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ACE-FTS ozone, water vapour, nitrous oxide, nitric acid, and carbon monoxide profile comparisons with MIPAS and MLS



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ABSTRACT

The atmospheric limb sounders, ACE-FTS on the SCISAT satellite, MIPAS on ESA's Envisat satellite, and MLS on NASA's Aura satellite, take measurements used to retrieve atmospheric profiles of O₃, N₂O, H₂O, HNO₃, and CO. Each was taking measurements between February 2004 and April 2012 (ACE-FTS and MLS are currently operational), providing hundreds of profile coincidences in the Northern and Southern hemispheres, and during local morning and evening. Focusing on determining diurnal and hemispheric biases in the ACE-FTS data, this study compares ACE-FTS version 3.5 profiles that are collocated with MIPAS and MLS, and analyzes the differences between instrument retrievals for Northern and Southern hemispheres and for local morning and evening data. For O₃, ACE-FTS is typically within $\pm 5\%$ of mid-stratospheric MIPAS and MLS data and exhibits a positive bias of ~ 10 to 20% in the upper stratosphere – lower mesosphere. For H₂O, ACE-FTS exhibits an average bias of -5% between 20 and 60 km. For N₂O, ACE-FTS agrees with MIPAS and MLS within -20 to $+10\%$ up to 45 km and 35 km, respectively. For HNO₃, ACE-FTS typically agrees within $\pm 10\%$ below 30 km, and exhibits a positive bias of ~ 10 to 20% above 30 km. With respect to MIPAS CO, ACE-FTS exhibits an average -11% bias between 28 and 50 km, and at higher altitudes a positive bias on the order of 10% ($> 100\%$) in the winter (summer). With respect to winter MLS CO, ACE-FTS is typically within $\pm 10\%$ between 25 and 40 km, and has an average bias of -11% above 40 km.

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1. Introduction

Satellite remote sounding of the Earth's limb is currently the only method of observing the atmosphere that allows for

near-global time series of atmospheric profiles from the upper troposphere to the lower thermosphere. However, each atmospheric limb sounder has its own sources of uncertainty and systematic biases. To get a true understanding of the state of the global atmosphere, these uncertainties and biases must be identified and characterized. Limb sounding instruments can exhibit different systematic differences from similar instruments depending on

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the observed latitudinal region and/or the observed local time. These biases must also be identified and characterized.

The Atmospheric Chemistry Experiment – Fourier Transform Spectrometer (ACE-FTS) instrument [1] on the SCISAT satellite is a solar occultation limb sounder that has extensive measurement overlap, both spatially and temporally, with the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument [2,3] on the Envisat satellite and the Microwave Limb Sounder (MLS) [4] on the Aura satellite. There are many hundreds of coincident measurements, in both the Northern and Southern hemispheres, covering the local morning and local evening, between ACE-FTS and both MIPAS and MLS, which are needed for determining any hemispheric and/or diurnal biases between the data sets. These instruments were chosen not only because they have extensive temporal overlap, but because they also retrieve many of the same atmospheric species. The five species they have in common are ozone, water vapour, nitrous oxide, nitric acid, and carbon monoxide. A complete overview of the roles of each of these important species in the different regions of the atmosphere is given by [5], and what follows is a brief synopsis.

Ozone (O_3) is one of the most important atmospheric constituents, as it absorbs harmful solar UV radiation, shielding living organisms on the Earth's surface. Ozone also plays a minor role as a greenhouse gas in the troposphere. Continuous monitoring of vertically-resolved O_3 is therefore vital for understanding O_3 depletion, long-term O_3 recovery, and, to a lesser extent, climate change.

Water vapour (H_2O) is the most important non-anthropogenic greenhouse gas in the Earth's atmosphere, and as such, it has a major influence on the lower atmospheric climate, chemistry, and energy budget. In the middle atmosphere, due to its long lifetime, it is often used as a dynamical tracer. The Brewer–Dobson circulation transports H_2O -rich air from tropospheric low latitudes to stratospheric mid latitudes. Water vapour is also produced in the middle atmosphere via the oxidation of CH_4 and is destroyed via photodissociation as well as via reactions with $O(^1D)$. The middle atmospheric sources and sinks tend to balance near the stratopause region, leading to a climatological H_2O peak in this region.

Nitrous oxide (N_2O) is a minor greenhouse gas, and in the stratosphere it is often used as a dynamical tracer. Moreover, it is the primary source of NO_x in the stratosphere. Stratospheric N_2O typically originates from the surface from different emission sources—agricultural, industrial, biomass burning, etc.—and is distributed throughout the middle atmosphere via the Brewer–Dobson circulation. Recently, it has been discovered that energetic particle precipitation in the upper atmosphere also produces N_2O , which can be transported down into the winter upper stratosphere – lower mesosphere (USLM) [6,7]. Once in the mid stratosphere, N_2O is destroyed by photodissociation and through reactions with $O(^1D)$, producing NO , which catalytically destroys O_3 .

Nitric acid (HNO_3) can remove NO_y from the stratosphere. The main production mechanism for HNO_3 is the three-body reaction between NO_2 , OH , and an air molecule, and the main destruction mechanisms are photolysis

in the UV and through reactions with OH . In the polar winter, N_2O_5 and $ClONO_2$ can react with H_2O or HCl on the surface of polar stratospheric cloud (PSC) particles, producing HNO_3 and chlorine molecules, which then lead to active chlorine species. HNO_3 is typically sequestered by the PSC particles, and subsequent sedimentation of the HNO_3 -containing particles can effectively remove NO_y from the stratosphere.

Carbon monoxide (CO) is a major pollutant and short-lived greenhouse gas in the troposphere. The main sources of CO in the lower atmosphere are surface emissions from fossil fuel combustion and biomass burning, and from hydrocarbon oxidation. In the stratosphere, CH_4 oxidation is the major source of CO and is more effective during sunlit hours, as the two main pathways to creating CO via CH_4 oxidation involve photolysis. In the USLM CO_2 photodissociation directly produces CO , and winter descent from the upper atmosphere also transports in CO -rich air. In the mesosphere and lower thermosphere, where CO is often used as a dynamical tracer, CO is primarily produced via CO_2 photodissociation. Throughout the atmosphere, CO is predominantly lost through chemical reactions with OH .

The following section gives an overview of the ACE-FTS, MIPAS, and MLS instruments, as well as the retrieval algorithms used to derive the data sets analyzed in this study, and Section 3 discusses the methodology used to compare the atmospheric data sets. Section 4 discusses the comparison results and the differences within those results due to diurnal and hemispheric biases. A summary is provided in the final section.

2. Instrumentation

2.1. ACE-FTS on SCISAT

The ACE-FTS instrument is a solar occultation Fourier transform spectrometer operating in the 750 to 4400 cm^{-1} spectral region, with a 0.02 cm^{-1} spectral resolution. It was launched into a high-inclination orbit in August 2003, and since February 2004, ACE-FTS has been providing temperature, pressure, and volume mixing ratio (VMR) profiles of over 30 atmospheric trace gases and over 20 subsidiary isotopologues. Twice per orbit, during sunrise and sunset, ACE-FTS takes measurements approximately every 2 s. Profiles are measured between ~5 and 150 km, with a vertical field of view of ~3 to 4 km and a vertical sampling of ~2 to 6 km, depending on the angle between the satellite's orbital plane and the look direction to the sun.

The ACE-FTS trace species VMR retrieval algorithm is described by [8], and the changes for the most recent version of the retrieval, version 3.5 (v3.5), are detailed by [9]. The retrieval algorithm uses a non-linear least-squares global-fitting technique that fits the ACE-FTS observed spectra in given microwindows to forward modeled spectra—based on line strengths and line widths from the HITRAN 2004 database [10] (with updates as described by [9]). The pressure and temperature profiles used in the forward model are the ACE-FTS derived profiles, calculated by fitting CO_2 lines in the observed spectra. The main

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