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Ozone chemical equilibrium in the extended mesopause under the nighttime conditions

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Abstract

For retrieval of atomic oxygen and atomic hydrogen via ozone observations in the extended mesopause region (~70–100 km) under nighttime conditions, an assumption on photochemical equilibrium of ozone is often used in research. In this work, an assumption on chemical equilibrium of ozone near mesopause region during nighttime is proofed. We examine 3D chemistry-transport model (CTM) annual calculations and determine the ratio between the correct (modeled) distributions of the O_3 density and its equilibrium values depending on the altitude, latitude, and season.

The results show that the retrieval of atomic oxygen and atomic hydrogen distributions using an assumption on ozone chemical equilibrium may lead to large errors below ~81–87 km. We give simple and clear semi-empirical criterion for practical utilization of the lower boundary of the area with ozone's chemical equilibrium near mesopause.
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1. Introduction

It is hard to deny that atomic hydrogen and atomic oxygen are essential minor chemical constituents in mesosphere-lower thermosphere region because they are chemically active, and involved in thermal and radiative processes of the MLT. Furthermore, they take part in the formation of emission layers. The relation of atomic oxygen with transient luminous events was found recently (Wu et al., 2017). Unfortunately, in situ rocket-borne measurements of O and H are very limited (e.g., Hedin et al., 2009). More commonly, an atomic oxygen and atomic hydrogen distributions are investigated in the MLT by satellite measurements (Russell and Lowe, 2003; Mlynczak et al., 2007, 2013a, 2013b, 2014; Smith et al., 2010; Siskind et al., 2008, 2015). These observations are based on two assumptions concerning: (1) ozone's chemical equilibrium (hereafter OCE) and (2) the main processes entered into the balance equation.

The single reaction of photochemical ozone formation is the three-body reaction of molecular and atomic oxygens $O_2 + O + M \rightarrow O_3 + M$, where M is the air number density. Without a doubt, the main loss process of ozone near

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mesopause region during the night is the reaction with atomic hydrogen, $O_3 + H \rightarrow O_2 + OH$. The reaction of ozone with atomic oxygen amounts to less than 10% of the total ozone loss and can be ignored (Smith et al., 2008). All other common for mesopause ozone balance reactions are even less important. In mesopause region ozone additionally reacts with OH, HO_2, NO, and NO_2. Assuming typical mesopause temperature 298 K one may assess corresponding reaction rates, which are $7.3 \cdot 10^{-14}$, $1.9 \cdot 10^{-15}$, $1.8 \cdot 10^{-14}$, and $3.2 \cdot 10^{-17}$ [cm^3 $molecule^{-1}$ s^{-1}], respectively (Table A.3.1, Brasseur and Solomon, 2005). Multiplying by maximal concentrations at 80–100 km (Table A.6.2, Brasseur and Solomon, 2005) we obtain loss rates $\sim 2.3 \cdot 10^{-7}$, $2.3 \cdot 10^{-9}$, $5.9 \cdot 10^{-7}$, and $3.5 \cdot 10^{-12}$ [s^{-1}], respectively, which are several orders lower than corresponding loss rate of the main reaction $7 \cdot 10^{-3}$ [s^{-1}]. Losses by reactions with Cl, Br, F are even less important due to lack of these chemical compounds in the mesopause (Shimazaki, 1985; Brasseur and Solomon, 2005). Thus, the second assumption is roughly valid for nighttime conditions. An applicability of the OCE assumption, for nighttime conditions is not as obvious, although it is of importance for other applications.

For several decades, OCE assumption has been used to study hydroxyl emission mechanisms, morphology, and variability in the extended mesopause region (Marsh et al., 2006; Xu et al., 2010, 2012; Kowalewski et al., 2014). Kulikov et al. (2006, 2009) proposed methods for the simultaneous retrieval of O, H, HO_2 and H_2O by joint OH and O_3 satellite measurements, where OCE assumption has been utilized. Mlynczak and Solomon (1991, 1993) and Mlynczak et al. (2013b) used this assumption to derive exothermic chemical heat. The OCE assumption was applied in order to study the mesospheric OH^* layer response to gravity waves (Swenson and Gardner, 1998). It is also applied in ultimately theoretical works (e.g. Grygalashvyly et al., 2014; Grygalashvyly, 2015). OCE assumption is used to derive the dependence of excited hydroxyl layer number density and altitude of atomic oxygen and temperature. Sonnemann et al. (2015), used it to analyze annual variations of OH^* layer. Very often this assumption is applied implicitly, when authors are equating the nighttime loss of ozone in reaction with atomic hydrogen and production of ozone by 3-body reaction of molecular and atomic oxygen (e.g., Nikoukar et al., 2007). However, except for several particular cases with rather narrow ranges of coordinates and local times (e.g., Smith and Marsh, 2005), the feasibility of these assumptions depending on time and coordinates has not been proved hitherto. Moreover, current knowledge regarding the chemistry of the MLT suggests that the lower boundary of applicability of the OCE can take place higher than 80 km. First, at 80 km, the concentration of H possesses considerable diurnal photochemical variations and during the night can decrease by approximately one order which leads to the corresponding growth of the characteristic time of ozone (Allen et al., 1984). Second, in the height range between 80 and 90 km, the photochemical system, and consequently ozone evolution, is essentially nonlinear (Konovalov and Feigin, 2000). In particular, in a nighttime phase space of the system, there are areas where characteristic ozone evolution time can be comparable to photochemical lifetime of ozone, which is equal to the invers loss term of ozone (Shimazaki, 1985; Brasseur and Solomon, 2005). From the physical point of view, obviously in such a case the condition of OCE will not be satisfied, and its application for the estimation of O and H via ozone and airglow measurements can lead to larger errors up to several orders. In this current paper we perform a global study of the OCE for the nighttime extended mesopause region based on 3D CTM. We calculate the relationship of "true" O_3 concentration to local equilibrium values O_{3eq} depending on height, latitude, and season. The paper is structured as follows: in the next chapter, the model is described; chapter three presents the results and discussion of our calculations; followed by concluding remarks in the last chapter.

2. Model and calculations

We use for our calculations the global 3D chemistry-transport model (CTM) of the middle atmosphere designed at the Leibniz Institute of Atmospheric Physics (IAP) particularly to study the spatio-temporal phenomena in the MLT region with focus on the extended mesopause region. Model calculates 3D advective and vertical diffusive transport (turbulent and molecular). The grid-point model extends from the ground up to 150 km (118 pressure-height levels). The horizontal resolution amounts to 5.625° latitudinally and 5.625° longitudinally. The chemistry module consists of 19 constituents, 49 chemical reactions and 14 photo-dissociation reactions. The chemical part has been described in numerous papers (e.g., Sonnemann et al., 1998; Körner and Sonnemann, 2001; Grygalashvyly et al., 2009, 2011, 2012). The CTM was validated with measurements, and particularly for ozone, in a number of papers (Hartogh et al., 2004, 2011; Sonnemann et al., 2006a, 2006b, 2007). Three-dimensional fields of the temperature and winds are used from the Canadian Middle Atmosphere Model (CMAM) (de Grandpre et al., 2000; Fomichev et al., 2002; Scinocca et al., 2008). We utilize dynamics and temperature of extended version of CMAM30-SD for year 2000 (http://climate-modelling.canada.ca/climatemodeldata/cmam/output/CMAM−Ext/CMAM30-SD/6hr/atmos/index.shtml).

We calculate the annual variation of the spatio-temporal distributions of the ratio

$$R = O_3/O_{3\,eq} \tag{1}$$

where O_3 are the ozone values calculated by the model, and $O_{3\,eq}$ are the values of ozone in photochemical equilibrium. The values of $O_{3\,eq}$ are calculated as a ratio of production to the reduced loss terms, which take into account all main sources and sinks:

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