

# Evaluation of processes that affect the photochemical timescale of the sodium layer

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## Abstract

We apply the method of the eigenvalue and eigenvector analysis (EEA method) to analyze the photochemical timescale of the sodium layer chemical system to perturbations. The results show that the timescales of three important sodium species, Na, Na<sup>+</sup> and NaHCO<sub>3</sub>, vary strongly with altitude and are significantly different during day and night. For these three species, the EEA method gives a photochemical timescale that differs strongly from the traditional lifetime, defined as the inverse of the photochemical loss rate. For other chemical species in the sodium layer, the two methods of predicting chemical lifetime give virtually identical results. Tests of the sensitivity of the photochemical timescale to changes of the background conditions indicate that the photochemical timescale of Na is sensitive to the changes of the background densities of O<sub>3</sub>, H, and O.

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## 1. Introduction

Sodium resonance lidar is a powerful tool for probing the mesopause region because of its very high vertical and temporal resolution. In addition to providing information about atmospheric density and temperature, the lidar signals give sodium density variations that can be used to infer transport. Because the sodium layer is a photochemical–dynamical coupled system, knowledge of the dynamical and photochemical timescales can be an important piece of information for interpretation of perturbations to the layer. Xu and Smith (2003) used the method of eigenvalue and eigenvector analysis (EEA method) to calculate the timescale for the photochemical

response of sodium to perturbations. The results indicated that the effective photochemical timescale is more than a day in the vicinity of the mesospheric sodium layer and is much longer than the traditionally defined chemical lifetime for atomic sodium of a few minutes.

At the same time that ongoing lidar measurements are improving our knowledge about the sodium layer and its variations, new laboratory measurements are helping to quantify the chemical system. The laboratory research indicates that some new photochemical reactions are important and must be included in chemical models of the sodium layer.

Self and Plane (2002) measured the photolysis cross-section of NaHCO<sub>3</sub>, which is believed to be a major sodium reservoir species. They found that the cross-section below 240 nm is sufficiently large that it cannot be ignored. This change would lead to a large increase in

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atomic sodium around 85 km during the daytime and a very large diurnal variation below 90 km. However, observations show that the diurnal variation of the sodium layer is small (Gibson and Sandford, 1972; Clemesha et al., 1982; States and Gardner, 2000). Plane (2004) proposed the permanent removal of  $\text{NaHCO}_3$  by the polymerization process in his new time-resolved model of the sodium layer. The dimerization leads to low density of  $\text{NaHCO}_3$  in the 75–85 km region and, therefore, the model no longer predicts unrealistically large diurnal variations of sodium. The results of Plane's new chemical model of the sodium layer are consistent with the averaged observed distribution of atomic sodium.

When the new photochemical processes of  $\text{NaHCO}_3$  photolysis and dimerization are included, there is a fundamental change in the chemistry of Na layer. Without these reactions, the accepted chemical model assumes that gas-phase chemistry is fast compared to the timescale of injection of sodium from meteoric sources and its redistribution through vertical transport and diffusion. In this case, the sodium is distributed among the chemical species by photochemical reactions and the chemical system is closed. However, the new chemistry proposed by Plane is an open system. According to this model, the sodium layer is supplied by the injection of Na from space and depleted by the dimerization of  $\text{NaHCO}_3$ . Transport, diffusion and photochemical reactions account for the actual structure and variability of sodium species in this system.

In this paper, we investigate how the proposed new chemical system affects the sodium photochemical timescale calculated by the EEA method. Section 2 presents the new photochemical model of the sodium layer. The timescale of every chemical species in the sodium layer system is given. Section 3 shows the sensitivity of the calculated photochemical timescales to changes in the concentrations of background chemical species. Conclusions are given in Section 4.

## 2. The photochemical timescale of the sodium layer

### 2.1. The time-dependent photochemical model of Na layer

Plane (2004) presented a new time-resolved model of the mesospheric Na layer based on recent laboratory studies. The important changes are inclusion of photolysis of  $\text{NaHCO}_3$  and of an associative self-reaction (dimerization) of that molecule whose product is chemically inactive. The new chemical system of the sodium layer becomes an open system; in other words, the distribution of mesospheric reactive sodium species cannot be understood without taking into account sources and sinks from outside the system. These external sources and sinks consist of the Na input

flux from space and the permanent removal of reactive sodium by the formation of the stable dimer ( $\text{NaHCO}_3$ )<sub>2</sub>.

The dimerization and photolysis reactions of  $\text{NaHCO}_3$  have been added to the photochemical model of the sodium layer of Xu and Smith (2003, 2004) using the coefficients determined by Self and Plane (2002) and Plane (2004). In addition, the ion chemistry in the model has been updated and expanded to better simulate the photochemistry in the topside of the layer (Collins et al., 2002; Cox and Plane, 1998). The model includes 12 sodium species: Na, NaO, NaO<sub>2</sub>, NaO<sub>3</sub>, NaOH, NaCO<sub>3</sub>, NaHCO<sub>3</sub>, Na<sup>+</sup>, NaN<sub>2</sub><sup>+</sup>, NaO<sup>+</sup>, NaCO<sub>2</sub><sup>+</sup> and NaH<sub>2</sub>O<sup>+</sup>. Table 1 lists the photochemical reactions.

The sodium layer chemistry is affected by the background neutral chemical constituents, the temperature profile and the background ion species in the ionospheric E and D regions. The background neutral chemical constituents, such as O<sub>3</sub>, O, and H, which have large effects on the distribution of sodium layer, are calculated by the middle atmospheric photochemical model of Xu et al. (2000, 2003). The model is a time-dependent 1-dimensional model that simulates the diurnal variation of the distributions of neutral chemical species. The background ion species in the ionospheric E and D regions are specified from the International Reference Ionosphere model (IRI-95) (available from <ftp://nssdcftp.gsfc.nasa.gov/models/ionospheric/iri/iri95/>) for the appropriate conditions and time. The vertical temperature profile is taken from NRLMSISE-00 (available from <http://nssdc.gsfc.nasa.gov/space/model/atmos/nrlmsise00.html>). The complete sodium layer chemical model couples together the background middle atmosphere chemical model, the ionosphere model and the sodium layer chemistry model. It is 1-dimensional and time-dependent and accounts for the variation of solar zenith angle during daylight hours.

Fig. 1 gives the simulated profiles of some sodium species at midnight and midday for 45°N during the Vernal Equinox after a model integration of 20 days. The figure shows that the peak sodium density at midnight is about 3000 cm<sup>-3</sup> at the altitude of about 92 km. At midday, the altitude of atomic sodium density peak moves to about 91 km. Below 90 km, there is no obvious diurnal variation in the atomic sodium density. However, the density of  $\text{NaHCO}_3$  at midnight is larger than at midday: a result of the diurnal variation of photolysis. Above the atomic sodium layer around 90 km, ion chemistry predominates. Because the plasma density at midday is much larger than at midnight in the ionospheric E and D regions, the density of Na<sup>+</sup> is larger during day. This leads to a diurnal variation in the density of atomic sodium on the topside of the sodium layer (Clemesha et al., 1982).

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