no significant temperature trend in the upper mesosphere/mesopause region at high latitudes in summer. In the same way, no trend was detected in the annual mean data of temperature derived by airglow measurements at Wuppertal (51°N, 7°E) (Offermann et al., 2004). These findings motivated us to study the mechanisms of stabilization particularly in the summer mesopause region which is astonishingly stable in spite of anthropogenic impacts.

Long-term trend calculations on the basis of the climatologic mean model COMMA-IAP (COlogne Model of the Middle Atmosphere of the Institute of Atmospheric Physics in Kühlungsborn) confirmed a considerable increase in the mesospheric water vapor mixing ratio over the past 120 years (Grygalashvyly and Sonnemann, 2006; Grygalashvyly et al., 2009). However, recently the trend of methane increase seems to have stopped or slowed down considerably (e.g. Dlugokencky et al., 2003, 2009; Khalil et al., 2007; Frankenberg et al., 2011). The discrepancy between water vapor and methane increase gave rise to the conclusion that only a certain portion of the observed increase in the stratospheric humidity could be attributed to the increase in methane. The particular exchange conditions of water vapor between the troposphere and stratosphere essentially influence the middle atmospheric humidity (Forster and Shine, 1999). The general mechanism of the water vapor exchange is an upward transport in the tropical convection zone and downward transport outside of the convection zone in connection with low pressure troughs (e.g. Junge, 1962; Johnson and Viezee, 1981; Holton et al., 1995; Holton and Gettelman, 2001; Fueglistaler et al., 2004; Hegglin et al., 2009, 2010; Randel, 2010; Gettelman et al., 2011). This process takes place particularly in the winter polar vortex. At the tropical tropopause layer, water vapor is subjected to a freeze drying effect. The water vapor mixing ratio decreases to about 4 ppmv at the cold trap. Above the hygropause, the mixing ratio of water vapor increases with increasing height due to the almost complete oxidation of methane in the middle atmosphere and reaches maximum values of 7 to 8 ppmv in the vicinity of the stratopause and occasionally in the middle mesosphere. Methane is not subjected to the freeze drying effect. Consequently, as the hydrogen escape flux is small compared with the flux of hydrogen atoms bound in methane which cross the tropopause, the globally averaged water vapor flux crossing the tropopause is downward directed (Sonnemann and Körner, 2003).

The mesospheric water vapor is controlled by the stratospheric water vapor concentration, but it also depends on the internal mesospheric dynamics changing from year to year. With an anthropogenic increase in methane, the mesospheric water vapor also increased monotonically with a certain time delay of few years. Only the variable solar activity modulates the trend in this case. Particularly in the upper mesosphere and above, the solar activity is increasingly mirrored in the water vapor mixing ratios by clear anti-correlation with the Lyman-α flux (Sonnemann and Grygalashvyly, 2005a).

The plan of this paper is as follows: In Section 2, we briefly introduce the model LIMA (Leibniz-Institute Middle Atmosphere) and the numerical experiments; the results are presented in Section 3; in Section 4, we discuss the results; and, finally, we give a short summary and draw some conclusions in the last chapter.

2. Introduction of the model LIMA and description of the model experiments

We employ the GCM LIMA for the numerical experiment. LIMA is a coupled model of the dynamics and chemistry of the MLT-region. The dynamical fields of the temperature, wind components and pressure calculated in the dynamical part of the model are used in the chemistry transport model (CTM), but the chemical fields are not interactively employed in the dynamical part, meaning all chemical fields used in the dynamical part such as ozone or carbon dioxide and others are based on a respective climatology. This model is briefly described in Berger and Lübken (2006) (dynamics only), Sonnemann et al. (2006, 2008) and Hartogh et al. (2010) (chemistry), as well as in other publications. A more thorough description of the dynamical part of the model is given in Berger (2008).

LIMA is a fully nonlinear global three-dimensional Eulerian grid point model extending from the surface to approximately 150 km with a vertical resolution of 1.1 km. The dynamical part of the model has a completely new architecture, using so-called reduced Gaussian or simplex respectively three-angle coordinates consisting of 41,804 horizontal grid points per layer and a mesh size of approximately 110 km, which helps avoid the pole singularities and the increase in the longitudinal grid point distance toward the equator as is the case with spherical coordinates. The advantage of the 3D-model LIMA lies in the fact that it calculates a dynamical and chemical output belonging to a specific date. The main difference between LIMA and its predecessor COMMA-IAP (Berger and von Zahn, 1999; Körner and Sonnemann, 2001; Sonnemann and Körner, 2003; Hartogh et al., 2004; Sonnemann and Grygalashvyly, 2005a,b, and the quotations therein) is the fact that COMMA-IAP calculates climatologic means, whereas LIMA employs realistic tropospheric and lower stratospheric temperature and horizontal wind