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RADIOMETRY OF ATMOSPHERIC CHEMICAL COMPOSITION FROM SOFIA

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ABSTRACT

Microwave radiometers are widely used to determine the vertical distribution of trace gases in the atmosphere. The Airborne Submillimeter Radiometer (ASUR), a 604-662 GHz sensor is designed to be operated on a high flying aircraft. An atmospheric application of ASUR or other microwave sensors proposed on SOFIA (preferrably on campaign basis) will include investigations of the mesosphere, the stratosphere and the UT/LS. One focus will be the study of the chemical composition and changes of lower mesospheric constituents, like O_3, HCN, HO_2, and lower thermospheric NO. In particular, during aurora the variability in NO would be an important objective. To measure the radical OH a different sensor, e.g. at 2.5 THz, has to be utilized. In the stratosphere ASUR is capable of measuring vertical profiles of trace gases related to the ozone chemistry, like ozone, HCl, ClO, and HNO_3 as well as dynamical tracers like N_2O and CH_3Cl. This makes it suitable to target numerous questions related to chemistry and dynamics of the stratosphere, including process studies, e. g. at the vortex edge, but also studies of the stratospheric circulation and possible long-term trends. Trace gas measurements by ASUR can also serve as a long-term validation of several satellite instruments over a large latitudinal range. To study the upper troposphere/lower stratosphere region new instrumentation in limb viewing geometry with frequency bands between 180 and 350 GHz is proposed.

INTRODUCTION

Passive microwave radiometry is a well established instrumental technique in astronomy and atmospheric sciences to measure molecular abundances remotely. However, atmospheric measurements are limited by continuum absorption primarily caused by water vapor which decreases with altitude. Only at lower frequencies (< 350 GHz) some atmospheric windows exist suitable for observations from ground. SOFIA which has a cruising altitude of 12-14 km enables stratospheric and mesospheric measurements even at submillimeter wavelenghts where important molecules have rotational lines. Vertical distributions of the trace gases are obtained from the pressure broadening of these thermally excited molecular lines.

The Airborne Submillimeter Radiometer (ASUR) has been flown onboard the DLR-FALCON and the NASA-DC8 research aircraft during most of the international campaigns concerning atmospheric chemistry associated with stratospheric ozone depletion and satellite validation, such as the SAGE III Ozone Loss and Validation Experiment (SOLVE), and the SCIAMACHY Validation and Utilization Experiment (SCIA-VALUE). It also participated in the LEONID Multi-Instrument Aircraft Campaign (LEONID MAC) 2002 observing mesospheric ozone, NO, and HCN.

THE ASUR INSTRUMENT

ASUR is a passive microwave receiver that operates in the 604 to 662 GHz frequency range using low noise SIS mixer technology (Küllmann et al., 2001). Sounding the atmosphere in an uplooking geometry at 78 degrees zenith angle, the sensor enables observations of the vertical distribution of atmospheric trace gases like ozone, ClO, HCl, N_2O, and HNO_3. The spectra also show signatures of NO, HCN, H_2O, HO_2, CH_3Cl, H_2CO, and BrO. Switching between molecules can be done within 1-2 minutes. For a profile retrieval the Optimal Estimation Method (Rodgers, 1976) is used by means of the pressure broadening of the rotational line spectra. A vertical resolution of 5-10 km in the lower and 10-20 km in the upper stratosphere and lower mesosphere is obtained. The altitude ranges from 15 to approximately 50 km with a horizontal resolution of 12-30 km depending on the molecule. Optionally, for some molecules, like ozone, the altitude may be extended to 65 km using an additional high resolution spectrometer. Above 65 km information on the partial column is available.

Fig. 1: The ASUR instrument integrated into the NASA DC-8 during the SOLVE/THESEO-2000 campaign.

The radiometer consists of two standard 19" racks approved for the German Falcon and the NASA DC-8 research aircraft. The dewar containing the submillimeter mixer has to be filled with liquid nitrogen and liquid helium before the flight and stays cooled up to 10 hours. The cold load used for calibration purposes has to be refilled with liquid nitrogen about every two hours.

ATMOSPHERIC CHEMISTRY AND DYNAMICS

Mesosphere and lower thermosphere

The mesosphere and lower thermosphere (MLT) is one of the least explored regions of the Earth's atmosphere. It has gained recent interest as it is an important link in the vertical transfer of energy and material in the atmosphere where phenomena like gravity wave breaking, charged particle precipitation and aurorae take place (Jarvis, 2001). Using its high resolution spectrometer, ASUR is able to obtain profile information on atmospheric trace gases up to altitudes around 65 km. Fig. 2 shows measurements of ozone volume mixing ratio in the mesosphere around 70° N during a flight across the terminator. The expected nighttime increase in ozone mixing ratio can be clearly measured with ASUR. The total measurement time for a mesospheric ozone measurement is only 180 seconds, corresponding to a horizontal resolution of about 40 km at typical aircraft speed. In addition to ozone, upper stratospheric/lower mesospheric HCN has been recently retrieved from ASUR spectra.

Another key species in the MLT is nitric oxide, which can be very abundant in the lower thermosphere. Using its high resolution spectrometer ASUR is capable of obtaining information on the NO profile in the stratosphere as well as on the NO column in the lower thermosphere. This makes it an interesting tool for the study of NO production due to charged particle precipitation and aurora. Fig. 3 shows preliminary retrievals of upper mesospheric/lower thermospheric NO columns during a flight from Madrid, Spain to Omaha, Nebraska on 18 Nov. 2002 in the framework of the Leonid Multi-instrument Aircraft Campaign (Leonid MAC). An aurora was detected visually and by optical cameras co-aligned with ASUR's line of sight between about 7 and 10 h UT. An enhancement in NO can be detected in this region. Flying out of Moffett field, the range of SOFIA will easily allow to reach the Northern auroral oval and enable similar measurements during different conditions of auroral activity. These measurements will be supported by cameras in the visible wavelength range, the deployment of which are also suggested for the SOFIA Upper Deck Facility. The time period SOFIA activities are planned for will even allow to make measurements in different periods of solar activity. Of particular interest will also be the NO production following enhanced particle precipitation during geomagnetic storms, which can be followed by a decrease in mesospheric and upper stratospheric ozone when the produced NO is transported to lower altitudes (Sinnhuber et al., 2003). ASUR will be able to detect both, the NO enhancement and the resulting deficit in ozone.

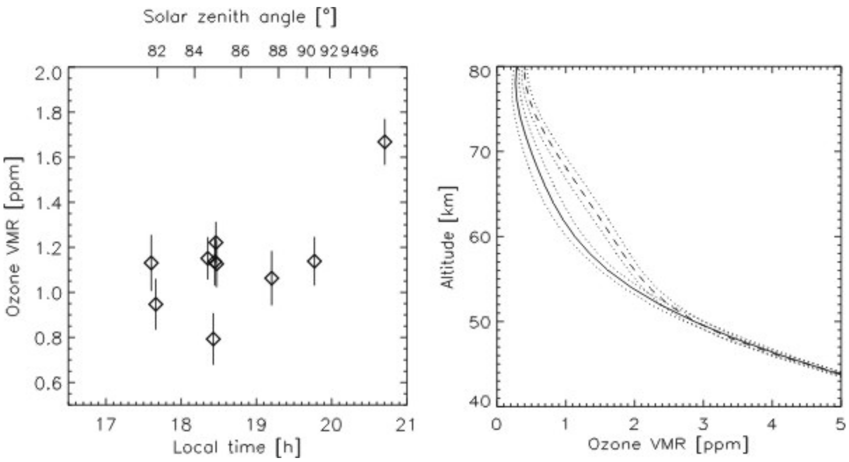
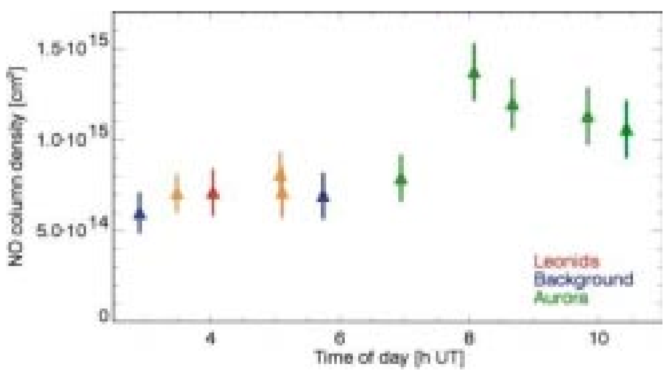


Fig. 2: Left: ASUR ozone at 60 km altitude during a flight across the terminator on 4 Sept. 2002 in the Arctic. Right: Retrieved daytime (SZA 81.6°, solid) and nighttime (SZA 97.6°, dashed) ozone profiles during this flight. From Kleinböhl et al. (2004).

Fig. 3: Preliminary retrievals of NO columns in a region above 70 km measured by ASUR during a Leonid Multi-Instrument Aircraft Campaign (LEONID MAC) flight from Madrid, Spain to Omaha, Nebraska on 18 Nov. 2002. Note the presence of an aurora between 7 and 11 h UT.



As an asset ASUR will be capable of measuring the variability of trace gases like HCN and ozone during meteor storms (Depois et al., 2000), which has been demonstrated in November 2002 during the Leonid shower. Another important radical OH which dominates the upper atmospheric chemistry is accessible at higher frequencies (2.5 THz or above) and has been measured successfully with an far-infrared airborne sensor (Titz et al., 1995). Such measurements could be performed in parallel to HO_2 measurements with ASUR.

Stratosphere

The potentials of ASUR flying at high altitudes abroad a long-range aircraft, like SOFIA, will allow stratospheric studies of variability and long term trends of many chemical species and tracers. Monitoring the stratospheric constituents is mandatory to understand the anthropogenic impact on the environment which is rapidly growing in our days. Ongoing objectives are climate change and ozone depletion, while the stratospheric chlorine loading has passed its maximum and is expected to decline during the next decades. Although present satellite instruments are able to detect many of the ASUR species, those measurements are mostly limited by the horizontal resolution and the sometimes patchy global coverage of the earth's surface. The horizontal resolution of ASUR is appropriate for many process studies and investigations of locally restricted events (e.g. vortex edge and subtropical barrier). For particular studies ASUR is complemented well by airborne aerosol lidar (von König, et al., 2002). Prolonged aircraft measurements also help to cope the important challenge of long term validation for the new generation of space instruments, as suggested for ASUR with regards to e.g. MLS on AURA, and MIPAS and SCIAMACHY on ENVISAT.

Figure 4 shows the vertical distribution of N_2O as a function of latitude observed during the validation campaigns for the instruments of the ENVISAT satellite at different seasons in 2002/03. The deployments spanned from Spitsbergen (80°N) to the Seychelles (5°S). The crosses indicate the locations of the aircraft during measurements, other molecules have been measured in between. The plots visualize the dynamical and photochemical behaviour of N_2O which is transported basically by the Brewer-Dobson circulation and depleted by photolysis. Thus a decrease of volume mixing ratio is observed towards higher latitudes. The strong variability around 65°N is due to large longitudinal excursions.

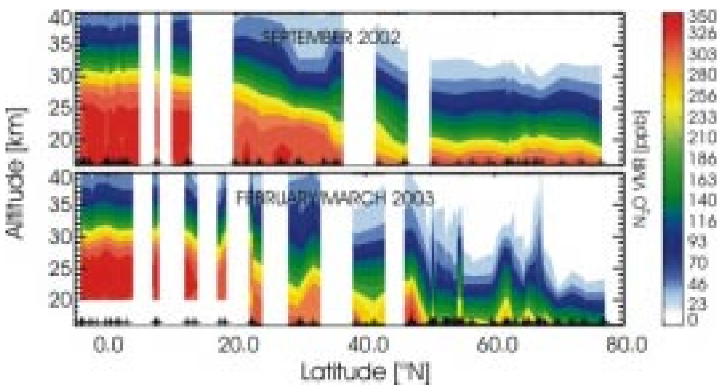


Fig. 4: Latitudinal cross-sections of N_2O vertical profiles measured by ASUR during the SCIAMACHY validation campaigns in autumn 2002 and winter/spring 2003 (Kuttippurath et al., 2004).

UT/LS region

The upper troposphere/lower stratosphere and in particular the tropical tropopause layer (TTL) has become a major region of interest during the recent years. Changes in the dynamics and composition of the TTL have the potential to strongly influence the stratosphere in the future, e. g. due to a change in the flux of water vapor or other chemical constituents from the troposphere to the stratosphere. To address these important questions it is suggested to operate microwave receivers in limb geometry from SOFIA. The frequency range of the ASUR instrument at about 600 GHz is not suited for this purpose as the strong water vapor absorption at these frequencies lead to a saturation of the measurement at a tangent altitude of ~12 km. However, there are windows at lower frequencies that will allow measurements down to a tangent altitude of about 6 km in the arctic and about 8 km in the tropics. The limb geometry provides highly resolved profiles below the aircraft altitude. In addition, the lines could be observed in uplooking geometry to provide vertical profiles above flight altitude in a comparable quality of the ASUR data products (depending on frequency resolution and integration time). The lower frequency and the increased accuracy in pointing necessary for the limb measurements would require an antenna dish larger than the ASUR antenna. However, due to the limited cruising altitude of SOFIA the tangent point of a limb observation is not very far from the aircraft such that an inside antenna observing through an aircraft window should be feasible. Three frequency bands are particularly interesting for UT/LS observations:

- 300 – 345 GHz: This frequency range chosen for the planned European satellite instrument MASTER contains spectral signatures of O_3 around 300 GHz, a water vapor line at 325 GHz and a CO emission at 345 GHz. The combination of these three species will be powerful for the study of stratosphere-troposphere exchange processes. O_3 can be used as a tracer for airmasses of stratospheric origin while CO can be used to track tropospheric airmasses. Measurements of these two species together with water vapor will provide a tool to investigate the ways of water vapor injection into the stratosphere. An instrument for the confirmation of the technical feasibility for radiometric measurements of this type has been recently constructed (Oldfield et al., 2001).
- ~ 270 GHz: This frequency range allows to access numerous species, in particular HCN, N_2O, ClO, HNO_3, and O_3. It would be particularly suitable for the combination of uplooking and limb geometries. The combination of observable species would be especially suited for studies of the lowermost stratosphere in the polar region during and after the polar winter. Ground-based instruments working at these frequencies are in operation, e. g. at Summit, Greenland, and at Merida, Venezuela.
- 183 + 203 GHz: These frequencies correspond to a strong water vapor line and an emission of the water vapor isotope H_2O-18, respectively. They are suitable for studies of the isotopic ratio in the UT/LS, which for example could provide information on the condensation history of the observed

airmasses. Ozone lines to be used for the identification of stratospheric airmasses are found in this frequency region as well.

CONCLUSIONS

We presented goals for the deployment of airborne microwave receivers on board the SOFIA research aircraft. The airborne submillimeter radiometer ASUR, which has performed already in numerous campaigns for atmospheric research, would be a suitable tool on the SOFIA Upper Deck Facility to target several questions in stratospheric and mesospheric chemistry and dynamics. Its ability to measure ozone and HCN in the lower mesosphere as well as NO in the lower thermosphere enables ASUR to study the variation of NO under the influence of auroral activity as well as ozone depletion following downward transport of NO produced by enhanced charged particle precipitation. In the stratosphere ASUR's ability to measure vertical profiles of trace gases related to the ozone chemistry, like ozone, HCl, ClO, and HNO_3 as well as dynamical tracers like N_2O and CH_3Cl makes it suitable to target numerous questions related to chemistry and dynamics. These include detailed process studies, where its good horizontal resolution is a unique advantage, e. g. studies of transport processes at the edge of the polar vortex or the subtropical barrier, or co-ordinated studies together with airborne lidar. In addition measurements of latitudinal cross sections of tracers will help refine models of the stratospheric circulation and identify possible long-term trends. Eventually, trace gas profiles measured by ASUR can also serve as a long-term validation of several satellite instruments over a large latitudinal range.

To extend the measurement range to the upper troposphere/lower stratosphere region we propose the use of new instrumentation in limb viewing geometry at millimeter wavelengths. This would allow the meaurement of highly resolved trace gas profiles down to 6-8 km. We suggest three frequency ranges between 180 and 350 GHz where observable species include ozone, H_2O, H_2O-18, CO, N_2O, HNO_3, ClO, and HCN. Questions to be targeted include current problems in stratosphere-troposphere exchange and water vapor injection through the tropical tropopause layer.

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