# AIRBORNE CHEMISTRY MEASUREMENTS

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#### ABSTRACT

The paper deals with the problems and technical opportunities of the measurement of air chemistry parameters in the atmosphere. These parameters include the concentration of trace substances (gases, aerosol), which are injected into the atmosphere by natural and anthropogenic effects, as well as meteorological parameters which are important factors influencing the space and time distribution of the trace substances. It is shown that atmospheric chemistry research can be performed from aircraft and spaceborne platforms and that in-situ measurement techniques were developed for the determination of trace substances at very low concentration levels (lower ppb range). Examples and results of measurements are demonstrated.

Keywords: air pollution, air chemistry, aircraft measurements, remote sensing, in-situ techniques, space-borne platforms

## 1. Introduction

Because of the increasing problems of atmospheric pollution by man's activities it becomes more and more essential to monitor and control the atmospheric physical and chemical parameters not only on a local but more urgently on a regional and even global scale. Although air pollution is produced at very defined local sources (point sources, line sources, aggregated sources), atmospheric transport and diffusion leads to a local, regional and at last global dispersion of the emitted substances.

As a consequence atmospheric composition has changed since man has begun to burn fuels and generated emission products which are released into the atmosphere. The most severe changing of our atmosphere happened during the last 10 to 30 years due to the enhanced consumption of energy and emission of exhaust gases into the atmosphere.

A changing of atmospheric composition does not only mean danger for all living by breathing toxic substances but also a change of the radiation properties of the atmosphere by constituents a part of which may be harmless for human health. These effects may lead to disturbances of the radiation budget with the consequence of dramatic climate changes (greenhouse effect, ozone hole etc.).

On the other hand many immissions in the atmosphere

undergo chemical and physical reactions by which they will be transformed or by which they will produce additional substances (photochemistry) or by which they will be removed from the atmosphere to ground (precipitation).

Future hazards from these effects can only be estimated by global numerical models. However, all those models need as realistic as possible input parameters which have to be provided by measurements. Furthermore, continuous monitoring of the state and the change of atmospheric conditions are necessary in order to detect possible sudden dangerous situations caused by accidental releases of toxic materials. It is, therefore, desirable to obtain informations on atmospheric conditions on a large scale because relevant events could remain undetected within the grids of a measuring network, at ground.

The only two possible methods to obtain quantitative results on the distribution of the concentration of air pollution constituents, of meteorological parameters and on physical and chemical processes in the atmosphere on a large scale are:

- 1. airborne sounding and
- 2. remote sensing.

Airborne sounding means the performance of any airborne instrument capable of the measurement of the above mentioned parameters. Remote sensing can be performed, of course, from fixed stations at the ground or from airborne or spaceborne platforms.

In the future the continuous global monitoring of the environment including chemical composition and processes in the atmosphere will be a more and more imperative necessity. Such global monitoring can only be performed by remote sensing from spaceborne platforms. Of course, this monitoring will have to be supplemented by airborne and surface-near ground-truth and atmosphere-truth measurements. Also during the development of the necessary technologies the airborne measurement methods have to be developed and performed in order to obtain knowledge on atmospheric composition and processes as well as for tests and calibration of the instrumentation.

# 2. STATE OF SPACEBORNE REMOTE SENSING INSTRUMENTATION FOR TROPOSPHERIC CHEMISTRY RESEARCH

At the moment only few methods are available for the spaceborne measurement of tropospheric trace constituents. A status review was given by a workshop of the NASA (1987) (Ref. 1). The workshop gave the following recommendations:

- to perform a three-layer measurement of CO (in the lower, middle, and upper troposphere) by a nadirviewing gas filter correlation spectrometer
- to perform a lower or middle troposphere measurement of CH<sub>4</sub> by a nadir-viewing gas filter correlation spectrometer
- to perform a survey of lower, middle and upper tropospherice tracer gases by a nadir-viewing high resolution interferometer operating in the thermal emission mode
- to make feasibility studies of the potential spaceborne lidar system for the measurement of aerosols, water vapor, ozone, carbon monoxide, methane, ammonia, sulphur dioxide, nitrogen dioxide, and nitric oxide
- research work in basic studies in tropospheric radiative transfer and spectroscopy
- basic research and development in detectors and associated technologies for the next generation of tropospheric spaceborne instrumentation
- basic research and development in required laser technology

Table 1 summarizes the spaceborne missions for atmospheric chemistry research which were already or shall be performed in the near future. It can be seen that spaceborne missions were already carried through on Space Shuttle, ERBS, and Spacelab. These measurements were performed by passive remote sensing methods revealing medium values or column content values along the light path. In the future efforts should be made to obtain range-resolved results. This can be done by active remote sensing methods. The present state of lidar technology is shown in Table 2, according to Ref. 1. It shows that LIDAR techniques are available for the measurement of aerosol distributions in the atmosphere utilizing Nd/YAG lasers. They were well demonstrated for airborne performance (for example Renger, W., 1989; Ref. 2) but a lot of research has to be done in order to develop techniques for the range-resolved measurement of other species. A DIAL (Differential Absorption Lidar) technique for range-resolved measurements of water vapor was demonstrated by Ehret et al. 1988 (Ref. 3) and just now even airborne measurements had been successful (Ehret, 1989; Ref. 4).

The problems of tropospheric chemistry measurements from space can be summarized as follows (Ref.1):

- electromagnetic waves will be scattered or absorbed by the presence of aerosols and clouds
- wide parts of the electromagnetic spectrum are masked by the presence of water vapor and carbon dioxide
- the very small temperature difference between the surface and the surface-near air is unfavourable

Table 1: Spaceborne missions for tropospheric chemistry research

Species	Platform	Instrument	Mission
со	Space Shuttle	gas filter corr. radiometer (nadir-view. mode)	MAPS 1981/84
H <sub>2</sub> O O <sub>3</sub> aerosols	ERBS	absorpt. spectr. (limb-view. solar occult. mode)	SAGE 11 1984
several trace species	Spacelab 3	high-resol. interferometer (limb-view. solar occult. mode)	ATMOS 1985
aerosols	POES GOES (NOAA) LANDSAT	AVHRR VISSR multispectral sensors	

### **Explanations:**

MAPS ERBS	Measurement of Air Pollution from Satellites Earth Radiation Budget Satellite
POES	Polar-Orbiting Operational Environmental
PUES	Satellites
GOES	Geostationary Environmental Satellites
SAGE	Stratospheric Aerosol and Gas Experiment
ATMOS	Atmospheric Molecules by Spectroscopy
AVHRR	Advanced Very High Resolution Radiometer
VISSR	Visible Infrared Spin Scan Radiometer

Table 2: Laser remote sensing technology

Species	Laser type	Wavel. μm	Comments
Aerosols	Nd/YAG	1.06 0.53 0.355	well demonstrated airborne demo completed
Aerosols	Excimer (raman shift)	0.3 10.6	airborne demo required airborne demo required heterodyne det.
Aerosols	Alexandrite Ti/Sapphire Diode pumped	0.7 0.7 0.5	to be demonstr. to be develop. to be develop.
H₂O	YAG pumped Alexandrite	0.72	to be develop. to be develop.
H₂O	Dye Nd/GLASS Ti/AL <sub>2</sub> O <sub>3</sub>	0.94	to be develop. to be develop. to be develop.
H₂O	CO <sub>2</sub>	10.6	to be develop. heterodyne det.
O <sub>3</sub>	Excimer	0.308	in development balloon test required
O <sub>3</sub>	CO <sub>2</sub>	10	column content balloon test required
NH <sub>3</sub>	CO2	10	to be devel. heterodyne det.
CH₄	no current lasers	7.7? 3.3?	to be develop.
со	no current lasers	2.3? 4.6	to be develop.

- spectral features are blended by the effects of high air pressure which makes the isolation of a particular signature difficult
- the tropospheric presence of ozone and nitrogen dioxide will be concealed by the high abundance of these species in the stratosphere.

During further development efforts airborne measurements of atmospheric parameters and processes have to be performed currently and are indispensable in the future.

The next chapters demonstrate the necessity of airborne measurements of tropospheric chemistry and meteorology and show several examples and problems.

# 3. GENERAL CONSIDERATIONS ON AIRBORNE CHEMISTRY MEASUREMENTS

Airborne sounding can be performed on any platform which is capable of carrying the necessary equipment. The following platforms can be equipped with air chemistry instrumentations:

- aircraft
- balloons
- airships
- dropsondes
- rockets

from which aircraft and balloons are most commonly utilized depending on the particular objectives. Instrumented airplanes are the best platforms to measure atmospheric parameters horizontally and vertically on a large scale while balloons can be employed best for vertically sounding the atmosphere.

Airships may carry heavy payloads of instruments, however they are too slow, too immobile and too expensive for practicable performance.

Dropsondes and rockets will be utilized only for special objectives and their application is limited.

For tropospheric measurements the aircraft has proven to be the best platform to carry air chemistry and meteorological measuring devices.

Not every instrument utilized at ground level is appropriate for airborne performance. Those devices have to fullfill certain conditions and demands (see e.g. Paffrath, D. 1988, Ref. 5 and Cooper, W.A., Baumgardner, D. 1989, Ref. 6). Although nearly all parameters which can be measured at ground can also be measured from aircraft platforms if the instruments are adapted for airborne performance. However, only a few particular atmospheric species, can be measured by remote sensing techniques. Furthermore, most of these systems barely fit in a van not to mention an aircraft with its limitations regarding payload and power requirements.

The basical difference with reference to application between these two alternatives airborne in-situ and remote sensing techniques is:

- airborne in-situ measurements allow a better space resolution and they are very sensitive with a detection limit of less than 1 ppb, but they can be performed only along single lines (flight path)
- remote sensing measurements on the other hand can provide us with informations on the distribution of atmospheric species in a plane through the flight path (mostly a vertical plane) or in a volume around

the platform if a scanner technique is applied. However, the sensitivity is worse than that of the in-situ instruments and many of remote sensing instruments do not allow range resolved measurements.

It is obvious that remote sensing techniques are required for a lot of applications in sounding atmospheric physical and chemical parameters but the state of the art is that presently reliable quantitative results can practically be obtained by airborne in-situ techniques and that remote sensing methods can be performed only in particular cases. Therefore it is very important and necessary to support the development of all remote sensing techniques for the measurement of atmospheric parameters.

# 4. OBJECTIVES AND APPLICATIONS OF AIRBORNE CHEMISTRY MEASUREMENTS

Speaking about airborne air chemistry instrumentation we should note that it is common use to combine chemical measurements simultaneously with the measurement of meteorological parameters because they are closely related with each other.

The objectives of airborne chemistry measurements can be listed up as followes:

- to measure the concentration of selected substances at certain places in the atmosphere (ascertainment of the actual state)
- to measure the two- or three-dimensional concentration field of air pollution components and its variation with time (large-scale overview of atmospheric properties)
- to determine transport and dispersion processes of pollutants in the atmosphere (scientific questions)
- to detect unknown sources of air pollution and to determine the origin of pollutants (environmental surveillance)
- to measure the mass fluxes from point sources (industrial stacks), line sources (highways), aggregated sources (industrial areas, cities) and to determine transboundary mass fluxes (environmental surveillance) transboundary mass fluxes
- to investigate chemical and physical reactions in the atmosphere (photochemistry, oxidation processes, washout and rainout) (scientific questions, tropospheric ozone chemistry)
- to provide data sets as input for simulation models and for the verification and validation of models (scientific questions)
- to forecast air pollution (smog) episodes (warning and emergency measures)
- to monitor the atmospheric conditions and transport of toxic pollutants after accidental releases
- to obain informations on the influence of pollutant components on atmospheric radiation budget and climate (scientific questions on climate research)
- to test and callbrate remote sensing instruments and to obtain ground-truth or atmospheric-truth measurements for remote sensing from spaceborne platforms (quality assurance).

A summarizing report on applications and results of chemistry measurements was given by Paffrath, D. and Peters, W. (1988), Ref. 7).

Fig.1 shows schematically the basic application of an airborne chemistry instrumentation. Looking at the effects which will be caused at the receptor (e.g. the forest) by deposition, it is very important to learn what is going on in the atmosphere between the source and the receptor. The air is the transport medium of any substance emitted by the source. But any material in the atmosphere is not only subject to transport but also to dispersion, transformation, coagulation, turbulent absorption to water droplets, washout, sedimentation and deposition. The effects of any emission from the source on the receptor is a function of all these processes in the atmosphere. The most important effect, of course, is transport and dispersion which will cause, at last, a world-wide distribution of the emitted substance, but also a decrease of the concentration. Many substances will react with others or will be photolyzed to generate new substances (oxidation processes, photochemistry). Thus generally the air composition at the receptor is quite different from that at the source. In order to judge the effects at any receptor we will have to investigate all these processes in the atmosphere, and this can only be done by airborne or remote sensing measurements.

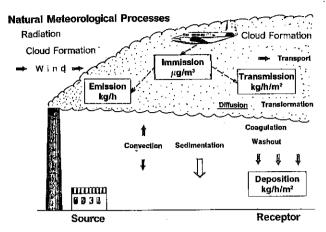


Fig. 1: Schematic view of the application of airborne chemistry measurements

### 5. AIRBORNE IN-SITU INSTRUMENTATION

A typical aircraft instrumentation for air chemistry and meteorological measurements is installed on the DLR Environmental Research Aircraft. Table 3 gives an over-

Table 3 DLR Environmental Research Aircraft equipment specifications

Parameter	Instrument	Range	Detection limit	Time resol.
air pressure	Rosemount Probe	04 800 m.s.l.	0.2 hPa	25 ms
alr temperature	PT-100	- 40 + 40 C	0.01 K	0.5 s
rel. humidity	Vaisala humicap	5100 %	1 %	1 s
SO <sub>2</sub> concentr.	flame photometer	01 ppm	0.5 ppb	1030 s
NO concentr.	chemiluminescence (ozone) analyzer	02 ppm	2 ppb	5 s
NO <sub>y</sub> concentr.	chemiluminescence (ozone) analyzer	02 ppm	2 ppb	5 s
NO <sub>2</sub> concentr.	chemiluminescence (luminol) analyzer	020 ppb	50 ppt	0.3 s
NO <sub>x</sub> concentr.	chemiluminescence (luminol/converter)	020 ppb	50 ppt	0.3 ธ
O <sub>3</sub> concentr.	chemiluminescence (ethen) monitor	05 ppm	4 ppb	5 8
CO concentr.	inrared absorption spectrometer	0100 ppm	100 ppb	10 s
CO <sub>2</sub> concentr.	infrared absorption spectrometer	0.1000 ppm	1 ppm	10 s
aitken nuclei concentration	conden- sation nucleus counter	5010 <sup>7</sup> m <sup>-3</sup>	50 cm <sup>-3</sup> -3	5 s
scatterg. coefficient/ visibility	integr. nephelometer	2 • 10 <sup>-6</sup> 4 • 10 <sup>-3</sup> m <sup>-1</sup> 1100 km	10 <sup>-5</sup> m <sup>-1</sup>	5 s
C <sub>n</sub> H <sub>m</sub> concentr.	air sampling/ gas chromatography	0100 ppb	0.011 ppb	1 s
ion content of aerosol - sulfate - nitrate - chloride etc.	particle collection/ lon chromatography	0100 μg · m <sup>-3</sup>	0.01 0.5μg · m <sup>-3</sup>	5 min 10 min
chemical composition of cloud water	cloud droplet collection (Mohnen collector)	0100 mg/l	0.00201 mg/l	2 min 10 min

view over the equipment specifications.

In the future the spectrum of constituents will be further extended, for instance, additional photooxidants, heavy metals, non-volatile hydrocarbons, halogenated hydrocarbons etc. There are, of course, techniques for the trace analysis of these constituents, however, sampling times are too large in most cases for practical use in aircraft. In the near future measuring methods will be available for the photooxidants hydrogen peroxide  $(H_2O_2)$  and PAN (peroxiacetylnitrate).

Of course, the place, weight and power available on an aircraft is limited and thus the equipment will be limited, too, and in most cases subsequent analysis of collected samples has to be performed in the laboratory at ground. This means that the analysis results can be obtained only after some time (one day and more). Real-time measurement data, - on the other hand - can be yielded by direct analyzing instruments and they may be transfered to ground by radio transmission.

# 6. EXAMPLES OF AIRBORNE MEASURE-MENTS

The surface-near concentration of any air pollutant is a function of

- emission rate
- wind speed and direction
- turbulence conditions
- atmospheric structure
- transformation rates
- removal processes

These effects can be investigated by airborne and partly also by remote sensing measurements. For example, emission rates of  $SO_2$  and  $NO_2$  can be determined by measuring mass fluxes of these components by means of a correlation spectrometer (COSPEC) remote sensing instrument receiving sky light which has penetrated the exhaust plume of a stack. Wind parameters may be measured by means of laser doppler anemometers (LDA). Turbulence may be investigated by a sonar

remote sensing technique which makes use of the back-scattering of sonic pulses from turbulence parcels in the atmosphere (SODAR). Atmospheric structure may be determined by LIDAR techniques by sensing the vertical aerosol distribution from which the formation and presence of atmospheric layers (inversion layers, mixing layer) may be deduced. Transformation rates can be determined by remote sensing only if it will be possible to measure simultaneously the concentration of several reacting substances. At present no available remote sensing method is capable of investigating atmospheric removal processes.

## 6.1 Plume tracking and emission rates

If at any place in the atmosphere a spectacularly high concentration is detected it is possible to discover the emitter by tracking the plume by aircraft in-situ or remote sensing measurement. This, of course, implies the real-time analysis of the recorded data on-board the airplane because the flight path of the airplane has to be fixed immediately during the flight according to the results of the concentration measurements. The emission rate of the idientified source can be estimated by measuring the mass flux of a certain component emitted from the source.

The technique of measuring the emission rate from a defined source as for instance a power plant or a city is demonstrated in fig. 2. At first a box is defined around the source. Measuring flights are then performed on horizontal traverses within vertical planes upwind and downwind of the source. Simultaneous measurements of the wind through these planes allow then the calculation of mass fluxes through the planes. Under certain conditions the calculated flux values may be equated with the emission rates of the sources. These conditions are:

- no large distance of the measuring planes from the source,
- no significant transformation of the investigated species,
- no significant removal of the investigated species from the atmosphere.

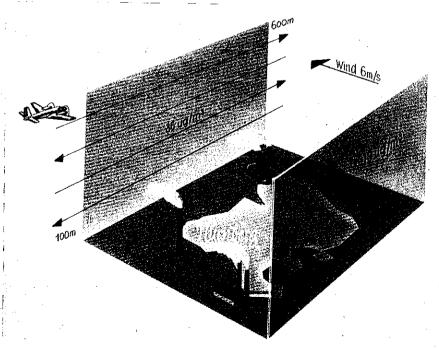


Fig.2: Principle of measurement of mass fluxes from a source (e.g. a city)

From several mass flux measurments we estimated the emission rates of some particular sources as follows:

Table 4 Results of mass flux determinations of some major sources measured in the plumes by aircraft

Source	Flux (t/h)	Date
power plant Schwandorf	11	(1980)
power plant Schwandorf	6	(1987)
power plant Arzberg	2.5	(1987)
industrial area Ingolstadt	4.5	(1987)
city of Regensburg	1.8	(1987)
city of Nürnberg + industry	17	(1987)
Stuttgart area	8	(1987)
Mannheim area	6	1987)
Frankfurt/Mainz area	17	(1987)

# 6.2 <u>Determination of long-range and transboundary</u> mass fluxes

Transboundary mass fluxes of  $SO_2$  and  $NO_2$  could be measured by means of correlation spectrometry remote sensing by Beilke et al. (1985) (Ref. 8) and by Rabl et al. (1987) (Ref. 9). The COSPEC instrument was mounted on a van which was moved along a road parallel to the boundary between two points. However, remote measurement of mass fluxes are restricted to those species

which can be detected by this techniques. These are  $SO_2$  and  $NO_2$ . It is, however, necessary also to measure mass fluxes of other pollutants. This can be done by aircraft measurements in a similar manner as was illustrated in fig. 2. Aircraft measurements along horizontal traverses in a vertical plane parallel to the border are performed simultaneously with measurements of the wind vector. Then the mass flux of any measured species through the plane is given by the product of the concentration c(x, y, z), the wind vector  $\vec{v}$ , the plane element dF, and the cosine of the angle between the wind vector and the normal unity vector of the plane.

In a special project on behalf of the German Umweltbundesamt we measured mass fluxes along the eastern and western borders of the Federal Republic of Germany of the following air pollution components:

- sulphur dioxide
- total nitrogen oxides
- nitrogen monoxide
- ozone
- total suspended particles
- sulfate
- nitrate
- chloride
- ammonium
- sodium and others

An example of the results is shown in fig. 3. A measuring flight was carried through on a route which is denoted by the broken line. (s. for example: Paffrath, D. et al. 1987) (Ref. 10).

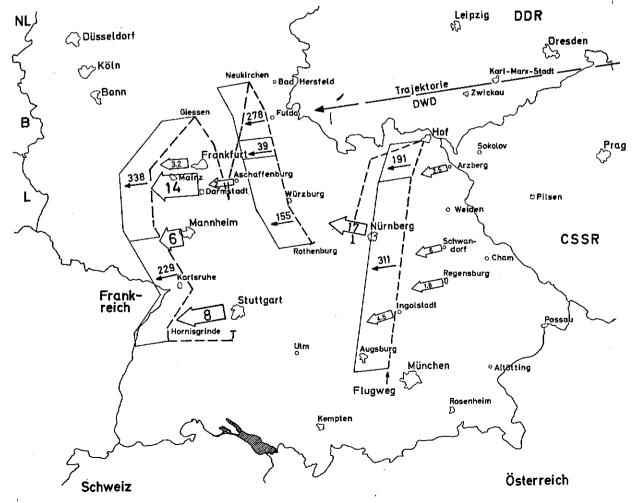


Fig. 3: Example of aircraft measurements of transboundary mass fluxes of SO<sub>2</sub>

The results of the calculation of mass fluxes are presented for long-range transport (numbers in boxes along the flight paths mean tons per hour of  $SO_2$ ) and for particular single sources in the Federal Republic of Germany, for instance power plants, cities and industrial areas (mass fluxes are denoted by numbers in t/h in the arrows). It can be seen that the contribution of the single sources to total long-range transport is rather weak. The total mass flux of  $SO_2$  between Munich and the northern top of Bavaria is about 500 t/h whereas the total contribution of significant single sources is 14.8 t/h which is less than 3 %.

This example shows how important it is to obtain quantitative results for environmental political negotiations regarding transboundary air pollution problems. Furthermore, it may demonstrate the importance of the development of remote sensing methods which would reduce the flight patterns to horizontal lines instead of an expensive scanning of the whole atmospheric mixing layer by aircraft.

# 6.3 Atmospheric structure

Atmospheric structure is operationally monitored by vertical ascents of radiosonde balloons equipped with temperature, pressure and humidity sensors. The data are transmitted by radio communication to the ground. However the resolution of the vertical profiles is poor. Thus in special cases of interest, for example, for judging dispersion of pollutants it can be useful to obtain more precise informations of the layers in the atmosphere. This can be done by means of vertical remote sensing by LIDAR using the aerosol as tracer for dispersion processes. However, not all pollutants will be dispersed and distributed in the same way as the aerosol. To obtain reliable results on the vertical distribution of pollutants they have to be measured directly by vertical ascents of an aircraft.

## 6.4 Investigation of transformation processes

Many investigations on chemical transformation processes have been performed in smog chambers and other artificial environments. However, if one wants to study the processes under natural atmospheric conditions the only possible means are airborne measurements. Measurements from groundbased fixed stations are not suitable for these investigations because the measurements are superimposed by natural variations due to changes of wind direction and velocity. With an instrumented airplane one can arbitrarily scan the concentration field in a pollutant plume and obtain informations on transformation processes between two defined points. Thus by horizontally traversing a power plant plume at different distances from the stack it was possible for instance to obtain reaction velocities for the oxidation of NO to NO. by ozone.

Overall reaction velocities of this reaction could be measured in plumes of a refinery in the Ingolstadt area. The obtained values were between 0.5 and 0.05 % -min<sup>-1</sup>. It should be mentioned that these overall reaction velocities are a function of the meteorological parameters, particularly turbulence and wind velocity because they include the entrainment of ozone from outside the plume into the plume. This means that, if the chemical reaction velocity of the reaction is known from laboratory experiments, for instance, then field measurement results of the overall reaction velocities will allow to draw conclusions on the entrainment phenomenon.

#### 7. Conclusions

In this paper only some selected examples could be presented from the broad spectrum of applications of airborne air chemistry measurements.

It is obvious that many of the airborne measurement results could also be obtained by remote sensing techniques - airborne or rather spaceborne - if only the technology for range-resolved and highly sensitive sensors would allow such measurements. Instead of investigating the three dimensions of the atmosphere by airborne sondes on airplanes which requires a lot of flight hours, it would be easier to scan the atmosphere by remote sensing methods. Thus, in the future, a lot of work has to be done to reach this goal.

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# Remote sensing and the Earth's environment

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