

European Commission

Directorate-General XII for Science,

Research and Development

Second European Stratospheric Arctic and Mid-latitude Experiment (SESAME)

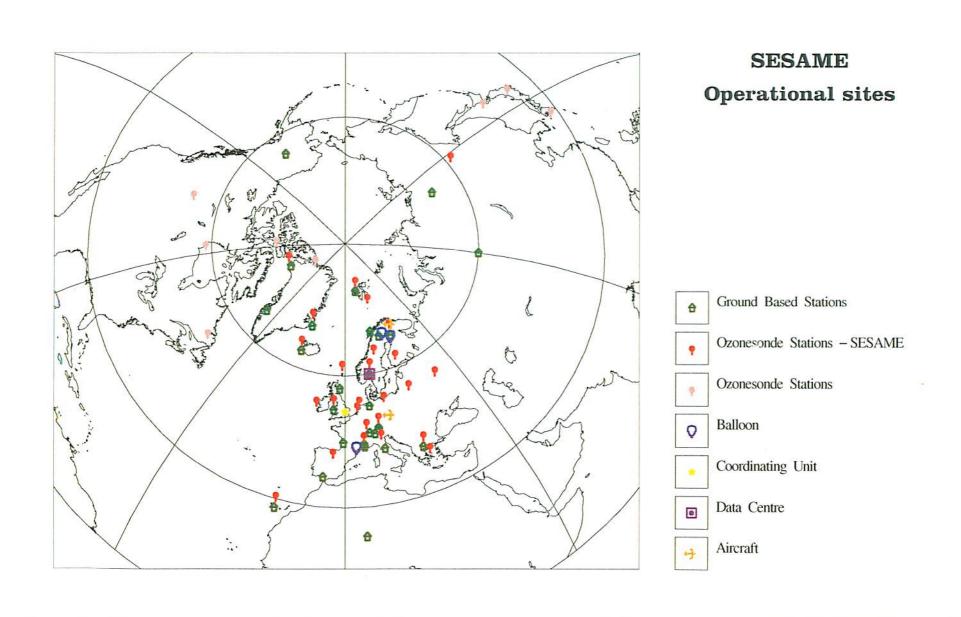
1994 - 1995

A European contribution to the stratospheric ozone issue



ENVIRONMENT PROGRAMME





INTRODUCTION

vidence that human activities affect the ozone layer has been building up over the last 20 years, ever since scientists suggested that the release of chlorofluorocarbons (CFCs) into the atmosphere could reduce the amount of ozone over our heads. First, the breakdown products of these gases were detected in the stratosphere. Next, the "ozone hole" was discovered and soon after it was proved that chlorine compounds were the cause (see Box 1). At around the same time the first firm evidence was produced that there had been an ozone decrease in the stratosphere over the heavily populated northern mid-latitudes (30-60°N). Many of these findings have since been reinforced by a variety of internationally supported scientific experiments involving satellites, aircraft, balloons and ground stations.

The European Commission, within the ENVIRONMENT research programme (1991-1994), supports scientific research activities designed to understand the causes and the consequences of stratospheric ozone depletion. The key components are:

- (a) <u>laboratory measurements</u> of basic properties, such as how fast different chemicals react and how light affects them:
- (b) <u>field measurements</u> of important atmospheric components, such as the concentration of chemical species and the nature of stratospheric clouds and aerosols;
- (c) model studies where our understanding of the stratosphere is fed into computer models and the results compared to available field measurements. Models can also be used to make predictions about the future state of the atmosphere.

In the framework of the ENVIRONMENT research programme, a major European campaign, the European Arctic Stratospheric Ozone Experiment (EASOE) was organised to study the north polar regions during the winter of 1991/92. EASOE studies have demonstrated quantitatively the extent of ozone depletion in the Arctic regions and that this ozone loss is consistent with stratospheric chlorine activation under particular meteorological conditions (see Box 3). Despite these findings, many intriguing questions remain: What are the causes of the mid-latitude loss? How are the losses over the Poles linked to those at mid-latitudes? While CFCs and the bromine-containing compounds known to destroy ozone over the Poles are strongly implicated in the mid-latitude loss, a few doubts remain.

In 1994 and 1995, the Second European Stratospheric Arctic and Mid-latitude Experiment (SESAME) is being organised to investigate the processes occurring at both high and mid-latitudes and to determine how they are linked. SESAME is built upon the experience gained during EASOE. At the same time, a US-led expedition is studying similar processes in the southern hemisphere. These two campaigns are expected to enhance our understanding of the stratospheric ozone layer on a global scale.

These EC stratospheric research activities are undertaken in conjunction with the research programmes of individual countries and contribute to a continuing scientific assessment, in liaison with the World Meteorological Organisation and the United Nations Environment Programme, of the causes and the consequences of stratospheric ozone depletion. This is directly related to the Montreal Protocol (see Box 2) and is to be considered as part of the obligations derived from the Vienna Convention, of which the European Union (EU) is a signatory. To ensure that maximum benefit is gained from both the national and EC programmes, the EC is advised by a Science Panel on Stratospheric Ozone, made up of experts from EU and EFTA countries. To assist specifically in the coordination of this European research effort, the EC, together with the UK Department of Environment, supports the European Ozone Research Coordinating Unit which is based in Cambridge, UK.



Balloon launch at ESRANGE, Kiruna, Sweden

BOX 1

CHEMICAL PROCESSES LEADING TO POLAR OZONE DEPLETION

The <u>depletion of ozone</u> in polar latitudes is attributed to a sequence of chemical reactions involving chlorine and bromine compounds. <u>The sources</u> of these are simple organic compounds containing chlorine, e.g. chlorofluorocarbons (CFCs), and/or bromine (e.g. halons). Nearly all of the chlorine and about half of the bromine in the stratosphere originates from human activities. These source compounds can be broken down in the stratosphere to form less stable products (e.g. HCl and ClONO₂). These in turn are broken down by sunlight to give high concentrations of reactive chlorine and chlorine monoxide radicals (Cl and ClO). This leads to catalytic cycles that can destroy ozone very rapidly, and in which a significant part is played by bromine and bromine monoxide radicals (Br and BrO).

The main long-lived inorganic carriers (reservoirs) of chlorine in the atmosphere are hydrochloric acid (HCl) and chlorine nitrate (ClONO $_{\circ}$). Dinitrogen pentoxide (N $_{\circ}$ O $_{\circ}$) is a reservoir of oxides of nitrogen. All the gas phase reactions of these reservoirs are relatively slow, but fast reactions can occur on a suitable surface or in solution (heterogeneous reactions). Polar stratospheric cloud (**PSC**) particles are believed to support such reactions. PSCs can form in the lower stratosphere at temperatures below about 195 K (-78°C), and are believed to contain solid phases of mixed nitric acid and water. Laboratory experiments suggest that the most important reactions are:

$$\begin{array}{ll} HCl + ClONO_2 \rightarrow HNO_3 + Cl_2 & (1) \\ ClONO_2 + H_2O \rightarrow HNO_3 + HOCl & (2) \\ HCl + HOCl \rightarrow H_2O + Cl_2 & (3) \\ N_2O_5 + HCl \rightarrow HNO_3 + ClONO & (4) \\ N_2O_5 + H_2O \rightarrow 2HNO_3 & (5) \\ \end{array}$$

The chlorine compounds formed in these reactions are readily photo-dissociated, even in low light conditions, releasing chlorine atoms (Cl).

The nitric acid (HNO₃) formed in these reactions remains in the particles, so that the gas phase concentrations of oxides of nitrogen are reduced. This reduction, `denoxification', slows down the rate of removal of ClO in the reaction:

```
ClO + NO_2 + M \rightarrow ClONO_2 + M (where M is any air molecule) and so helps to maintain high levels of active chlorine. (6)
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It is now known that heterogeneous reactions can also take place on, or in, sulphate aerosol particles (droplets of sulphuric acid in solution), which are always present in the lower stratosphere. The droplets are formed following the oxidation of carbonyl sulphide (OCS), a naturally occurring sulphur compound that has a long enough lifetime to survive the slow transport to the stratosphere. Another, highly spasmodic, source of aerosol is sulphur dioxide, injected directly into the stratosphere during some volcanic eruptions. This can raise aerosol concentrations to many times the background.

Sulphate aerosol is known to support reactions (2) and (5), and possibly (3), leading both to the release of active chlorine, and to denoxification. Thus there is the potential for widespread ozone destruction. However the temperature dependence of the heterogeneous reactions on the aerosol seems to be such that rapid destruction is possible only at temperatures close to those required to form PSCs, and the effects of the background aerosol are thought to occur slowly.

The production of active chlorine requires sunlight, and sunlight can drive the following catalytic cycles:

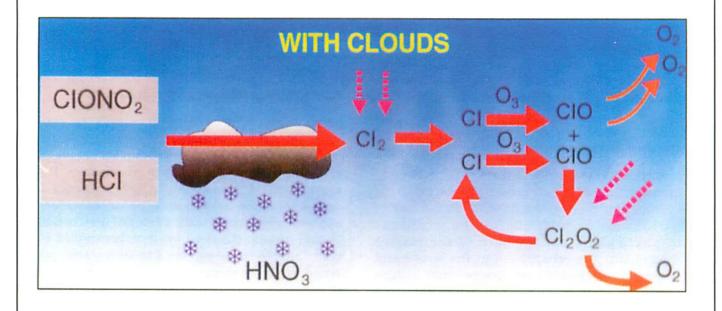
$$Cycle\ I \ \begin{cases} & ClO + ClO + M \ \rightarrow Cl_2O_2 + M \\ & Cl_2O_2 + h\sqrt{\rightarrow} Cl + ClO_2 \\ & ClO_2 + M \rightarrow Cl + O_2 + M \\ & 2\ x\ (Cl + O_3 \rightarrow ClO + O_2) \\ & net\ 2O_3 \rightarrow 3O_2 \end{cases}$$

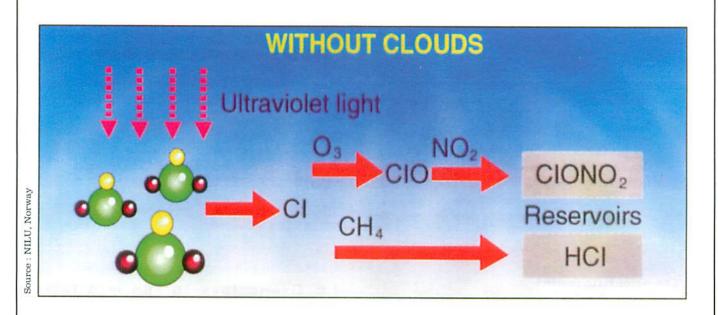
$$and$$

$$ClO + BrO \rightarrow Br + Cl + O_2 \\ & Cl + O_3 \rightarrow ClO + O_2 \\ & Br + O_3 \rightarrow BrO + O_3 \\ & net\ 2O_3 \rightarrow 3O_2 \end{cases}$$

The dimer (Cl2O2) of the chlorine monoxide radical involved in Cycle I is thermally unstable, and the cycle is most effective at low temperatures. It is thought to be responsible for most (70%) of the ozone loss in Antarctica. In the warmer Arctic a large proportion of the loss may be driven by Cycle II.

Activation of stratospheric chlorine with and without the presence of polar stratospheric clouds





BOX 2

INTERNATIONAL REGULATORY POLICY

The first global agreement to restrict CFCs came with the signing of the **Montreal Protocol** in **1987**. Two revisions of this agreement have been made in the light of advances in scientific understanding, the latest being in 1992 at Copenhagen. Agreement has been reached on the control of industrial production of many halocarbons until the year 2030. The main CFCs will not be produced by any of the signatories after the end of 1995, except for a limited amount for essential uses, such as for medical sprays.

The countries of the European Union have adopted even stricter measures than are required under the Montreal Protocol agreements. Recognising their responsibility to the global environment, they have agreed to halt production of the main CFCs from the beginning of 1995. Tighter deadlines for use of the other ozone-depleting compounds are also being adopted.

Provisions of Copenhagen amendment to the Montreal Protocol

PRODUCT	ACTION								
CFCs	Annual production of each CFC must be reduced by 75% by 1 January 1994, and eliminated by 1 January 1996. (For the five CFCs controlled since 1989, the reductions relate to 1986 production figures. For the ten CFCs added to the Protocol since then, the reductions relate to 1989 production figures.)								
Halons	A total phase-out of halons is required by 1 January, 1994. Three halons are controlled - Halons 1211, 1301 and 2402.								
Carbon tetrachloride	Annual production of carbon tetrachloride must be reduced by 85% by 1 January 1995, and by 100% by 1 January 1996 (in relation to 1989 production figures).								
Methyl chloroform	Annual production of methyl chloroform must be reduced by 50% by 1 January 1994, and by 100% by 1 January 1998 (in relation to 1989 production figures).								
HCFCs (hydrochlorofluorocarbons)	A freeze in consumption of HCFCs is required in 1996, and 100% reduction must be achieved by 2030.								
HBFCs (hydrobromofluorocarbons)	A complete phase-out of production and consumption of HBFCs is required by 1 January 1996.								
Methyl bromide	A freeze in consumption and production is required in 1995. A review will be carried out before 1995, so that an agreement can then be made on further controls, probably a 75% reduction by the year 2000.								

BOX 3

EUROPEAN ARCTIC STRATOSPHERIC OZONE EXPERIMENT E A S O E

EASOE took place in the winter of 1991/92. Its overall objective was to study the evolution of the Arctic stratosphere in that winter. Particular goals included:

- a) to measure the change in ozone concentration with altitude throughout the winter;
- to measure the concentrations of other trace chemical species (especially chlorine and nitrogen species);
- to investigate the role of polar stratospheric clouds, and in particular to study dehydration and denitrification of the Arctic stratosphere;
- d) to study the meteorological processes which move chemically-perturbed air southward.

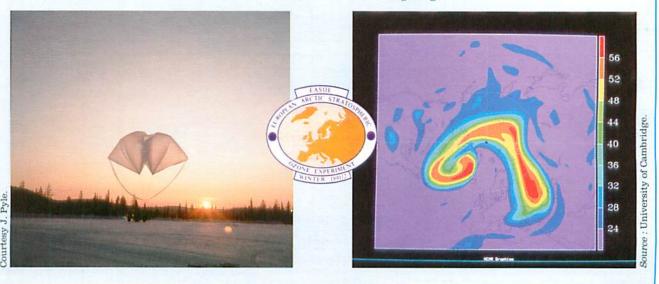
Measurements were made using ground-based instruments at 16 sites ranging from the Arctic Circle to southern Europe, from large stratospheric balloons and from three research aircraft. 43 stratospheric balloons, capable of carrying scientific payloads of up to 500kg directly into the ozone layer, were launched from the Swedish Space Corporation rocket range ESRANGE near Kiruna, Sweden. In addition, over 1000 ozonesondes were launched on small balloons from more than 20 sites. A wide variety of meteorological information was provided by the various European meteorological services, including some information especially provided for EASOE scientists.

While scientists from a number of non-European countries took part, EASOE was primarily an European activity (with over 250 participating European scientists). As well as being scientifically successful, EASOE was extremely effective in forging new collaborations between many research groups. Links between stratospheric scientists now cross national boundaries with ease, allowing more beneficial collaborations to occur in the future.

Many of the results of EASOE are published in a special issue of Geophysical Research Letters (June 22, 1994, Vol. 21, No. 13) containing over 70 scientific papers. They include measurements of the important chemical species involved in ozone destruction and analyses of the interplay between the dynamic motions of the stratosphere and the chemistry which occurred. For instance, it is clear that there was a large region of air over the Arctic in which the chlorine had been activated. EASOE studies have shown that the ozone loss in the Arctic vortex, measured in the experiment, is consistent with the chlorine activation. Other studies are providing important clues to the reasons for the middle latitude ozone decline.

Auxiliary balloons used in the main balloon launch during EASOE.

Meteorological map showing the distorsion of the polar vortex in mid-January 1992 from a forecast run of U.K. Universities Global Atmospheric Modelling Programme.



SECOND EUROPEAN STRATOSPHERIC ARCTIC AND MID-LATITUDE EXPERIMENT, SESAME

The detailed cause of the long-term midlatitude ozone decrease is controversial. A number of ideas have been advanced, giving possible explanations of the chemical and dynamical processes leading to ozone destruction. These need to be confirmed, by measurements and by theoretical studies, before we can make confident predictions about future changes. Accordingly, the EC has now funded a major field campaign to build upon the experience gained during the European Arctic Stratospheric Ozone Experiment (EASOE, see Box 3) and its Franco-German predecessor, Chemistry of Ozone in the Polar Stratosphere (CHEOPS), as well as from the various US aircraft campaigns. The Second European Stratospheric Arctic and Mid-latitude Experiment (SESAME) started in January 1994 and will continue until the end of 1995. The majority of the scientists involved in SESAME will, as in EASOE, be from Europe but scientists from many other countries are also involved. In all, over 55 research groups from 21 countries will take part.

OBJECTIVES

The SESAME strategy calls for measurements at a variety of times throughout the year of processes occurring in, and connecting, the lower stratosphere of high and middle northern latitudes. This requires high quality measurements of ozone, active chemical species and stable species which are useful for tracing stratospheric transport processes, in conjunction with detailed meteorological studies. Scientific goals are to study:

- the stratospheric chemistry of the chlorine, nitrogen, hydrogen and bromine families;
- the roles of polar stratospheric clouds and of sulphate particles in abetting the chemical destruction of ozone:
- the evolution of ozone concentrations through the two winters and to see exactly when and where the ozone loss occurs;
- the meteorological conditions most likely to favour ozone destruction.

Year	1994										1995														
Month			F	M	A	M	J	J	A	S	0	N	D	J	F	M	A	M	J	J	A	S	0	N	D
Ballon flights	Kiruna/Andoya (=70N)																								
	Aire sur l'Adour/ GAP (=45N)																								
Aircraft	TRANSALL																								
	ARAT																								
	FALCON																								
	GEOPHYSIKA																								
Ground-based measurements	Ozone																								
	SCUVS (12 sites)																								
	ESMOS (2 Arctic sites)																								
	ESMOS (10 Alpine sites)																								
	Arctic (5 sites)	•																							
	Mid-laditudes (5 sites)																								
	UVB intensive (4 sites)																								
Ozonesondes	26 sites																								

Depth of shading indicates the intensity of the particular activity.

Full details of these projects are found in 'Stratospheric Chemistry and Ozone Depletion. Field measurements: SESAME, Laboratory measurements, Modeling, Instrument development', A planning document for the period January 1994-December 1995, European Commission DGXII D-1. AirPollution Research Report n°51 EUR 15658 EN

INSTRUMENTATION

A wide variety of instruments are being used. The strategy is to have a background set of measurements made by instruments operated throughout 1994 and 1995. On top of this. there will be three intensive periods of study in January-March 1994 (see SESAME Phase I, in page 10), September-October 1994 (Phase II) and December-April 1995 (Phase III). The first and last of these are periods when ozone loss is expected to occur, while the middle phase will be used to monitor conditions prior to the winter, a previously under-studied period. A schematic showing when measurements will be made is given in Table 1.

GROUND-BASED INSTRUMENTS

The large year-to-year variation in sratospheric conditions makes it necessary to study several years in order to fully understand the factors underlying the long term trends and so the EC. through its ESMOS and SCUVS have projects, been supporting ground-based measurements

in the Arctic region and in Europe as part of the Network for the Detection of Stratospheric Change. Many of the instruments have been making measurements since EASOE (1991/92) - and a few for longer - and a valuable record is being built up. The scientists in these projects will be working in SESAME. In addition, a number of instruments will be deployed for shorter periods during the intensive phases of SESAME to investigate specific phenomena, such as the chemical activation by PSCs. Over 50 ground-based instruments are being deployed at over 30 sites in Greenland, Scandinavia, Siberia and mainland Europe (see map), measuring many nitrogen and chlorine species. Measurements will also be available from 117 instruments in the global ozone observing network.

An important consequence of changes in stratospheric ozone is the resulting change in the fluxes of ultraviolet (UV) radiation reaching the surface. This will be studied at a number of sites during SESAME, making both regular and intensive spectral irradiance measurements during the more active phases of the campaign.



UV-visible spectrometers are widely in use for the monitoring of ozone, nitrogen dioxide, chlorine dioxide and PSCs. Because the measurements are performed in the visible during twilight, they are able to provide daily data, up to the latitude of the polar circle in winter, in all weather conditions, when more traditionnal UV instruments are blind. The picture shows two automatic SAOZ spectrometers developed at CNRS, under parallel testing in extreme conditions at ESRANGE, Kiruna, Sweden, in winter.



The launch of a combined KFA-Julich/JPL payload during SESAME in February 1994.

OZONESONDES

The vertical distribution of ozone is measured routinely at many locations using small balloons carrying lightweight electrochemical sensors (ozonesondes). During EASOE, the launch frequency was greatly increased with measurements being made at more than 20 stations. Studies of the natural variability and of the ozone loss were made which were previously not possible because of the more limited data available.

A network of ozonesonde sites will operate during SESAME (see map inside front cover). Cooperation with scientists in Japan and Canada will further enhance the coverage of measurements. About 2000 ozonesondes will be launched, mostly in the winter months.

RESEARCH BALLOONS

During SESAME 57 large balloons will be launched, 36 from ESRANGE, near Kiruna, Sweden, and Andoya, Norway, and 21 from mid-latitude sites in France and Spain. Measurements will be made of ozone and of the major reactive species in the nitrogen and chlorine families, including NO, NO₂, HNO₃, ClO, HCl, ClONO₂ (see Box 1). In addition, the sources of these compounds, the N₂O and the CFCs, will be measured along with other important stratospheric species including water vapour. Polar stratospheric clouds and sulphate aerosol droplets will also be investigated

using balloon-borne instruments. A greater range of chemical species will be measured than during EASOE.

For the first time during an Arctic campaign, it is planned to fly three long duration balloons which will circle the globe. Measurements will be made over many days, compared with flights of several hours by conventional high altitude balloons. The long duration balloons will be operated by the Centre National d'Etudes Spatiales (CNES), France, who also have responsibility for the large research balloons.

AIRCRAFT

Three aircraft will contribute to SESAME. They will mainly be based at Kiruna airport during their field campaigns and so will be able to make complementary measurements to the balloon measurements. The German TRANSALL (range 4000km; maximum altitude 9km) will make measurements of aerosols and many chemical constituents in its 5 or 6 field campaigns. The German FALCON (range 2000km; max. alt. 12km) will have two field campaigns in February of each year and will concentrate on making measurements of the vertical profiles and horizontal gradients of the reactive chlorine species. The French ARAT (range 1600km; max. alt. 8km) will spend much of the 1994/95 winter in the field making measurements of aerosols and PSCs in northern Scandinavia.

One of the French CONCORDE aircraft will fly into the cold Arctic stratosphere during early 1995 (SESAME Phase III) in the Zebre project. Study of the contrails produced by the aircraft should provide valuable additional information on a number of the processes involving polar stratospheric clouds.

A fifth aircraft will make measurements after the end of SESAME - the Russian GEOPHY-SIKA, capable of flying with a variety of instruments at high altitudes within the stratosphere (up to 20km). Italian scientists are organising a first mission which will address pre-winter conditions (as in SESAME Phase II).

THEORETICAL COMPONENTS

High quality meteorological information is essential to ensure that the measurements made during SESAME are interpreted correctly. SESAME can call upon the active involvement of the scientists who model the stratosphere using some of the most powerful computers available. These computer models are used in our assessments of what the future holds; tests against observations are a critical component in their validation.

Computer models have advanced greatly in the last few years, in part as a result of the influx of new information from field measurements in campaigns like EASOE. Three-dimensional chemical models of the stratosphere have now achieved such a degree of maturity that they will be widely used in interpreting SESAME

results. In addition, two models will be used in SESAME to produce chemical forecasts of the stratosphere in real-time. These forecasts should help the planning of the measurement strategy for balloon and aircraft flights. The modellers played an important role in EASOE, with many going into the field so that they could be more closely involved with the measurements. This will be repeated in SESAME.

During EASOE a large amount of meteorological support came from the European Centre for Medium-range Weather Forecasts and from the national meteorological services. This information was invaluable in both the planning of experiments (e.g. aircraft and balloon flights) and in the post-mission analysis and interpretation of the measurements. Similar information will be available during SESAME.

Satellite information will be available during and after SESAME to help in operations and in post-mission data analysis. Instruments such as the Total Ozone Mapping Spectrometer and the High Resolution Infrared Sounder on the TIROS Operational Vertical Sounder will provide measurements of the column amounts of ozone. Measurements will continue to come from the Upper Atmosphere Research Satellite, and it is anticipated that the European Remotesensing Satellite 2 - Global Ozone Monitoring Experiment (ERS2-GOME) will be launched and provide useful information on stratospheric ozone during SESAME.



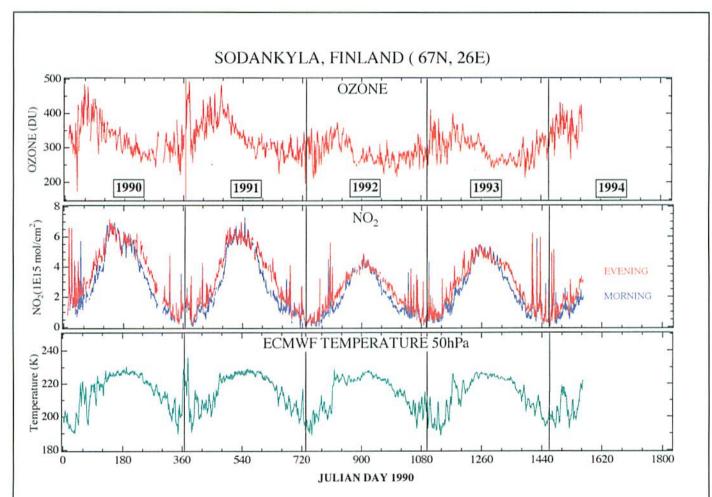
The TRANSALL and FALCON aircraft on deployment at Kiruna airport.

Courtesy J. Büsing

PRELIMINARY RESULTS OF SESAME PHASE I

The first phase of SESAME took place in early 1994. Eight balloons were launched from Kiruna and both the TRANSALL and Falcon were also based there for short campaigns. Early 1994 has been quite different from both the EASOE winter (1991/92) and that of 1992/93. The early part of the winter was warmer than the previous winters and the activation of chlo-

rine appears to have been sporadic. However, stratospheric temperatures dropped in early March and more widespread production of ozone-destroying chemicals could have occurred. The SESAME ozone data is now being examined in detail to find evidence of ozone depletion in the late winter.



The interannual variation 1990-94 of the total ozone, the stratospheric NO_2 (morning and evening) and the air temperature at 50 hPa observed at Sodankyla, Finland.

LIST OF INSTITUTIONS CONTRIBUTING TO SESAME

World Meteorological Organisation, Geneva European Centre for Medium-Range Weather Forecasts

AUSTRIA

University of Innsbruck

BELGIUM

Belgian Institute for Space Aeronomy Belgian Meteorological Institute University of Liège

DENMARK

Danish Meteorological Institute

FINLAND

Finnish Meteorological Institute

FRANCE

Centre Nationale d'Études Spatiales
Laboratoire de Physique et Chimie de L'Environnement
CNRS - Laboratoire Physique Moléculaire et
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Universitè Claude Bernard, Lyon
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AERODATA Flugmeßtechnik Aerologisches Observatorium Lindenberg Alfred Wegener Institute (AWI) for Polar and Marine Deutsche Wetterdienst Deutsche Forschungsanstalt für Luft und Raumfahrt Forschungszentrum Jülich Free University of Berlin Fraunhofer Institut Kernforschungszentrum Karlsruhe Max-Planck Institut für Aeronomie, Lindau Max-Planck Institut für Chemie, Mainz Max-Planck Institut für Kernphysik, Heidelberg Meteorologisches Observatorium, Hohenpeissenberg University of Munich University of Bremen University of Essen University of Cologne University of Hannover University of Leipzig University of Heidelberg

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University of Athens Aristotle University of Thessaloniki

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IRELAND

University College Dublin Irish Meteorological Service

ITALY

CNR - Istituto di Ricerca sulle Onde Elettromagnetiche Istituto FISBAT - CNR University of "La Sapienza", Rome University of l'Aquila

JAPAN

National Institute for Environmental Studies University of Nagoya

NETHERLANDS

Rijksintituut voor Volksgezondheid en Milieuhygiene (RIVM) Laboratory for Space Research, Groningen Royal Netherlands Meteorological Institute

NEW ZEALAND

National Institute of Water and Atmospheric Research

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Bergen Remote Sensing Institute Norwegian Institute for Air Research University of Bergen University of Oslo University of Tromso

POLAND

Institute of Meteorology and Water Management, Aerological Centre

RUSSIA

Central Aerological Observatory Polar Geophysical Institute State Optical Institute

SPAIN

Instituto Nacional de Technica Aerospacial Instituto Nacional de Meteorologia

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NASA Goddard Space Flight Center
NCAR
NOAA Aeronomy Laboratory
State University of New York at Stony Brook
University of Alaska
University of California at Irvine
University of Denver
University of Wyoming
SRI International

SESAME is a campaign of coordinated scientific observations of the stratospheric ozone layer supported by the European Commission. The investigations will make use of combined data sets obtained by many scientists, in collaboration with colleagues from around the world, at a large number of locations, over extended periods of time and using a broad range of instruments. This combination of many different studies has a much greater potential for a significantly improved understanding of global ozone depletion than limited studies carried out in isolation. As such it represents a major European contribution to the resolution of a vital environmental question.

Members of the Science Panel on Stratospheric Ozone:

Prof G.Mégie (Chair)	Université Pierre et Marie Curie, Paris	FRANCE
Dr M.Ackerman	Belgian Institute for Space Aeronomy, Brussels	BELGIUM
Dr B.Carli	IROE/CNR, Firenze	ITALY
Dr R.A.Cox	Natural Environment Research Council, Swindon	UK
Dr J.Farman	University of Cambridge	UK
Prof G.Fiocco	University "La Sapienza", Rome	ITALY
Dr O.Hov	University of Bergen	NORWAY
Prof I.Isaksen	University of Oslo	NORWAY
Prof K.Künzi	University of Bremen	GERMANY
Dr J-P.Pommereau	Service d'Aeronomie, Verrières le Buisson	FRANCE
Dr J.A.Pyle	University of Cambridge	UK
Dr U.Schmidt	Forschungszentrum, Jülich	GERMANY
Prof P.Simon	Belgian Institute for Space Aeronomy, Brussels	BELGIUM
Dr R.P.Wayne	University of Oxford	UK
Prof R.Zellner	Universität Essen	GERMANY
Prof C.Zerefos	Aristotle University of Thessaloniki	GREECE

Members of the SESAME Operational Core Group:

DRWAY
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7
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RMANY
LANCE
RMANY
LGIUM
ALY
2

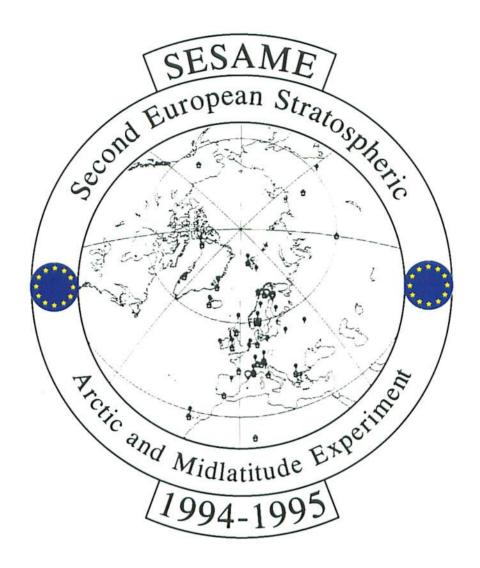


European Commission

Dr H. Ott, Dr G.T. Amanatidis

DGXII/D-1 Environmental Technologies Rue de la Loi 200,B- 1049 Brussels, Belgium Tel.: +32 2 2951182 - Fax: +32 2 2963024

Editing: J. Acevedo, European Commission, DG XII /D1 Environmental Technologies



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Cover: Polar stratospheric clouds in the arctic sky - Courtesy T. Deshler

