Aerosol/Clouds microphysics and Heterogeneous chemistry studIes in the loWEr stratosphere by Models and Multi-intrument Measurements. (ACHIWE-MMM)

Category and Objectives: Proposal Objective and Type (Priority 1): Scientific Research Atmosphere

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Experience

All the investigators have a proved experience in the studies of the stratospheric aerosol and polar stratospheric clouds by means various experimental and theoretical techniques as documented in the current scientific literature. For such purposes they have been using data coming from different kinds of instrumentation on satellite, airborne, balloon-borne and groundbased. Some of them have developed and successfully used models for studying the optical properties, the microphysics, the chemistry of the aerosol and the polar stratospheric clouds.

Some of the partners are currently involved in a study of the polar stratospheric clouds using a combination of Lidar data and models along with Upper Atmospheric Research Satellite (UARS) and the Improved Limb Array Spectrometer (ILAS) measurements collected in the previous years. Such project is funded by the European Commission.

Executive Summary and Schedule

Executive Summary

The aim of the project is to combine atmospheric measurements of polar stratospheric clouds (PSCs) obtained from different instruments and atmospheric models to investigate PSCs formation and the heterogeneous chemistry involved in stratospheric ozone depletion. The proposal will involve the use of measurements from some ENVISAT instruments, which would provide aerosol and chemical data, from 2 Arctic, 2 Antarctic and 4 European lidar stations, and from in situ laser backscatter sonde and optical particle counter.

A sampling satellite data along Lagrangian trajectories would allow the study of the evolution and decay of a PSC within given air parcels. The project would permit advancements in:

- a description of the spatial scales of PSC properties,
- the microphysical properties by polar stratospheric clouds,
- the chemical processing within a polar stratospheric cloud,
- improved chemical modelling in the lower stratosphere.

Schedule

3-year activity starting from the date of availability of Envisat data after the validation and calibration phase.
1st year:

-collection of the data from satellite and other instruments,
- -selection of the case studies on the basis of the measurements,
- -preparation of the models for their integration with experimental measurements.

2nd and 3rd years:

- -continuation of the measurements and selection of the case studies,
- -integration of the observations and the models,
- -integration of the models with experimental measurements.

Progress meetings every 6 months from the start of the project, preparation of a progress report every year.

Detailed Description

Innovative Character of the Proposal:

This proposal involves the synergistic combination of atmospheric measurements and models in which individual groups have already developed their particular and complementary expertise. The proposed advances in the knowledge of the atmosphere are achieved through a interdisciplinary approach and these advances could not be attained at an individual group level. The composition of the complex picture in which all the aspects of the polar ozone depletion from the microphysics to the chemistry and the dynamics of the polar vortices would be investigated. A clear innovation in this proposal is the sampling of satellite data along Lagrangian trajectories so that the evolution and decay of a polar stratospheric cloud within an airparcel can be examined. In doing so the chemical changes within an airparcel are effectively decoupled from the dynamics allowing a clear picture of PSC formation and heterogeneous processing to be obtained.

Detailed Description:

PSCs consist of micron-sized particles formed from the condensation of gaseous HNO3 and H2O in the winter polar stratospheric vortices. The current understanding is that PSCs consist of a variety of phases and compositions. At the lowest temperatures ice crystals are formed, however the formation of PSCs may encompass several intermediates consisting of amorphous or crystalline hydrates of nitric acid or supercooled droplets of H2SO4-HNO3 -H2O solutions. Liquid particles may undergo supercooling by several degrees in order to form frozen particles and then, once formed, the solid particles can exist at higher temperatures. This hysteresis in the temperature of PSCs formation and evaporation has large implications since not only are local temperature variations important in the determination of halogen activation, but so is the temperature history of the air parcel. In addition to PSC composition,
particle size and physical phase are critical parameters in the estimation of the reaction rates leading to ozone depletion. If temperatures are cold enough then PSC particles grow large enough to fall to lower altitudes. This has the effect of permanently removing the condensed nitric acid from the air. Under denitrified conditions halogen can remain active longer giving more prolonged ozone destruction. A consequence of the poor understanding of the PSCs properties is that there is limited ability to predict the future evolution of the ozone layer.

Climate model calculations have recently indicated that increased concentrations of greenhouse gases may lead to lower temperatures in the stratosphere and stronger and long-lasting polar vortices with fewer minor warmings possibly extending toward midlatitudes. Together with increasing trends in stratospheric water vapour this may lead to more widespread and long-lasting PSC formation and changes in PSC properties, in particular in the Arctic regions. A positive feedback on stratospheric temperatures may occur due to consequently enhanced ozone depletion which may delay the spring time warming. In particular in the Arctic, where winter temperatures hover around the PSC thresholds, it is important to assess if PSC formation and PSC properties tend to change, resembling conditions over Antarctica.

Instruments and models

ENVISAT instruments (data managed by IFA, OU, CAU,) are able to perform measurements on aerosol and chemical species relevant in the frame of the project. The aerosol data will be complemented by Lidars, LABS and OPC data. A 4D-Var assimilation of aerosols will be used in this framework tofacilitate the comparison and analysis of data and model results. Meteorological and chemical data will give the constrains in which the microphysical, chemical and transportation models should be applied. The ENVISAT data relevant to the project are: pressure (P), temperature (T), O3, H2O, CH4, N2O, and HNO3 from MIPAS; O3, NO2, NO3, O2, H2O, T and aerosol density and extinction from GOMOS; profiles of P, T, O3 NO2 and aerosol from Sciamachy.

The PSC/aerosol microphysical model (DMI,AU) permits the study of PSCs formation and evolution. It will include stratospheric sulphate aerosol, liquid supercooled ternary solution and solid nitric acid trihydrate particles, solid water ice, and frozen sulphate aerosol. Such a model will allow consistent calculations of heterogeneous reaction rates on particles and consequently ozone destruction rates. Microphysical models are also needed for more accurate simulations of denitrification and dehydration to be compared with observations.

The interpretation of the lidar and satellite data requires a PSC/aerosol optical model (OU, CAU, IFA) to relate the backscatter and extinction data to the particle fundamental properties (i.e. size parameter and refractive index). The Mie theory, which describes the scattering and absorption of radiation by homogeneous spheres, will be used to analyse the PSC measurements. To account for the non-sphericity of the solid PSCs, corrective factors will also be considered. The PSC/aerosol optical model will also be required to predict PSCs infra-red emission spectra which depend very strongly on PSC composition.

The detailed calculation of heterogeneous reaction rates is based on a different treatment of liquid and solid particles. The characteristics of the available PSC/chemical box model (BIRA) includes an easy choice of the microphysical theory (pure liquid, NAT on presupposed frozen SAT, different freezing and melting conditions of the particles) and detailed or simple calculation of heterogeneous reaction rates. The Chemical Transport Model, CTM (SA, CU, AU), is a 3D model which is driven by realistic dynamical fields from either ECMWF analysis or forecast and provides the link with PSC chemical model. The model has the ability to be used with different spatial resolutions and variable particle size distributions. This CTM is used in a 4D-Var assimilation (BIRA): the misfit between observations and model results is minimised with respect to CTM initial conditions as to reproduce the observations. Currently only aerosols and dynamics are taken into account and the assimilation of chemical species is under development.

Lidars can provide aerosol and PSC optical properties by measuring depolarisation and backscatter. These parameters are directly determined by the particles' composition (through refractive index), phase and size distribution. Lidars provide instantaneous profile data with a very high vertical resolution, so that is able to resolve fine layered structures, as often observed in PSCs. Lidars are also able to measure profiles of stratospheric temperature and ozone. A small network of European Lidars – in the Arctic, in Antarctica and in Europe - will be involved in the project to provide different data - A is for Aerosol and PSC:

- Ny-AAlesund, AWI: A, T, O3

- Andoya, SA & LIAP: A, T, wind
- McMurdo, IFA: A, T
- Dumont D'Urville, SA: A, O3
- Observatory of Haute Provence, SA: A, T, O3
- Garmish, IFU: A, O3
- Rome, IFA: A, T
- L'Aquila, AU: A, O3

Besides the obvious importance of the polar Lidars, the European ones are not less important to the project. In fact, the Arctic vortex, unlike the Antarctic one, is extremely dynamic and can extend to the midlatitudes. A LAser Backscatter Sonde, LABS (IFA), can measure backscatter and depolarisation from atmospheric particles. Joint LABS/OPC (Optical Particle Counter, WU) balloon flights can measure the concentration, size distribution, backscatter coefficient and phase (liquid or solid) from the particles.

The activity

PSC data from ENVISAT, Lidars and LABS/OPC will be selected under a range of winter conditions. Through wavelength conversion, the parameters measured by Lidar can be compared to ENVISAT extinction data. The PSC models will then be used to calculate infra-red emission spectra to be compared to MIPAS data. It will be evaluated how well satellite limb measurements reflect the changes that occur in PSCs. Lidar data will be used to derive correlation length scales of PSC density and phase in the vertical. An estimate of the uncertainty in the retrieved value of extinction due to PSC fine scale structure will be provided.

Experimental results show that PSCs can consist of liquid ternary solution, frozen and perhaps some metastable particles which can be present at the same time. The exact formation, speciation and evolution of solid particles is not yet fully understood. The improvement of existing theory involves clear feedback from model/measurement comparison into the development of the PSC model.

One important element of the planned model studies is a box model with a detailed implementation of PSC/aerosol microphysics. This can be used to calculate PSC microphysical parameters relevant for the calculation of PSC optical properties and heterogeneous reaction rates together with rates of denitrification and dehydration.

The microphysical model will be linked with a model for detailed heterogeneous processes and standard gas phase chemistry of the stratosphere. This activity will thus be concentrated on the implementation of detailed determination of heterogeneous processes into the general chemical mechanism using the characteristics from the PSC microphysical model. Results from the PSC model will be compared with satellite data of PSC extinction and chemical species in order to optimise and validate the microphysical assumptions of the model. The PSC model will be run along Lagrangian trajectories through PSCs.

Coincident measurements of temperature, gaseous HNO3, and H2O will be used as inputs. The sensitivity of chlorine activation and ozone destruction on aerosol/PSC characteristics and heterogeneous reaction rates will be evaluated. The activation of halogen on different types of particles will serve as an additional validation of the PSC formation mechanism. CTM calculations tend to underestimate the large ozone depletion rates. This could be due to insufficient knowledge of the heterogeneous processes. Hence, detailed particle simulations, based on the actual temperature development in the air parcels where the particles form, as given from a 3D CTM, are required to investigate the effects of heterogeneous chemical reactions, and for studies of PSC formation.

Data Requirements

Instruments:

- MIPAS
- GOMOS
- SCIAMACHY

MIPAS

Geolocated Calibrated Spectra (MIP_NL_1P)
MIPAS Temperature, Pressure and Atmospheric Constituent Profiles
(MIP_NL_2P)

GOMOS

GOMOS Temperature, Atmospheric Constituents Profiles (GOM_NL_2P)

SCIAMACHY

Vertical Profiles and Total Column Amount of Temperature, Pressure and
Various Trace Gases (SCI_NL_2P)

Date Range / Geographical Area

Date Range:

01-07-2000 - 31-12-2000
Area Name:
Winter Southern Stratosphere 2000

Rectangle:

Top left Latitude (deg min) / Top left Longitude (deg min): 0° 00'/ 0° 00'
Bottom right Latitude (deg min) / Bottom right Longitude (deg min):
-90°00'/360°00'

This geographical Area is related to:

Comments:

As the processes we are going to study can be strongly connected to the dynamics of the stratosphere on a large scale, we request data for a full coverage of the respective winter hemispheres. The data would cover the time from the start of PSC formation to the end of the ozone hole phenomenon. In the vertical range the data should cover at least all the atmosphere up to the lower mesosphere (50-60 km).

Date Range:
01-11-2000 - 30-06-2001

Area Name:
Winter Northern Stratosphere 2000-2001

Rectangle:
Top left Latitude (deg min) / Top left Longitude (deg min): +90° 00'/ 0° 00'
Bottom right Latitude (deg min) / Bottom right Longitude (deg min):
0°00'/360°00'

This geographical Area is related to:

Comments:
apply the previous comment

Date Range:
01-05-2001 - 31-12-2001

Area Name:
Winter Southern Stratosphere 2001

Rectangle:
Top left Latitude (deg min) / Top left Longitude (deg min): 0° 00'/ 0° 00'
Bottom right Latitude (deg min) / Bottom right Longitude (deg min):
-90°00'/360°00'

This geographical Area is related to:

Comments:
apply the previous comment

Date Range:
01-11-2001 - 30-06-2002

Area Name:
Winter Northern Stratosphere 2001-2002

Rectangle:
Top left Latitude (deg min) / Top left Longitude (deg min): +90° 00'/ 0° 00'
Bottom right Latitude (deg min) / Bottom right Longitude (deg min): 0°00'/360°00'

This geographical Area is related to:

Comments:
apply the previous comment

Date Range:
01-05-2002 - 31-12-2002

Area Name:
Winter Southern Stratosphere 2002

Rectangle:
Top left Latitude (deg min) / Top left Longitude (deg min): 0° 00'/ 0° 00'

Bottom right Latitude (deg min) / Bottom right Longitude (deg min): -90°0'/360°00'

This geographical Area is related to:

Comments:
apply the previous comment

Date Range:
01-11-2002 - 30-06-2003

Area Name:
Winter Northern Stratosphere 2002-2003

Rectangle:
Top left Latitude (deg min) / Top left Longitude (deg min): +90° 0'/ 0° 00'
Bottom right Latitude (deg min) / Bottom right Longitude (deg min): 0°00'/360°00'

This geographical Area is related to:

Comments:
apply the previous comment