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A model study of the plasma chemistry of stratospheric Blue Jets

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ABSTRACT

A numerical plasma chemistry model has been used to simulate the chemical processes in stratospheric Blue Jets. It was applied to Blue Jet streamers in the altitude range 18–38 km. Additionally, the chemical processes in the leader part of a Blue Jet have been simulated for the first time. The model results indicate that there is considerable impact on nitrogen species and ozone. The chemical effects of the streamers predicted by our model are by orders of magnitude larger than in previous model studies. In the leader channel, driven by high-temperature reactions, the concentration of N_2O and NO increases by several orders of magnitude, and there is a significant depletion of ozone.

1. Introduction

Stratospheric Blue Jets (BJs) are upward propagating discharges in the altitude range 15–40 km above thunderstorms. Such events have been observed from aircraft (Wescott et al., 1995, 2001), space (Boeck et al., 1995), and ground (Lyons et al., 2003). They appear as conical bodies of blue light originating at the top of thunderclouds and proceed upward with velocities of the order of 100 km/s. Typically, the diameters of BJs are a few hundred metres at their base and increase with altitude, e.g. Mishin and Milikh (2008).

Early theories related BJs to conventional electric breakdown above charged thunderclouds, and the propagation of streamers of positive (Pasko et al., 1996) or negative (Sukhorukov et al., 1996a) polarity. Also runaway electron breakdown was proposed to cause BJ discharges (Roussel-Dupré and Gurevich, 1996). Petrov and Petrova (1999) suggested that BJs are similar to the streamer zone of a leader (streamer corona). Three-dimensional model studies of streamer coronas in the stratosphere are in good agreement with BJ observations (Pasko and George, 2002). The currently most accepted theory of BJs was proposed by Raizer et al. (2007). It associates BJs to the development of a bi-leader which consists of two leaders of positive and negative polarity growing in opposite directions. Streamers emitted from the upward propagating part of a bi-leader can be sustained by relatively moderate cloud charges (Raizer et al., 2006, 2007, 2010). Streamers are ionised channels which propagate in relatively weak ambient electric fields. They grow due to ionisation caused by the strong electric fields at their tips. In developed BJs, the streamers can travel for a few tens of kilometres predominantly in the vertical direction (Raizer et al., 2006). For more details on the underlying mechanisms of BJs see e.g. Siingh et al. (2012) and Pasko et al. (2012).

The terminal altitude of an upward propagating leader above thunderstorms increases with the electric current carried by the leader (Milikh et al., 2014). From the results of da Silva and Pasko (2013a) it can be concluded that the maximum terminal altitude of a stratospheric BJ is about 30 km. Streamers emitted from leaders at higher altitudes connect to the lower ionosphere, and lead to the development of the so-called Gigantic Jets, for details see e.g. da Silva and Pasko (2013a), Milikh et al. (2014), and references therein.

Electric discharges in the atmosphere are known to have chemical effects. Energetic electrons cause ionisation, dissociation and excitation mainly of the highly abundant N_2 and O_2. As a result, reactive nitrogen and oxygen species are produced which initiate rapid ion-neutral reactions, see e.g. Kossyi et al. (1992). Of particular interest from the atmospheric chemistry point of view is the formation of ozone, and the production of ozone depleting nitric oxide (NO). In recent years, the chemical effects of electric discharges occurring in the mesosphere (so-called sprites) have been investigated in a number of model studies, e.g. Enell et al. (2008), Gordillo-Vázquez (2008),Hiraki et al. (2008), Sentman et al. (2008), Evtushenko et al. (2013), and Parra-Rojas et al. (2013). On the other hand, there are only a few investigations on the chemistry of BJs. However, chemical perturbations due to BJs are of interest as they might influence the stratospheric ozone layer.

To the authors' best knowledge, there are only two publications on the plasma chemistry of BJ streamers: Mishin (1997) has estimated the chemical impact of a single Blue Jet by means of a basic

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nitrogen and oxygen plasma chemistry model. For an altitude of 30 km, the model indicates a 10% increase of nitric oxide, and a 0.5% increase of ozone (peak values within 100 s of model time). Smirnova et al. (2003) used a more detailed chemistry model of 33 species together with modified electron impact rate coefficients to study BJ discharges. Their simulations predict a larger increase of NO, and a smaller relative increase of O_3 compared to the values of Mishin (1997).

The streamer parameters used by Mishin (1997) and Smirnova et al. (2003) were based on an early BJ model (Sukhorukov et al., 1996b). It was pointed out by Mishin and Milikh (2008) that the strength of the streamer tip electric field was unrealistically small. As a result, ionisation, dissociation and excitation processes were significantly underestimated. Therefore, also the calculated chemical effects of BJ streamers might be underestimated.

Furthermore, it is expected that the high temperatures in the leader channel give rise to additional chemical perturbations (Mishin and Milikh, 2008). As far as the authors know, the chemical processes in BJ leaders have not been simulated before.

In this paper, the chemical effects of BJs are investigated by means of a plasma chemistry model. Single BJ streamers are simulated using realistic electric field parameters based on Raizer et al. (2007). Additionally, a first attempt is made to simulate the processes in a BJ leader. For this purpose, leader parameters are estimated from the model results of da Silva and Pasko (2013b). The aim of this study is to investigate the short-term impact of both streamers and leader. The longer-term effects including the mixing of the BJ body with ambient air are not addressed here.

2. Approach

In order to simulate the chemical processes in BJs, a numerical plasma chemistry model is used. It accounts for more than 1000 reactions and calculates the evolution of the 88 species listed in Table 1. The model is similar to the one described in Winkler and Notholt (2014). The only difference of the chemical scheme is that the chlorine ion clusters $Cl^-(H_2O)$, $Cl^-(CO_2)$, and $Cl^-(HCl)$ are not simulated because of the uncertainties concerning these species (Winkler and Notholt, 2013). This is of minor importance for the present study because the negative ion chemistry of the stratosphere is dominated by HCO_3^-, and CO_3^-, NO_3^- and their hydrates, see e.g. Winkler and Notholt (2013), and references therein.

The model considers the electric field driven processes given in Table 2. The reaction rate coefficients have been calculated with the Boltzmann solver BOLSIG+ (Hagelaar and Pitchford, 2005) dependent on the reduced electric field E/N, where E is the electric field strength, and N the number density of air molecules. A detailed description of the model, including the full reaction scheme and the rate coefficients, can be found in Winkler and Notholt (2014).

Table 1
Modelled species.

Negative species
e, O^-,O_2^-, O_3^-,O_4^-,NO^-, NO_2^-, NO_3^-,CO_3^-,CO_4^-,
$O^-(H_2O)$, $O_2^-(H_2O)$, $O_3^-(H_2O)$, OH^-, HCO_3^-, Cl^-, ClO^-

Positive species
N^+, N_2^+, N_3^+, N_4^+,O^+, O_2^+,O_4^+,NO^+,NO_2^+,N_2O^+, $N_2O_2^+$, $NO^+(N_2)$, $NO^+(O_2)$,
H_2O^+, OH^+, $H^+(H_2O)_{n=1-7}$, $H^+(H_2O)(OH)$, $H^+(H_2O)(CO_2)$, $H^+(H_2O)_2(CO_2)$,
$H^+(H_2O)(N_2)$, $H^+(H_2O)_2(N_2)$, $O_2^+(H_2O)$, $NO^+(H_2O)_{n=1-3}$,
$NO^+(CO_2)$, $NO^+(H_2O)(CO_2)$, $NO^+(H_2O)_2(CO_2)$,
$NO^+(H_2O)(N_2)$, $NO^+(H_2O)_2(N_2)$

Neutral species
$N(^4S)$, $N(^2D)$, $N(^2P)$, $O(^3P)$, $O(^1D)$, $O(^1S)$, O_3, NO, NO_2, NO_3,
N_2O, N_2O_5, HNO_3, HNO_2, HNO, H_2O_2,
$N_2(A^3\Sigma_u^+)$, $N_2(B^3\Pi_g)$, $N_2(C^3\Pi_u)$, $N_2(a^1\Pi_g)$, $N_2(a'^1\Sigma_u^-)$, O_2(a), O_2(b),
H_2O, HO_2, OH, OH(v), H,
HCl, Cl, ClO,
N_2, O_2, H_2, CO_2

Table 2
Electric field driven processes in the model.

Ionisation
$e+N_2 \rightarrow N_2^+ + 2e$
$e+N_2 \rightarrow N^+ + N(^4S) + 2e$
$e+N_2 \rightarrow N^+ + N(^2D) + 2e$
$e+O_2 \rightarrow O_2^+ + 2e$
$e+O_2 \rightarrow O^+ + O(^3P) + 2e$

Attachment
$e+O_2 \rightarrow O^- + O(^3P)$

Dissociation
$e+N_2 \rightarrow N(^4S)+N(^4S)+e$
$e+N_2 \rightarrow N(^4S)+N(^2D)+e$
$e+N_2 \rightarrow N(^4S)+N(^2P)+e$
$e+O_2 \rightarrow O(^3P)+O(^3P)+e$
$e+O_2 \rightarrow O(^3P)+O(^1D)+e$
$e+O_2 \rightarrow O(^3P)+O(^1S)+e$

Excitation
$e+N_2 \rightarrow N_2(A^3\Sigma_u^+) + e$
$e+N_2 \rightarrow N_2(B^3\Pi_g) + e$
$e+N_2 \rightarrow N_2(a'^1\Sigma_u^-) + e$
$e+N_2 \rightarrow N_2(a^1\Pi_g) + e$
$e+N_2 \rightarrow N_2(C^3\Pi_u) + e$
$e+O_2 \rightarrow O_2(a)+e$
$e+O_2 \rightarrow O_2(b)+e$

Detachment
$O^- + N_2 \rightarrow N_2O + e$

2.1. Initialisation

The model is applied at seven altitude levels between 18 km and 38 km, the typical BJ altitude range. It is initialised with results (pressure, temperature, and trace gas concentrations) of the two-dimensional atmospheric chemistry and transport model B2dM (Winkler et al., 2009) for mid-solar activity conditions. For more information on the B2dM and its performance see Funke et al. (2011), and references therein. The model was applied to tropical conditions (latitude 10°N; March 1) as Blue Jets might be favoured in the tropics in comparison with mid-latitudes (Pasko et al., 2002).

After initialisation, the model is run for five days without electric fields applied in order to achieve a stable (repeating) diurnal cycle of all chemical species. Loss and production terms have been added to the model to avoid a slow drift of the model due to missing atmospheric transport processes. Fig. 1 shows the pre-breakdown profiles of O_3, NO, and NO_2 which are of interest for the BJ simulations. While there are considerable differences between the night-time and day-time values of the nitrogen species, the diurnal changes of ozone in the considered altitude range are small.

In a next step, the model is used to calculate the pre-breakdown electron and ion composition, i.e. the steady state concentrations due to galactic cosmic ray ionisation. This is the same approach as in Winkler and Notholt (2014). It provides the atmospheric background profiles of electrons and ions for the BJ simulations. The pre-breakdown concentration of the free electrons (seed electrons) is shown in Fig. 2. Due to photo-electron detachment, the day-time concentration of free electrons is higher