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Re-evaluation and trend analysis of the Payerne ozone soundings

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ABSTRACT

The ozone soundings performed at Payerne with the Brewer-Mast sonde constitute one of the longest time series available in Europe. Instrumental and operating changes have been responsible for inhomogeneities in this time series. This report gives an overview on the historical documentation, as well as the re-evaluation procedures and the homogeneity checks performed. It also provides an ozone climatological overview and an updated trend analysis for the whole measurement period between 1967 and 2000.

This re-processing and re-evaluation of the ozone time series have reduced and partly explained the inhomogeneities found in the earlier versions of this dataset. Comparisons with simultaneous soundings at Hohenpeissenberg and Uccle reveal a few periods when data at Payerne are still suspicious at various altitude ranges. These periods are linked to changes in the operating procedures and instrumental performance which occurred at Payerne. However, the overall assessment of the ozone dataset homogeneity concludes that any further re-evaluation or statistical homogenisation would certainly not bring significant improvements neither in data quality, nor on the trends over the whole data record.

Ozone trend models have been applied to the full dataset, as well as to a subset satisfying a higher quality criterion but with a reduced number of soundings. The trend model with atmospheric explanatory variables delivers more consistent results when based on the full dataset than when based on the subset. Furthermore, ozone trends computed without and with removal of the influence of natural processes do not differ much, with the exception of a thick layer around the tropopause.

In the stratosphere, between 100 and 10 hPa (16 - 31 km), the trends over these 34 years were negative and varied between -2% and -6% depending on the altitude and the season. Trends were not significantly different from zero between 200 and 100 hPa (11 - 16 km). Within the troposphere, the trends were positive and varied between + 10% and more than +15% depending on the altitude and the season. Trends uncertainty is largest in the troposphere, especially in the winter planetary boundary layer.

The variability of the Payerne ozone time series can be largely attributed to dynamic processes. In particular, the variation of the tropopause height is the main natural cause of ozone variability in the lower stratosphere and in the upper troposphere during Winter and Spring. Furthermore, the annual cycle of the ozone trend seems to be related to the one of the tropopause height. The stratospheric aerosol content, the ozone depletion factor (ODF), the solar flux and the Quasi-Biennal Oscillation have less importance in the ozone variability and trend. The combination of the variables used in the model allows explaining the stratospheric ozone trend decrease between the mid 90’s and 2000. These variables may be either natural variables including the stratospheric aerosol load, or natural variables and ODF.

RÉSUMÉ

La série de sondages d’ozone effectués à Payerne avec la sonde Brewer-Mast constitue l’une des plus longues d’Europe. Des changements d’instrumentation et de procédures d’exploitation ont introduit des ruptures dans la série des données, mettant en cause son homogénéité à long terme. Ce rapport présente une synthèse de l’historique de la station, ainsi que les travaux de ré-évaluation et les tests d’homogénéité effectués. Il fournit aussi un aperçu climatologique et une nouvelle analyse de l’évolution à long terme de l’ozone atmosphérique (trend) portant sur l’ensemble de la période de mesure entre 1967 et 2000.

Les procédures de retraitement et de ré-évaluation ont partiellement réduit, voire expliqué, les ruptures présentes dans des versions antérieures de la longue série temporelle. Des comparaisons avec des sondages d’ozone effectués simultanément à Hohenpeissenberg et à Uccle révèlent quelques périodes où le qualité des données de Payerne restent affectées d'un certain doute à différents domaines d'altitude. Ces périodes sont liées à des changements dans les procédures d'exploitation ou dans les performances des instruments. Toutefois, l'évaluation globale de l'homogénéité de la série des sondages d'ozone permet de conclure que toute poursuite de sa ré-évaluation et de son homogénéisation statistique n'apporterait que peu d'améliorations significatives, aussi bien dans la qualité de ses données que dans les trends calculés sur la période complète.

Des modèles de trend ont été appliqués sur le collectif complet des sondages, ainsi que sur un collectif réduit satisfaisant un critère de qualité plus sévère, mais qui réduit sensiblement le nombre de sondages pris en compte. Le modèle de trend faisant appel à des variables explicatives atmosphériques fournit des résultats plus consistants lorsqu'il est appliqué au collectif complet (par rapport au collectif réduit). Par ailleurs, les trends d'ozone obtenus ne diffèrent que peu, selon que l'on introduise ou pas les paramètres d'influence naturels dans les calculs, ceci à l'exception d'une épaisse couche autour de l'altitude de la tropopause.

Dans la stratosphère entre 100 et 10 hPa (16 - 31 km), les trends calculés sur ces 34 ans ont été négatifs et ont varié entre - 2% et - 6% en fonction de l'altitude et de la saison. Ils n'ont pas été significativement différents de zéro entre 200 et 100 hPa (11 - 16 km). Dans la troposphère, les trends ont été positifs et se sont situés entre + 10% et plus de + 15% selon l'altitude et la saison. La marge d'incertitude de ces trends a été la plus grande dans la troposphère, et tout spécialement dans la couche limite planétaire hivernale.

La variabilité que montre la série temporelle des sondages d'ozone de Payerne peut être en bonne partie attribuée à des processus dynamiques. En particulier, la variation de l'altitude de la tropopause est la cause naturelle principale de la variabilité de l'ozone dans la basse stratosphère et dans la haute troposphère en hiver et au printemps. De plus, le cycle annuel du trend de l'ozone semble être relié à celui de l'altitude de la tropopause. Les aérosols stratosphériques, le facteur de destruction de l'ozone (ODF), le flux solaire et l'oscillation quasi-biennale (QBO) jouent un moindre rôle dans la variabilité de l'ozone et de son trend. La combinaison des variables utilisées dans le modèle permet d'expliquer la diminution du trend d'ozone stratosphérique constaté depuis le milieu des années 90, que l'on sélectionne uniquement les variables naturelles (incluant les aérosols stratosphériques), ou les variables naturelles et l'ODF.

ZUSAMMENFASSUNG

Die in Payerne mit der Brewer-Mast Sonde durchgeführten Ozonsondierungen bilden eine der längsten Zeitreihe in Europa. Messtechnische und betriebliche Änderungen waren für Inhomogenitäten in der Messreihe verantwortlich. Der vorliegende Bericht vermittelt einen Überblick über die Stationsgeschichte, sowie über die Neubearbeitung der Reihe und die durchgeführten Homogenitätsanalysen. Er stellt einen Abriss zur Klimatologie des Ozons dar und liefert eine überarbeitete Trendanalyse über die volle Messperiode von 1967 bis 2000.

Die nach einheitlichen Kriterien erfolgte Neuberechnung und -bearbeitung der Ozon-Messreihe haben die in den früheren Datensätzen festgestellten Inhomogenitäten vermindert, beziehungsweise teilweise erklärt. Vergleiche mit gleichzeitigen Sondierungen in Hohenpeissenberg und Uccle zeigen vereinzelte Zeitperioden, während denen die Payerne-Sondierungen weiterhin mit grösseren Unsicherheiten behaftete Werte in gewissen Höhenbereichen aufweisen. Diese Perioden sind mit Änderungen verknüpft, die in den betrieblichen Prozeduren bzw. in den instrumentellen Spezifikationen der Payerne-Sondierungen vorgenommen wurden. Dennoch lässt diese eingehende Untersuchung den Schluss zu, dass weitere Überarbeitungen und statistische Homogenisierungen der Datenreihe keine signifikanten Verbesserungen sowohl in der Qualität dieser Datenreihe wie in den Trends über die volle Messperiode mehr bringen würden.

Zur Berechnung der Ozontrends wurden statistische Modelle verwendet, und zwar auf den vollständigen Datensatz sowie auf einen Teildatensatz, welcher einem strengeren Qualitätskriterium zu genügen hat, aber dementsprechend viele Sondierungen ausscheidet. Das Trendmodell, das auf atmosphärischen Erklärungsvariablen beruht, ergibt konsistentere Resultate, wenn es auf den vollständigen Datensatz angewandt wird, dies im Vergleich zum reduzierten Datensatz. Weiter sind die Trendresultate wenig davon abhängig, ob natürliche Einflussgrössen im statistischen Modell eingeführt werden oder nicht; nahmhafte Unterschiede werden lediglich in einer ausgedehnten Schicht um die Tropopausenhöhe festgestellt.

In der Stratosphäre zwischen 100 und 10 hPa (16 - 31 km) ergeben sich über die gesamte Messperiode negativ Trends, mit Werten zwischen - 2% und - 6% je nach Höhe und Jahreszeit. Zwischen 200 und 100 hPa (11 - 16 km) haben sich die gerechneten Trends als nicht signifikant von Null verschieden erwiesen. In der Troposphäre waren die Trends jeweils positiv, mit Werten zwischen + 10% und mehr als + 15% je nach Höhe und Jahreszeit. Die Unsicherheit, mit welcher diese Trends behaftet sind, ist in der Troposphäre die grösste, besonders in der planetarischen Grenzschicht im Winter und im Frühling.

Die Variabilität der Ozon-Sondierungszeitreihen kann weitgehend auf dynamische Prozesse der Atmosphäre zurückgeführt werden. Der zeitliche Verlauf der Tropopausenhöhe erwies sich als die wichtigste natürliche Ursache für den zeitlichen Ozonverlauf in der unteren Stratosphäre und in der oberen Troposphäre, sowohl im Winter als auch im Frühling. Der Jahresverlauf des Ozontrends scheint ferner mit demjenigen der Tropopausenhöhe verbunden zu sein. Der stratosphärische Aerosolgehalt, der Ozon-Zerstörungsfaktor (ODF), die Sonnenaktivität und die Quasi-Biennale Oszillation (QBO) wirken sich weniger auf die Variabilität des Ozons und deren Trends aus. Die Kombination der untersuchten Variablen gestattet, die Abnahme des Ozontrends in der Stratosphäre seit Mitte 1990 zu erklären, ungeachtet ob nur natürliche Variablen (inklusiv der stratosphärische Aerosolgehalt), oder die natürlichen Variablen und der ODF, benutzt wird.

RIASSUNTO

I sondaggi di ozono effettuati a Payerne con la sonda Brewer Mast costituiscono una delle serie temporali più lunghe d'Europa. Cambiamenti strumentali e operazionali sono responsabili di inomogeneità presenti nella serie. Questo rapporto dà una visione d'insieme della documentazione storica, delle procedure di rivalutazione e dei controlli d'omogeneità effettuati. Il resoconto della climatologia dell'ozono e un'analisi delle tendenze temporali a lungo termine per il periodo compreso tra il 1967 e il 2000 sono inoltre presentati.

La rivalutazione e il trattamento successivo della serie temporale di ozono hanno ridotto, e in parte spiegato, le inomogeneità riscontrate nella precedente versione del campionario di dati. Paragoni con sondaggi simultanei a Hohenpeissenberg a Uccle rivelano periodi in cui i dati di Payerne sono ancora privi di certezza a varie altitudini. Questi periodi sono legati a cambiamenti nelle procedure operazionali e a cambiamenti nella esecuzione strumentale verificatisi a Payerne. Nonostante ciò, la valutazione generale della serie di dati dell'ozono induce a credere che ulteriori rivalutazioni o correzioni statistiche non porterebbero miglioramenti sensibili né nella qualità della serie né nei valori delle tendenze temporali a lungo termine della serie completa.

Modelli di tendenze temporali a lungo termine sono stati applicati alla serie completa di dati così come ad una scelta ridotta in modo da aumentare la qualità della serie temporale, ma ridicendone il numero di sondaggi. Il modello comprendente le variabili esplicative atmosferiche forniscono risultati più attendibili quando sono basati sulla serie completa piuttosto che sulla serie ridotta. Inoltre, il modello non dà risultati significantemente differenti che includa l'influenza di processi naturali o meno, con l'eccezione di uno spesso strato nei pressi della tropopausa.

Nella stratosfera, tra 100 e 10 hPa (16 - 31 km), le tendenze temporali a lungo termine sono negative e variano tra - 2% e - 6% a dipendenza dell' altitudine e della stagione, mentre non sono significatamene differenti da zero tra 200 e 100 hPa (11 - 16 km). Nella troposfera, le tendenze temporali a lungo termine sono positive e variano tra + 10% e più di + 15% a dipendenza dell'altitudine e della stagione. L'incertezza delle tendenze temporali a lungo termine è maggiore nella troposfera, in particolare nello strato limite planetario invernale.

La variabilità della serie temporale dell'ozono di Payerne può essere largamente attribuita a processi dinamici. In particolare, la variazione dell'altezza della tropopausa è la più importante causa naturale della variabilità dell'ozono nella bassa stratosfera e nell'alta troposfera in inverno e primavera. Inoltre, il ciclo annuale dell'ozono sembra essere collegato al ciclo annuale dell'altezza della tropopausa. Il carico stratosferico in aerosol, il fattore di dissolvimento dell'ozono (ODF), il flusso solare e l'oscillazione quasi biennale hanno un'importanza minore per quanto riguarda la variabilità dell' ozono e per le relative serie temporali a lungo termine. La combinazione di variabili naturali, come il carico stratosferico in aerosol o l'ODF, possono spiegare la diminuzione delle tendenze temporali a lungo termine dell'ozono stratosferico a partire dalla metà.

TABLE OF CONTENTS

1 INTRODUCTION

Ozone in the atmosphere is of great importance because it prevents the short-wave solar UV-radiation from reaching the Earth's surface in large amounts, where it would be harmful to any form of life. The stratospheric ozone which includes nearly 90% of the Earth's ozone, is referred to as the ozone layer or the ozone shield. Ozone research began more than 100 years ago and strengthened with Chapman who explained in 1930 the ozone layer by photochemical theory. In the early 70's, researchers began to investigate the effects of various anthropogenically released chemicals on the ozone layer, particularly chlorofluorocarbons (CFCs). CFCs are very stable chemically and there are no natural processes that remove them from the lower atmosphere. They are so stable that only exposure to the intense UV radiation prevailing in the stratosphere breaks them down. When that happens, the CFC molecule releases atomic chlorine. One chlorine atom can destroy over 100,000 ozone molecules before being destroyed itself. The net effect is to destroy ozone faster than it is naturally produced by the solar UV-radiation. Large fires and certain types of marine life produce one stable form of chlorine that does also reach the stratosphere. However, CFCs and other widely-used chemicals produced in the 90's roughly 85% of the chlorine in the stratosphere, while natural sources contribute only 15%.

The thickness of the Earth's ozone shield significantly decreased over the last three decades not only over polar regions, but also over mid-latitudes (see review article of Staehelin et al., 2001 and references therein; Harris et al., 1997; Stolarski et al., 1992). The initial concern about the ozone layer in the 70's led to a ban on the use of CFCs as aerosol propellants in several countries. However, production of CFCs and other ozone-depleting substances grew rapidly afterward as new uses were discovered. Through the 80's, the world's nations became increasingly concerned that these chemicals would further harm the ozone layer. In 1985, the Vienna Convention was adopted to formalize international cooperation on this issue. Additional efforts resulted in the signing of the Montreal Protocol in 1987. The original protocol would have reduced the production of CFCs by half by 1998. After the original Protocol was signed, new measurements showed worse damage to the ozone layer than was originally expected. In 1992, reacting to the new scientific assessment of ozone layer, the Parties decided to completely end production of halons by the beginning of 1994 and of CFCs by the beginning of 1996 in developed countries. Because of measures taken under the Protocol, emissions of ozone-depleting substances are already decreasing. The levels of stratospheric chlorine and of most of the other ozone-depleting substances are currently reaching their highest values. They will decrease very slowly as most of them have lifetimes between 50 and 100 years.

In the troposphere, the natural ozone concentrations are low. Tropospheric ozone is formed in the presence of sunlight by reactions with ozone precursors: nitrogen oxides (NO_X) and volatile organic compounds (VOCs). Natural sources, such as vegetation and soils, release these compounds at low concentration. But human activities have strongly increased the amounts released: VOCs from petroleum, chemical industries, and transportation and nitrogen oxides from combustion in power stations and automobiles. Consequently, the ozone concentrations have increased to more than twice their pre-industrial levels in the low atmosphere over densely populated and industrial regions, as well as far away from them through complex atmospheric transport and chemical processes (Staehelin et al., 1994). Ozone concentration varies there quickly in response to precursor's emissions and to weather changes; it peaks by hot sunny day periods. The health and environmental hazards of tropospheric ozone and associated photochemical smog have prompted most countries to impose limits on emissions of nitrogen oxides and VOCs into the atmosphere.

Changes in the atmospheric ozone concentrations also have climate implications (IPCC, 1995). A decrease in stratospheric ozone tends to cool the stratosphere through a negative radiative forcing and an increase of tropospheric ozone has the inverse tendency. In addition, other indirect effects could also impair the natural atmospheric greenhouse balance.

Several techniques, both satellites and ground based, have been developed to measure ozone in the atmosphere. Some detect absorption and emission of solar and atmospheric radiation, and others use chemical titration. Measurements of the ozone layer have been performed since 1920, using spectrophotometers. Ozone profile in situ measurements with balloon-borne small ozonesondes using chemical titration began in the sixties. Long-term satellite based ozone measurements started near the end of the 60's. Microwave and lidar instruments started operation in the second half of the eighties. The systematic atmospheric ozone monitoring has been successively integrated in the framework of different international programmes, especially into the WMO's Global Atmosphere Watch (GAW). The goal of GAW is monitoring the long-term evolution of the atmospheric composition on global and regional scales in order to assess its contribution to climate change and environmental issues.

Switzerland has a long tradition in research and monitoring of atmospheric ozone (Dütsch, 1970, 1992). At Arosa, total ozone (ozone layer) has been monitored since 1926 and this time series has been recently homogenised and its trend re-evaluated (Staehelin et al., 1998, 1999). Swiss ozone profile measurements with ozonesondes started in 1966. The present report is devoted to the re-evaluation and update of the trend analysis of this latter time series, which has been performed in the framework of a national GAW project between 1996 and 2000. It is worth mentioning that the ozonesondes provide the longest and most reliable datasets in the free troposphere and in the low to middle stratosphere up to 25 km (WMO, 1998). Until 1990, the ozonesondes represented the unique operational monitoring technique in the free troposphere.

During the 34 years of balloon ozone soundings, different changes in the measurement site, daily sounding time, operating procedures and data processing occurred. They all have influenced the measured ozone concentrations. Before any trend analysis of such a long time series can be performed, a thorough investigation through the history of the measurements and an appropriate data reprocessing are necessary (SPARC, 1998). Other nearby ozone soundings stations like Hohenpeissenberg and Uccle have already undergone a re-evaluation (Köhler and Claude, 1998; De Backer, 1999).

The present report gives an overview of the different efforts undertaken since 1995 to check the homogeneity of the Payerne ozone sounding time series and to re-evaluate it in order to enable an appropriate trend analysis. This time series is one of the longest of that type in the world and contains the largest number of ascents (more than 4000). Nevertheless its initial goal was not oriented towards long-term monitoring of trends between 0 and 10% per decade, but rather towards stratospheric ozone research (Dütsch, 1986; Staehelin et al., 1991). Chapter 2 of this report is devoted to the history of the time series, to the operating procedures and to the data processing. Chapter 3 provides the different re-evaluation steps performed in the project in order to improve the homogeneity of the time series. This is followed by a check of the re-evaluated Payerne ozone data set through statistical tests and other complementary analysis in chapter 4. Chapter 5 provides a short climatological analysis of the ozone profiles and of the tropopause pressure. Chapter 6 is devoted to trend analyses for the whole period 1967 - 2000 and chapter 7 ends the report with a conclusion.

2 HISTORY OF THE TIME SERIES

Regular soundings with the Brewer-Mast electrochemical ozone sonde have been carried out in Switzerland since November 1966; initially for 2 years as a research project in Thalwil near Zürich, and then since 15 August 1968, as a routine at the aerological station of MeteoSwiss at Payerne. During the first 2 years, these measurements have been performed completely by the Laboratory for Atmospheric Physics, ETH, at Zürich (Dütsch, 1974), then for 20 years with the operational help of the aerological station, and after a few years of increasing involvement in the data handling, under the whole responsibility of the Payerne staff. The research group at Zürich keeps a scientific support and research activities for these soundings.

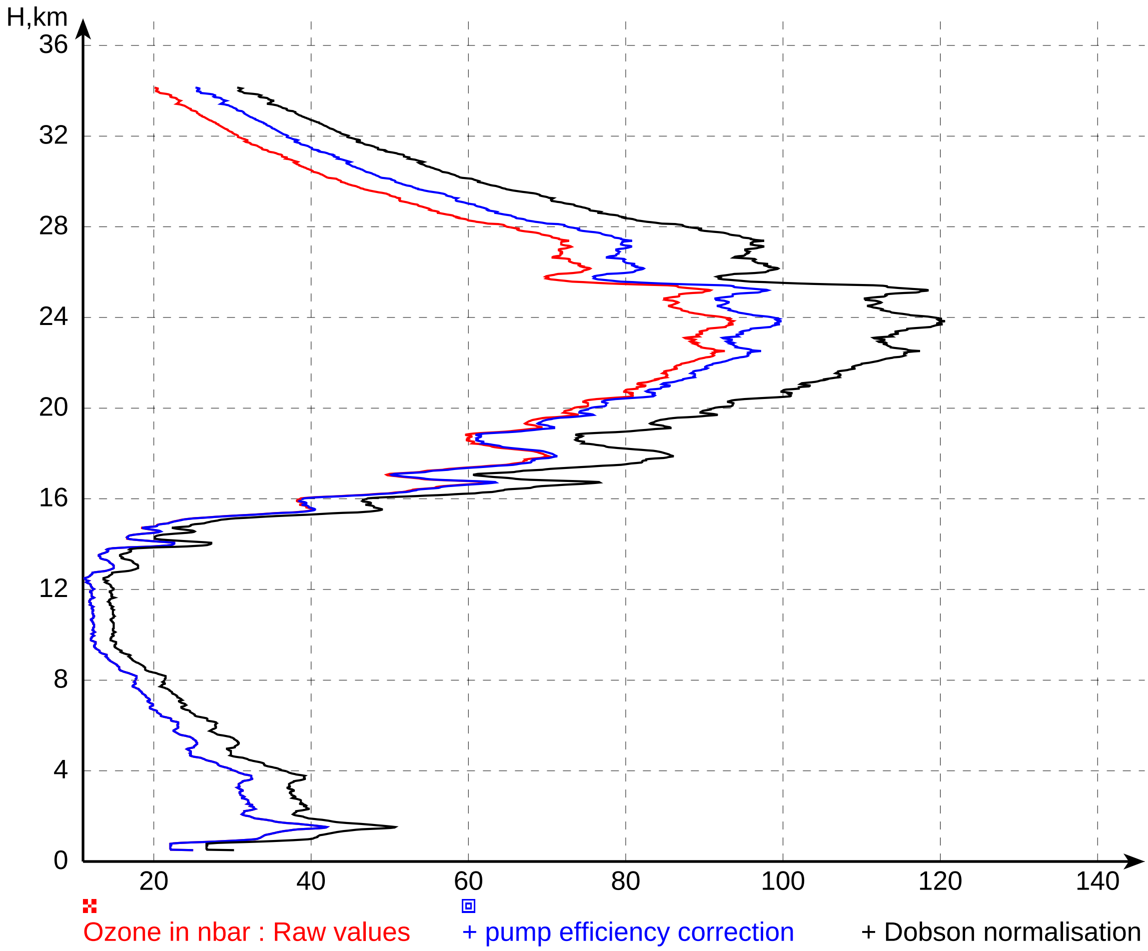
Despite many publications on these measurements since 1966 (e.g. Dütsch, 1974), most of which taking into account measurement uncertainties (Dütsch, 1979; Dütsch and Staehelin, 1988, 1992; Staehelin and Schmid, 1991), a comprehensive written overview on all changes affecting the data quality was still not available a few years ago. Therefore, an “historical” review of all operation procedures, notes, correspondence, invoices and publications related to the Payerne series has been carried out in parallel to an oral questioning of the persons responsible for data acquisition during the 30-year soundings series until 1996. The most important changes have been several modifications of the Brewer-Mast electrochemical cell preparation and calibration procedure, changes of the meteorological sonde, changes in electronics and acquisition system, variations of release time, modifications of the data processing and modifications of the integration programme (Stübi et al., 1996). An exhaustive history of the time series and a description of the data reprocessing performed in 1996 are now available (Giroud, 1996; Bugnion, 1996: internal reports). The most relevant information is summarised hereafter.

2.1 Measurement technique: general overview

The measurement of the vertical ozone profiles is performed using a small sonde carried out by a meteorological balloon that generally bursts at an altitude of 30 - 35 km. The ozone sensor is an electrochemical cell based on the reaction of ozone with potassium iodine (KI). The ambiant air is pumped through the potassium iodine solution and an electrical current is produced between the two electrodes. The ozone sensor is electronically coupled to the meteorological sonde measuring pressure, temperature and humidity. The raw values are transmitted to the ground station with a radio-emitter. The vertical resolution is approximately 50 meters and provides a very detailed vertical ozone profile. The accurate measurement of very low ozone concentrations with such a technique is a challenging task. The Brewer-Mast sensor that is used at Payerne requires very meticulous prelaunch procedures. The data processing takes into account the ozonesonde behaviour during the ascent, especially:

- the loss of pump efficiency at low pressure and temperature (high altitude) by using a pump efficiency equation,
- uncontrolled or unexplained ozone losses, using a scaling with the total ozone column measured with a Dobson spectrophotometer. The total ozone column measured by the sonde up to the balloon burst level is extrapolated up to the top of the atmosphere and the result is compared to the more accurate Dobson value.

Figure 2.1 shows pictures of the sonde as well as ozone profiles for a typical sounding after the main steps of the data processing: after conversion of the measured current to the raw ozone concentration, after correction for the loss of pump efficiency, and finally after the scaling (or normalisation) to the Dobson measurement.



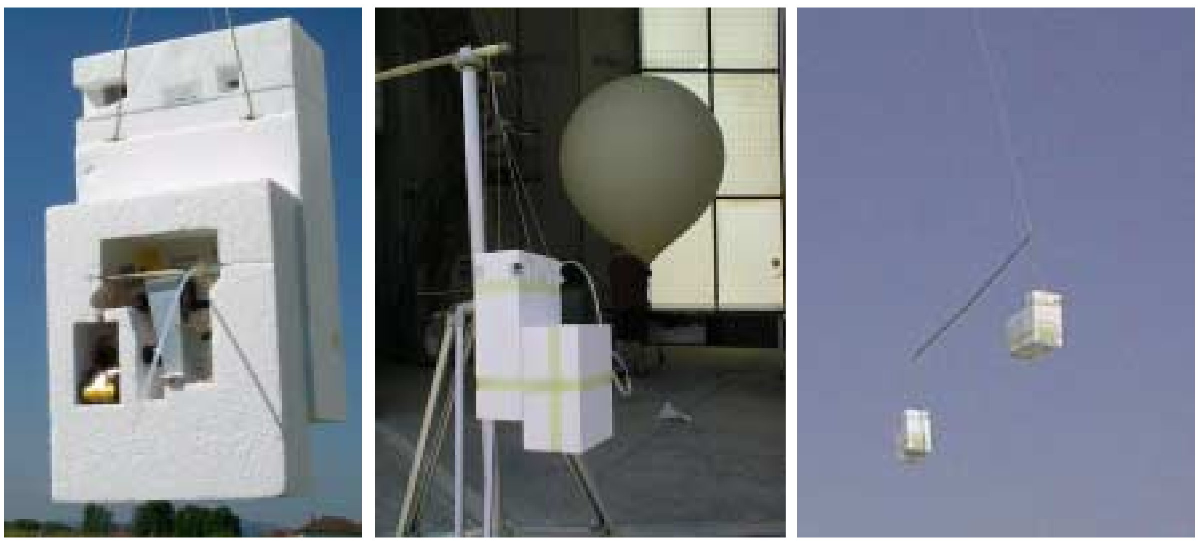


Figure 2.1: Ozone profile of a typical ozone sounding: in red, after the conversion from current to ozone concentration (raw values), in blue, after the correction for the pump efficiency loss, in black, after scaling to the total ozone column measured by a Dobson spectrophotometer.
The pump efficiency is considered not to be impaired until 16 km (100 hPa) and the first two curves overlap between ground level and that altitude. The scaling factor is constant over the whole profile and is 1.2 for this particular sounding.
Pictures below from left to right: ozone sonde without front styrofoam protection, the meteorological sonde is on the other side; sonde and balloon before launch; two ozone sondes under the same balloon soon after launch.

2.2 Measurement programme

Table 2.1 provides an overview of the measurement programme. The aerological station at Payerne is located at an altitude of 490 m (MSL) on the Swiss Plateau. To the northwest are the Jura mountains, at a distance of 25 km, where the highest elevation is approximately 1600 m. The Alps begin 50 km to the southeast, peaking over 4000m. Thalwil is located 140 km ENE from Payerne.

Table 2.1: Main features and changes of the measurement programme (see text in chapters 2.2 to 2.4 for acronyms and explanations).

Period	Site	Ozone sonde and interface	Data acquisition and resolution	Meteo. Sonde	Launch time (UTC)	Data processing and QC
Nov. 1966 - 31th Jul. 1968	Thalwil	Original BM 730-5 VIZ interface	Analog chart Only characteristic points	VIZ	09	At Zürich T_p = 300K (air pump temperature)
15th Aug. 1968 - 08th Oct. 1968	Payerne	“	“	“	04	“
09th Oct. 1968 - 31th Dec. 1971	“	“	“	“	15-16	“
01st Jan. 1972 - 04th Apr. 1977	“	“ BM 730-8 since 1976	“	“	14-15	“
12th Apr. 1977 - 18th Nov. 1980	“	“	“	“	08:30	“
19th Nov. 1980 - 31st Dec. 1981	“	BM 730-8 Special styrofoam box (too thin for a few months) Special interface	Analog + digital (on paper) 30 s (~165 m), but only characteristic points in data files	CH	09:45	At Zürich T_p=280K
05th Jan. 1982 - 31st Dec. 1986	“		“	“	11	“
01st Jan. 1987 - 30th Mar 1990	“	“	Digital (computer) 30 s (~165 m)	“	11	At Payerne T_p = 280K
01st Apr. 1990 - Summer 2002	“	BM 730-8 New interface New styrofoam box	Digital (computer) ~ 8 s (<~ 50 m)	SRS	11	“

Since the beginning there have been usually three soundings a week (normally Monday, Wednesday and Friday). There are only a very few months without any valid sounding. The launch time changed several times, as can be seen in Table 2.1; this introduced significant disruptions in the ozone time series at the lowest levels in the planetary boundary layer.

Since November 1980 the ozone soundings have been included into the upper air operational aerological soundings. The ozone soundings have then no more been carried out with a dedicated system, but have used the infrastructure of the operational aerological sounding (balloon, new dedicated interfacing, meteorological sonde, sonde radio-transmission unit, ground reception unit). The meteorological parameters have been checked with the standard operational quality control. A new styrofoam box has been designed for the ozone sonde at the end of 1980. The next year, it has been improved in order to hinder the icing of the bubbler solution in the middle stratosphere, thanks to a thicker thermal protection and a small water bag placed against the bubbler acting as a latent heat supply.

From 1966 to November 1980 the data were analogically registered on a chart and the characteristic points of the different parameters determined manually on the chart by the operator. From November 1980 to March 1990 the ozone values were also registered in digital form every 30 seconds (until 1986 only on paper strips). The data were then represented on the ozonagramme and the characteristic points selected manually on it. Since April 1990 the ozone values are sampled and stored on computer every 6-10 seconds. The selection of the ozone characteristic points is performed automatically with a dedicated programme.

Up to the end of 1986 only characteristic points exist in the digital archive, and the data handling were performed at Zürich. From January 1987 to March 1990, the measurements were stored on computer at a temporal resolution of 30 seconds (corresponding to a height resolution of roughly 165 m). Since April 1990, the temporal resolution of the stored data is approximately 8 seconds (corresponding to a height resolution of approx. 50 m).

2.3 Ozone sonde and pre-flight operating procedures

The ozone profile time series have been measured up to now with the Brewer-Mast (BM) ozone sensor (Figure 2.2). It is based on the chemical reaction of ozone with a 0.1% buffered iodine solution ($KI + H_2O$) that produces free iodine. This free iodine then causes an electrical current of 2 electrons per ozone molecule between the two electrodes (platinum gauze cathode and silver wire anode) immersed in the solution (Brewer and Milford, 1960). Ambient air is continuously forced into this solution by a small pump. A polarising potential of 0.42 V is applied between the electrodes such that no current will flow unless free iodine is present.

The sonde manufacturer is the Mast-Keystone Corporation. The BM sonde has been only slightly modified during the past 3 decades. The 730-8 model differed from the previous one by an electronic noise free air pump motor and an improved interfacing to the radiosonde. The sonde manufacturing’s quality lowered first in 1989 (see change of the calibration slope on Figure 2.3) and then a second time in 1999, in both cases after relocation of the factory.

A work intensive pre-flight preparation and conditioning of the BM-sonde is vital for getting reproducible measurements. For example, before any launch the sonde has to be exposed to high ozone concentrations. However, in the earlier years this practice was performed only qualitatively. Basically the procedures recommended by the WMO (Claude et al., 1987) have been applied. Changes in pre-flight procedures occurred in 1977, 1983 and the most important one in 1993. Before 1976 the sondes were prepared in Zürich and then transported to Payerne, where they were launched. In 1983 the improved pre-flight protocol of the Hohenpeissenberg group was taken in to account. In 1993 the procedures in exercise were fully re-examined and improved in several points (Hoegger and Levrat, 1993), including complete disassembly and meticulous cleaning of new sondes followed by repeated calibration with fresh sensing solution on each occasion. Each of these improvements were followed by a decrease of the correction factor (CF). 1993 is characterised by the best improvement ever achieved at Payerne.

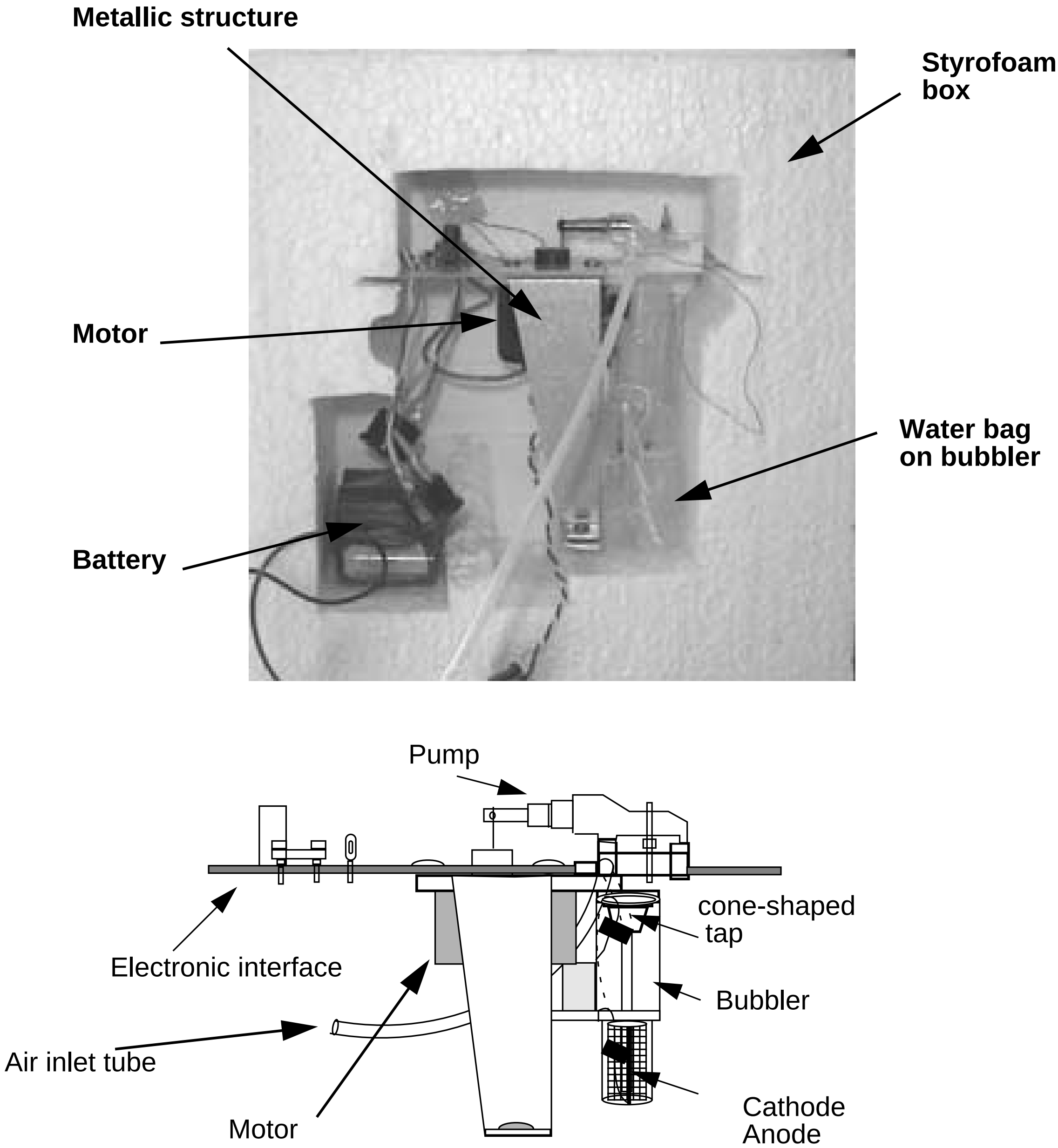


Figure 2.2: Brewer-Mast ozone sonde in the special Payerne rectangular styrofoam compartment used since November 1980. The external dimension of the box is 240 mm. The Brewer-Mast sensor is sketched again below.

Since January 1983, the pre-flight procedure has routinely included a laboratory calibration with a reference ozone UV-photometer. This procedure delivers a calibration slope and an offset for the sonde based on 4 concentration levels. The Figure 2.3 displays these results and gives useful historical information on the sonde preparation work, but does not quite reflect the true sonde behaviour in the atmosphere. Drifts of the ozone UV-photometer impaired sometimes the results of the calibration in the past. This calibration also permits an evaluation of the background current (calibration offset). These different results have been used in the data processing of an alternative time series that is not presented in this report.

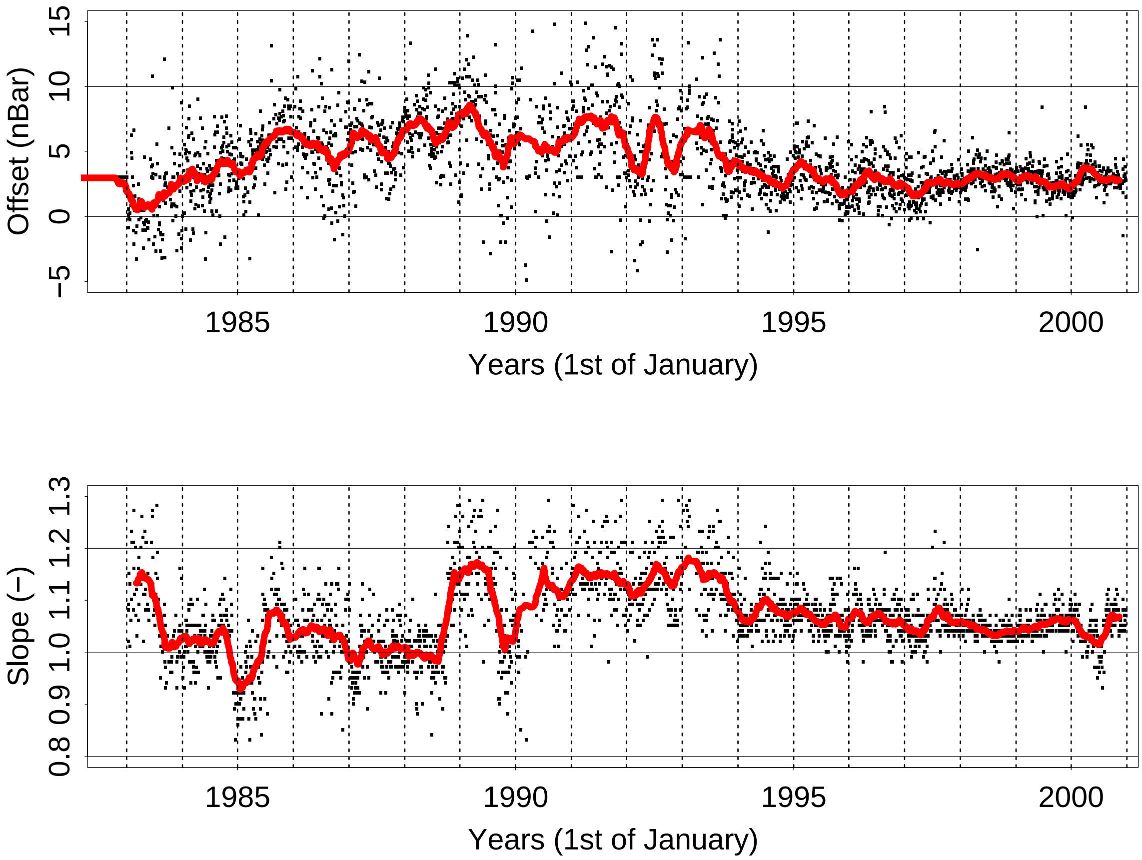


Figure 2.3: Offset and slope of the Brewer-Mast sonde pre-flight calibration for successful soundings with final CF between 0.9 and 1.4 (see Fig. 2.4). Each calibration is represented by a data point. Smoothed curves are drawn. The “normal” offset value of 3 nbar is marked left on the first graph.

From the beginning to 1984, the pump air flow was measured with a bubble meter. Since 1984 this flow is being measured with an electronic Hastings flow-meter. In order to homogenize the flow data, adjustments for earlier systematic errors were applied to the dataset (Bugnion, 1996). However, this mainly adjusted the correction factor CF and had little influence on the final data quality.

From 1966 to November 1980 the Brewer-Mast ozone sonde was connected to the meteorological sonde VIZ. During this period the Brewer-Mast sonde was placed in the original Brewer-Mast styrofoam box.

From November 1980 to March 1990 the Brewer-Mast sonde flew together with the old mechanical Swiss radiosonde (CH). The ozone sonde was integrated in a separate compartment of the radiosonde styrofoam box (Figure 2.2) and got a new electronic interface to the electronics of the Swiss Radiosonde. Nevertheless the ozone data were processed on a special acquisition system and were separated from the meteorological data.

The new electronic Swiss Radiosonde (SRS) was introduced in April 1990. The ozone sonde is packed jointly with a new electronic interface in a separate compartment (Figures 2.1 and 2.2) of the styrofoam box containing the meteorological sensors and the electronics. The ozone data are then received and processed in concert with the meteorological data.

2.4 Data processing

The equation used to determine the ozone concentration from the measured current is:

$$P_{O3} = 4,31 \cdot 10^{-3} \cdot t \cdot T_p \cdot i$$

where:

P_{O3} = Ozone partial pressure [nbar]
t = Time that 100 cm^3 of air at T_p needs to pass through the miniature pump [s]
T_p = Air temperature inside the pump [K]
i = Electrochemical current through the cell [µA]

In this equation, the number of ozone molecules through the cell is converted to the ozone volume concentration in the ambient air. The air temperature inside the miniature pump is therefore needed, but has been seldom measured during the Payerne soundings. The recommended constant value of 300K (Claude et al., 1987) has been used for the original BM-VIZ package from November 1966 to November 1980. As it has been experimentally shown, the special BM packages designed for the two Swiss meteorological sondes (BM-CH and BM-SRS) do not thermally protect the BM pump in the same way as the original BM-VIZ package. According to different experiments in the past, a constant value of 280K has been retroactively applied since the end of 1980. Newer experiments confirm that the quality of the Payerne ozone profiles is improved when measuring T_p during each sounding (Stübi et al., 2002). It is worth mentioning that this temperature is also largely controlled by the battery heat loss and by the styrofoam package design. Consequently, changes in the battery type as well as adjustments in the styrofoam package have already modified the temperature balance of the pump.

The time t in the equation is measured in the pre-flight calibration and introduced in the data processing. The pump efficiency diminishes with decreasing air pressure during the sounding. This has been taken into account over the whole period by the standard pump efficiency correction recommended in Claude et al. (1987). Other groups proposed alternative pump efficiency functions (De Backer, 1999; Steinbrecht et al., 1997; WMO, 1998). Experiments are also under way at Payerne (Stübi et al., 2002).

According to the standard operation procedure (Claude et al., 1987), the integrated ozone column from each single sounding is scaled to the total ozone measured by a Dobson spectrophotometer (daily means) located at Arosa, which is about 200 km to the east of Payerne (Arosa is 90 km to SE of Thalwil). As the accuracy of the Dobson measurement is approximately ± 2%, this correction factor (CF) allows a valuable quality improvement of the sounding profiles measured with the Brewer-Mast sensor. By strong horizontal gradient of the ozone distribution, the distance between Arosa and Payerne leads to additional errors in the adjustment of single soundings; this should however cancel out in mean values. The height difference between Payerne and Arosa is taken into account by integrating the ozone soundings only above the height of Arosa. The ozone column above burst level of the sounding up to top of the atmosphere is approximated by assuming that the ozone mixing ratio is constant above that height. The exact Payerne procedure is as follows:

- For soundings with a balloon burst height above 8 hPa, the mean ozone value between 10 and 8 hPa is used.

- For soundings with balloon burst heights between 17 and 8 hPa, a constant ozone mixing ratio set on a averaged value over the last 2 hPa is used.
- For soundings with balloon burst height between 30 and 17 hPa, a SBUV climatology for northern latitudes between 40 and 50 degrees is used (Solar Backscatter UV Radiometer, onboard NOAA satellites, McPeters et al., 1997).

The error introduced by these assumptions should be rather low as soundings not reaching 30 hPa (24 km) are rejected. Nevertheless, the influence of ozone trends in the upper atmosphere are not duly taken into account (Mateer et al., 1996).

The recently homogenised Dobson101AD series (Staehelin et al., 1998) has been applied to re-evaluate the whole sounding time series. In case Dobson values were missing for the period 1978-1993, TOMS values were used taking into account an adjustment based on the regression between the Arosa Dobson values and the TOMS values. For other years, values had to be estimated when no Dobson measurements are available (Dütsch, 1979). Since 1994, estimated values are mostly based on the measurements from new satellite instruments available on the internet; they still represent more than 10% of total ozone values.

2.5 Data quality

The evaluation presented in this chapter is based on the state of the Payerne time series after having re-evaluated it according to chapters 2 and 3.

The correction factor (CF) can be used to check the quality of ozone soundings. It is generally accepted that the CF from Brewer-Mast sonde should be slightly higher than 1 (Tiao et al. 1986, Staehelin et al., 1991 and 2001). Large deviations from unity generally indicate measurement problems. However, a value close to unity can also occur when ozone is overestimated in the troposphere and underestimated in the middle stratosphere. More or less stringent limits of CF's have been used worldwide for the rejection of ozone soundings: e.g. CF out of the range 0.9 - 1.2, 0.9 - 1.25, 0.9 - 1.35, or 0.8 - 1.4 (WMO, 1982, 1987; Staehelin et al., 1991; Miller et al., 1995; Bojkov and Fioletov, 1997; Logan, 1985; Logan et al., 1999). Brewer-Mast ozone profiles with CF outside the range 0.8 - 1.4 should be rejected (WMO, 1982). The annual statistics for the data of Payerne show a decrease of the CF during the past 30 years indicating a general improvement of the data quality throughout this time. Applying the stringent limits of 0.95 - 1.25 to the Payerne series leads to severe loss of data. 1200 soundings out of 4000 are lost and several months up to 1976 have no or too few soundings.

Finally, two datasets have been selected in order to enable a comparison. The range of 0.9 - 1.4 has been retained as normal selection criteria and the range of 0.95 - 1.25 for a higher quality dataset. Ozone soundings off the first range have been left out in this report as well as in the data submission to the world data centre. 1972 is the year with the fewest soundings satisfying the first quality criterion (38 soundings). The outcoming statistic for this selection is presented in Figure 2.4. Three breaks are clearly visible in this figure, the first between 1975 and 1976, the second between 1983 and 1984 and the third between 1993 and 1994. Those breaks coincide with changes in the preflight procedures as explained in chapter 2.3. Not only the median values show improvements, but so do also their distribution. The interquartile range remains under 0.1 since 1994.

Figure 2.5 is similar to Figure 2.4, but applies to the higher quality dataset. For all years until 1983, the number of soundings drops very strongly.

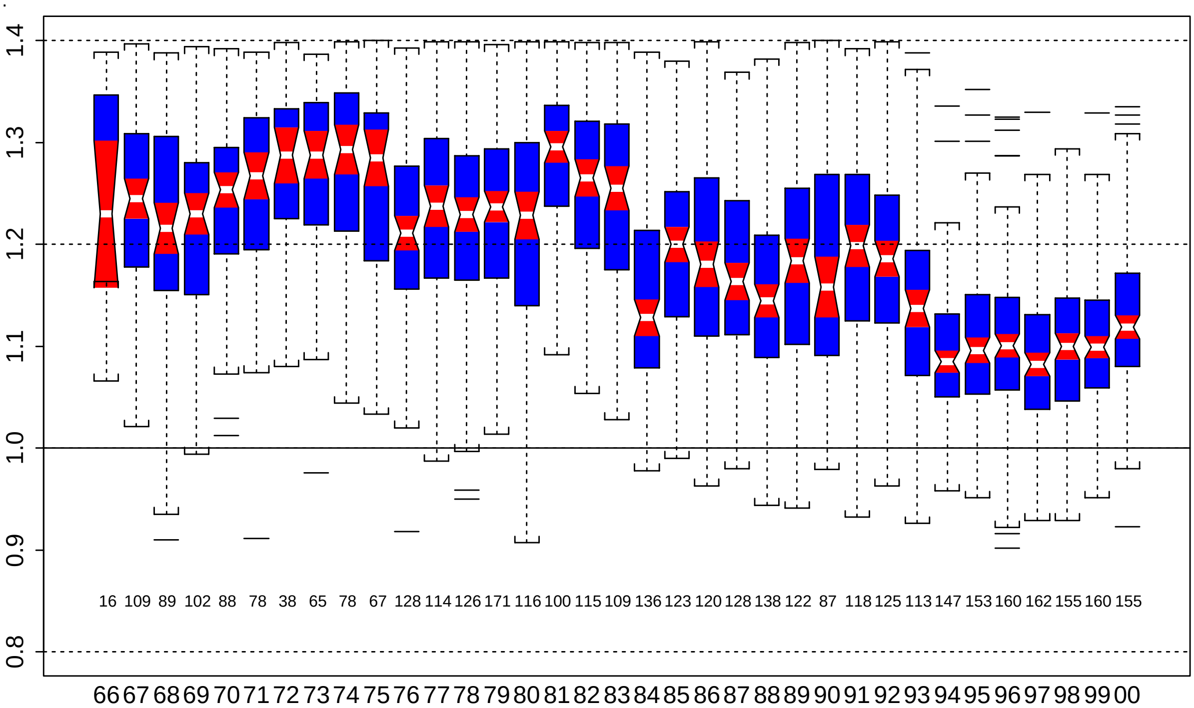


Figure 2.4: Yearly statistical distributions of the correction factors (CF) of the re-evaluated ozone time series. The median values are depicted by the horizontal thick blank lines and their confidence intervals by the notches. If the notches of two boxes do not overlap, this indicates a significant difference. The box limits correspond to the quartiles 25% and 75%. Only soundings with a CF between 0.9 and 1.4 have been selected. The yearly numbers of selected soundings are given below each boxplot.

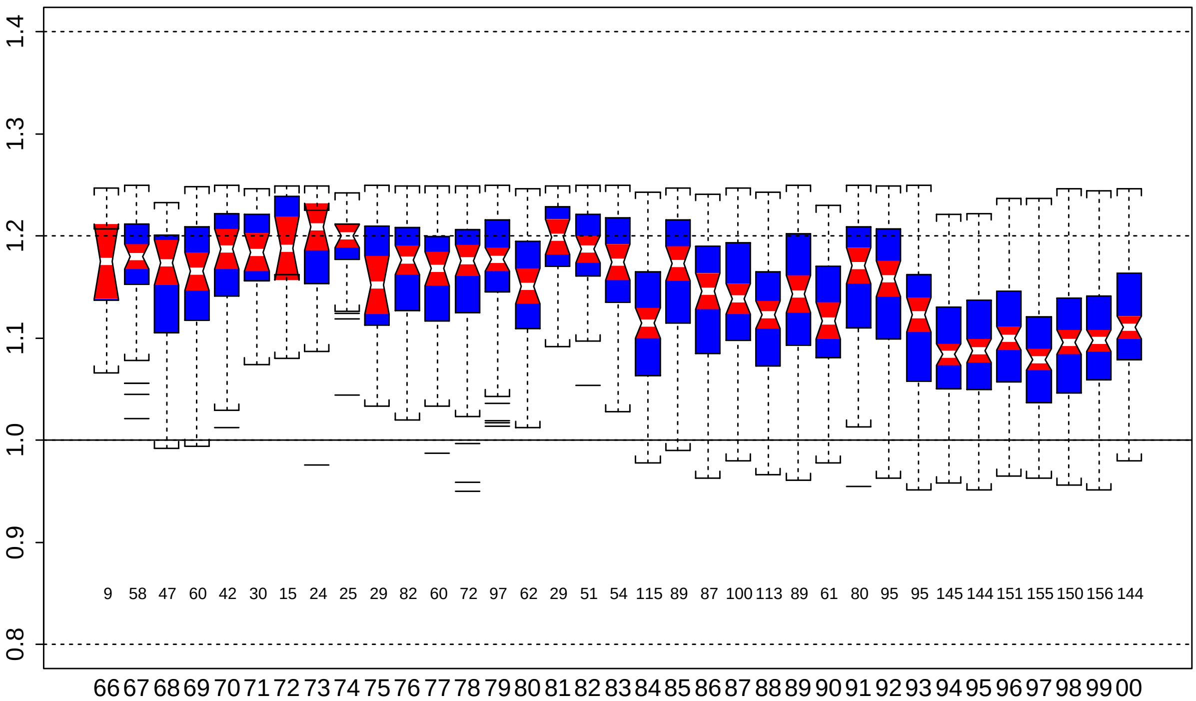


Figure 2.5: Yearly statistical distributions of the correction factors (CF) for the higher quality dataset with a CF between 0.95 and 1.25. Compare to Figure 2.4.

As stated in chapter 1.8.6 of the SPARC/IO$_3$C/GAW report (WMO,1998):

- “The correction factor derived from normalisation of the integrated profile to the total ozone should not be the sole determiner of sonde quality.”
- “Normalisation of the profile using total ozone may provide stability of the ozone time series but also produces some risks that regions with smaller ozone amounts (the troposphere and the stratosphere above 30 km) will be adversely affected if altitude-dependent corrections are not adequately known.”

The pre-flight calibration results could be used as another quality control of the ozone soundings. However, as already stated in chapter 2.3, the pre-flight calibration rather reflects the quality of the sonde preparation work. Furthermore, the calibration slopes only poorly correlate with the CF factors. Large calibration offset values certainly impaired measurement quality in the troposphere. Between 1985 and 1993, it is worth noting that the measured offsets have mostly been well over the value of 3 nbar, which characterises a well prepared Brewer-Mast sonde. Hence, the tropospheric part of the ozone soundings is characterised by a large uncertainty. Taking the offset into account in the data processing brings some improvements when comparing the results with those of high Alpine stations.

Table 2.2: Quality changes at the Payerne ozone sounding

Year or Period	Main problems or changes	Consequences
< Aug. 1968	Site near the town of Zürich	Interferences with SO_2 and NO_2 (Schenkel and Broder, 1982) resulting in too low ozone values in the winter boundary layer (several nbar)
1966 - 1982	Launch hour changed several times	Important in the boundary layer (up to several nbar), medium in the lower troposphere
1976	Brewer-Mast cell preparation moved from Zürich to Payerne	Mean annual correction factor (CF) drops by 5-10%
End of 1980	Change of meteo. sonde, as well as ozone interfacing, package and acquisition systems	Mean correction factor again 5-10% higher Pump temperature change Occasionally problems with the air inlet tube strongly hampering the pump efficiency (until 1995) with main consequences in the stratosphere
End of 1980	Change in the data processing: - pump temperature reduced to 280K	Mean correction factor decreased by ~ 7%
1981	Sonde package upgrade	Mean correction factor 2-3% lower
1983	Operation procedure improved Flowmeter replaced and misused up to 1996	Mean annual CF drops by 5-10% Raw data overestimated by ~ 3% (retroactive correction applied)
1990-1993	Change of meteo. sonde and interfacing problem	Raw data much too high in the troposphere (statistical correction has been performed)
1993	Major improvement of the operation procedure	Mean correction factor stabilized around 1.1 (10% better than before)

Chapter 2.5.6 of the SPARC/IO_3C/GAW report (1998) summarises as follows the quality of the ozone soundings:

- "In general all intercomparison studies have indicated that in the lower to middle stratosphere between the tropopause and ~ 28 km the three sonde types [BM, ECC,...] show consistent results provided the individual measured sonde profiles have been normalised to ground based total ozone column measurements at the launch site. In this altitude range the precision of the various sonde types is within ± 3%, while any systematic bias to other ozone sensing techniques are smaller than ± 5%."
- "For altitudes above 28 km the results are not so conclusive... and the BM sonde show systematic under-estimation with altitude (-15% at 30 km)...".
- "For the troposphere where the ozone concentration are much smaller the results are not consistent at all.... The BM-sonde... showed precision in the range of (± 10-20)%, but there are no indications of any bias larger than ± 5%...".

It is rather difficult to quantify the impact of each change introduced in the Payerne ozone soundings on the measurement errors and uncertainties. Table 2.2 points out some of the encountered problems (Stübi et al., 1996). Claude et al. (1987) estimate errors for the Brewer-Mast sondes by considering their different possible origins. Crude estimates of the final errors on trend calculations have been carried out in the SPARC/IO_3C/GAW report (WMO, 1998). The Table 2.3 below reproduces the preliminary results presented for the Brewer-Mast sonde. As stated there: "They are considered likely estimates for the ensemble of stations using a particular type of sonde. The uncertainty could be much larger, for a particular factor or a individual station."

Table 2.3: Measurement uncertainties influencing trends for BM sondes (%/year), 1970-1996, from the SPARC/IO_3C/GAW report (WMO, 1998), as well as potential drift errors (WMO, 1999).

Trend uncertainty factor	At 5 km	At 20 km	At 30 km
Normalisation to total ozone	0.07	0.05	0.07
Pump efficiency correction	0.20	0.04	0.16
Ozone zero signal (background current)	0.01	0.00	0.00
Pump temperature	0.01	0.00	0.01
Boundary layer / SO_2 pollution	0.05	0.01	0.04
Pressure/altitude measurements (radiosonde)	0.05	0.01	0.04
Unknown procedural changes	0.03	0.01	0.03
Sum of uncertainties	0.42	0.11	0.31
Potential drift errors (WMO, 1999)	0.23	0.066	0.123 at 27 km

Table 2.3 does not apply specifically to Payerne and is even a preliminary attempt (WMO, 1998). Considering the specific history of the Payerne time series, the most important trend uncertainty factors are certainly the procedural changes, the suspected pump efficiency drifts, as well as the sonde "poisoning" in the troposphere. The scaling to total ozone with the re-evaluated time series of Arosa reduces somewhat its detrimental consequences on trend calculations. This correction factor has been improved by ~ 15% over the measurement

period. If systematic error drifts decrease linearly between the begin and the end of the 34 year Payerne time series and amount to 15% at 5 km, to 3% at 20 km and to 10% at 30 km, the systematic error on the trend calculation would be 0.45%/year at 5 km, 0.1%/year at 20 km and 0.3%/year at 30 km. This is in rather good accordance with the sums of uncertainties of Table 2.3. According to WMO (1999), only part of these uncertainties results in potential drift errors in the data (see last line of Table 2.3)

At the beginning of the Swiss measurements (1966), the Brewer-Mast sonde was the only choice. Nevertheless, its pump design (plastic components assembled with glue, metal piston, lubrification oil, dead volume,...) is not optimal for accurate and reproducible ozone measurements due to the high reactivity of ozone and the low air pressure encountered in the stratosphere. The difficulties to prepare the BM sonde has also been since a long time a concern. The alternative is the newer ECC sonde, which is easier to handle and delivers better results. However, the risk of introducing a large discontinuity in the time series delayed the change of the Payerne ozone sensor to Autumn 2002, after having completed intensive comparisons between BM and ECC sondes (Stübi et al., 2002). The international comparison at Vanscoy (1991) already showed that the precision of the ECC sonde was better than that of the BM. The "precision of the ECC was found to be about 2 nbar for most pressures greater than 10 hPa,...". The results of the STOIC experiment were similar: "1.5 nbar between the surface and 100 hPa, decreasing to 3 nbar at 50 hPa, and then increasing and remaining constant at 2.5 nbar between 30 and 4 hPa" (Komhyr, 1995). The BM precision proofed also to be less, especially in the troposphere (poisoning by other atmospheric constituents) and in the middle stratosphere (pump deficiencies). The conclusions of the JOSIE experiments are the following: "The Non-ECC type sondes show considerably lower precision in both the troposphere and stratosphere compared to the ECC types" (Smit, 1996).

3 HOMOGENISATION AND RE-EVALUATION

A pre-requisite to trend calculations is a homogeneous, or at least uncertainty-characterised time series. Quoting Conrad and Pollak (1950): “A numerical series representing the variations of a climatological element is called homogeneous if the variations are caused only by variations of weather and climate”. Actually long-term atmospheric time series are never homogeneous. Two complementary ways allow for a reduction of discontinuities:

- Re-evaluation (or experimental homogenisation):
  Re-evaluation encompasses procedures that improve a series on the basis of new theoretical or experimental results. In the field of upper air ozone soundings, this technique is usually based on laboratory experiments and on simultaneous soundings with different ozone profiling systems. In the case such experiments benefit from a reference instrument, it is comparable to a calibration.
- Statistical homogenisation:
  Statistical homogenisation procedures remove inhomogenities in a time series with statistical tools, based on a station history and a reference time series. Finding a reference time series is the main problem. Climatologists have developed a wealth of such homogenisation procedures for surface time series (see for example Bosshard (1996) and Baudenbacher (1997) who compiled and applied such techniques to Swiss surface meteorological stations). The effect of statistical homogenisation is strongly dependent on the scheme used (see e.g Gaffen et al., 1999) and can influence the results of trend calculations. A correlation between the corrected and the reference times series is also introduced. Such methods should therefore be used with caution.

A frequent source of inhomogeneities is a change in the instrumentation process, which introduces a break in the time series. It can be handled at best by adhering to the first 2 of the 10 climate monitoring principles of GCOS (WMO,2000):

1. The impact of new systems or changes to existing systems should be assessed prior to implementation.
2. A suitable period of overlap of new and old observing systems should be required.

Break removal should then first be dealt with through a re-evaluation, and - if still necessary - using in a second step statistical homogenisation methods.

In the present attempt to provide a homogeneous series of the Payerne ozone soundings, the use of pure statistical homogenisation procedures has been avoided as much as possible and corrections have been introduced only under certain conditions. Consequently, some breaks have been left in the time series and are documented in the station history (chapter 2) or in the homogeneity analysis (chapter 4).

The re-evaluation and statistical homogenisation are presented in the next sections, considering first the ozone measurements, and then the meteorological parameters (pressure and temperature).

3.1 Ozone

3.1.1 Systematic re-processing

The different data sources (ozone and meteorological parameters) have been checked and merged after removal of the inconsistencies. In the cases where temperature and ozone have not been measured at the same pressure values, interpolations have been carried out. Missing altitudes have been calculated in order to have both pressure and altitude for all soundings.

Each sounding has been visually checked and as far as possible compared to the Hohenpeissenberg sounding on the same day. Soundings showing a sudden drop of ozone concentration in the middle stratosphere, most probably due to the freezing of the chemical solution (KI), have been truncated.

The data processing presented in chapter 2.3 has been applied to all soundings (Bugnion, 1996). Where needed, the raw ozone data have been corrected ("Dütsch" factor of 1.04 removed, correction of the pre-flight air flow measurement errors). The programme used for the final ozone computation has been modified in order to reproduce the processing defined in section 2.3. The final result is characterised by quality flags on total ozone, profile extrapolation and integration, special corrections introduced during the first decade, sounding's top, data transmission and electronic noise, and finally calibration.

It should be noted than the raw ozone values have been left unchanged in all database versions. The additional correction modules have been added as needed in the chain of the evaluation programmes. By that mean, the tracability of the whole calculation procedure for the final values is maintained, but no trace of "corrected" raw values is kept in the database. The ozone concentrations before normalisation can be reproduced (= final ozone /CF).

3.1.2 Measurement errors between 1990 and 1993

A major and unexpected ozone break was introduced in April 1990 in conjunction with the change of the meteorological sonde (see chapter 2). The operational amplifier for the current-tension conversion of the ozone electronic interface has been replaced by a 100 Ω Shunt resistance. Simultaneously the load resistance of the previous electronic was suppressed and a new styrofoam box was introduced as well as other minor changes. Between the end of 1992 and 1994, tests were made to explain the break (Kegel, 1995). Major improvements have then been introduced in the operational procedure between March and November 1993, especially related to the sonde conditioning (Hoegger and Levrat, 1993; Giroud, 1996). A load resistance of 4.7 kΩ was added to the new electronic in June 1993. Despite laboratory and flight tests (Kegel, 1995), it was not possible to assign the measurement errors to a specific origin. The search for technical correction factors did not lead to satisfactory results. The load resistance of the electronic interface could not explain the measurement errors. As can be seen on Figure 3.3, the ozone measurements were much too high in the troposphere, but too low in the middle stratosphere; this suggests also possible pump efficiency changes. The impossibility to re-evaluate this break led to use a statistical homogenisation procedure.

The comparison with Alpine surface ozone monitoring stations (Zugspitze, Jungfraujoch) confirmed the anomaly of the Payerne ozone soundings between 1990 and 1993. The anomaly began near April 1990 and ended near March 1993. Its shape can roughly be approximated by a rectangular function, but the second half of this period shows more complexity than the first one. The trend model of Tiao (see Bojkov et al., 1990, Staehelin et al., 1994) was adapted to accept a break function (Kegel, 1995). Its main features are the following ones:

- autocorrelation of order 2,
- seasonal cycle of the ozone concentrations (monthly values),
- break function between April 1990 and March 1993,
- linear trend for the studied period (January 1984 - December 1998),
- external forcing function (either total ozone, tropopause height or stratospheric aerosol),
- model applied on the raw ozone values taking into account the bettering of the normali-

zation factor since 1993, or applied on the final values for verification purpose,

- separate calculations for 25 pressure levels.

The total ozone has been introduced as an external function, because it expresses various forcing processes and is determined using nearby simultaneous measurements. The obtained corrections for the 25 pressure levels are displayed in Figure 3.1, with their uncertainty (one standard deviation) and statistical significance (p-value). In the troposphere, the corrections correspond to almost 20% of the raw ozone values and are highly significant. At 10 hPa and above, the corrections are also highly significant. In the lower stratosphere, the corrections are smaller and not statistically significant.

A purely subtracting and unique correction factor for all these anomalous 36 months was considered as too crude. Therefore a slight annual cycle has been introduced by expressing the jump in percent of the corresponding mean ozone concentration during these 36 months, as well as some minor simplification and adjustments. After applying these proportional correction factors to the raw ozone values of each sounding and normalising them again with total ozone, the same statistical model was applied to the new final monthly ozone values. Small statistically not really significant residual errors remained between 80 and100 hPa as well at 7 hPa. They have been left unchanged, because of the inherent uncertainties of the whole procedure.

Comparisons with the ozone values at Hohenpeissenberg before and after completion of this homogenisation confirm the large improvement of the new Payerne time series, but still show small anomalies at some levels (e.g at 15 hPa). The high ozone values in the low stratosphere before the Pinatubo volcanic eruption remain doubtful (see Figure 3.4). Despite the previous corrections, the ozone values of the Payerne soundings for the time period between April 1990 and Mid of 1993 exhibit larger uncertainties than the rest of the time series.

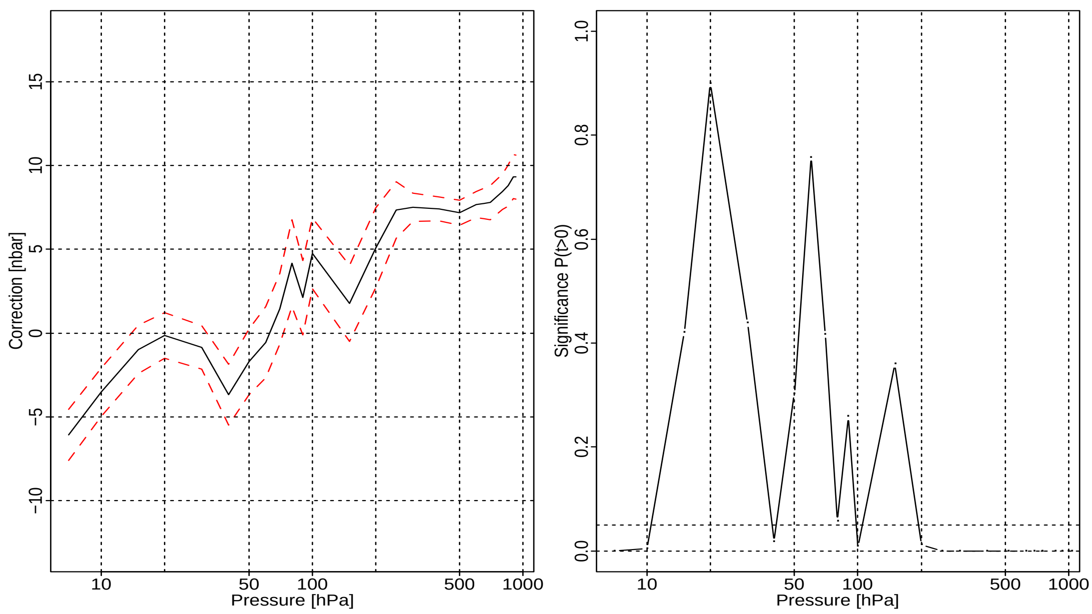


Figure 3.1: Statiscal correction for the ozone measurement errors between April 1990 and March 1993.
Left panel: Correction fonction (to be subtracted to the sounding) with ± one standard deviation.
Right panel: Statistical significance of the correction function (> 95% under the 0.05 line).

3.1.3 Launch time changes

Figure 3.2 shows a decrease of the measured ozone concentrations at 925 hPa during the years 1977 - 1981. At that time, soundings were launched early in the morning. Other changes occurred between 1966 and 1981, according to Table 2.1 and Figure 6.10. Staehelin and Schmid (1991) determined the first corrections on an annual basis. Neuhaus (1998) developed two correction methods for monthly means between 1966 and 1981. The first one is based on the diurnal ozone cycles measured at surface stations at the following altitudes: 500, 1100, 3000 and 3500 m asl. These monthly diurnal cycles are considered equivalent to what soundings could have measured at the same altitudes, as a gross approximation. At 925 hPa, the resulting monthly correction factors vary between 0.85 and 1.6 in Summer. At 650 hPa, these values decrease to 0.98 - 1.03. The smallest corrections apply to winter, summer months require the largest corrections.

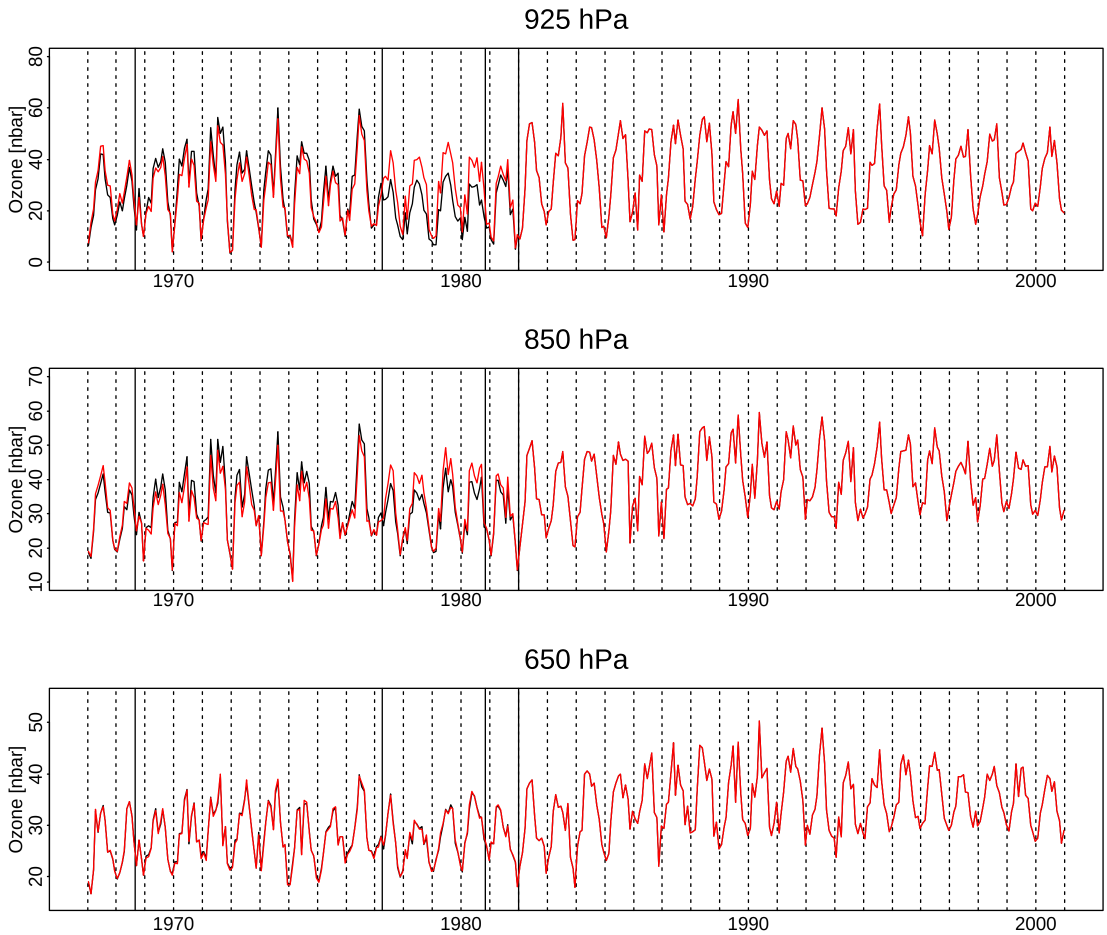


Figure 3.2: Original ozone time series at 925, 850 and 650 hPa (black), as well as corrected time series for the launch time changes between 1968 and 1981(red). The launch time changes taken into account are marked with vertical lines. At 650 hPa, the corrections are so small that the original series disappears behind the corrected one.

Figure 3.2 compares the original series with the corrected one at three pressure levels. The corrected summer values between 1977 and 1980 are still rather low compared to the other years and 2 breaks remain between 1976 and 1977 as well as between 1981 and 1982. Other measurement uncertainties may be suspected between 1976 and 1981 (see chapter 4.2).

The monthly correction factors have not been applied to the individual soundings in the database, because they do not represent measurement errors. Therefore they do not appear in the Figure 3.4. They have been taken into account for the trend calculations (see chapter 6).

3.2 Meteorological parameters

The main sources of inhomogeneities in the meteorological parameters are due to the changes of the meteorological sonde (see chapter 2), whose characteristics are summarised on Table 3.1.

Table 3.1: Instruments of the meteorological sondes used for the Payerne ozone soundings.

Period	Type	Pressure	Temperature	Humidity Wind
Before Nov. 1980	VIZ 1292-1392	1 aneroid (+ hypsometer above 30 hPa once a week 1968- 1980)	Thermistor rod	No No
Nov. 80 - March 90	Swiss CH	2 aneroids	Bimetallic spiral	Goldbeaters skin Radar
Since April 1990	Swiss SRS	Water hypsometer	Cu-Co thermoele-ment	VIZ hygristor Radar

Pressure measurement errors have the largest impact on the ozone profiles in the middle stratosphere. Although 1 hPa error corresponds to only an 8 m altitude error at Payerne level, it expends to a 650 m altitude error at the 10 hPa level (31 km). As the ozone concentrations vary strongly with altitude in the stratosphere, any pressure error sets an ozone measurement to the wrong pressure level. An ozone time series at a specific pressure level is then in fact a time series of ozone values measured at different levels, which depends on the accuracy and precision of the different meteorological sondes (e.g. Claude, 2000). This introduces an additional uncertainty on the ozone trend computed at a given level.

This is also a concern for temperature within atmospheric layers where the vertical temperature gradient is steep. Temperature errors do not affect the accuracy of aneroid pressure sensors as long as the aneroid is mechanically temperature compensated. Therefore a temperature homogenisation does not always directly affect pressure time series. Errors in the pressure measurements are hard to detect and to correct without extensive studies. At temperate latitudes, a pressure sensor giving too high pressure values in the stratosphere, e.g +0.5hPa from 100 hPa up, causes ozone concentrations being 6% too low at levels higher than 20 hPa (Claude, 2000).

Temperature measurement errors represent the main error source for the geopotential altitude at given pressure levels (Richner and Viatte, 1995). This can be explained by the hydrostatic equation which is integrated over pressure slices for the altitude computation. Each pressure introduced in the equation is considered as exact. Only the errors on the virtual temperature impair the altitude calculation and they have two origins: the temperature error itself and the incorrect height and - consequently - temperature of the sonde at the time it registers that pressure. The former is mainly due to a thermal imbalance of the thermometer compared to the air temperature. The latter is not relevant within isothermal layers (e.g ~ lower stratosphere), but each non isothermal layer contributes to the altitude error through the integration procedure. A homogenisation of the temperature leads therefore to an improvement of the geopotential altitudes at pressure levels, but still leaves almost unchanged vertical distortions in the ozone profile due to erroneous pressure measurements. As temperature measurement errors increase with decreasing pressure, the effect of the temperature homogenisation will be most significant in the middle stratosphere. Temperature homogenized geopotential altitudes allow some additional check of the homogeneity of the temperature profiles.

One can conclude that systematic pressure errors have first to be corrected, as they can cause rather large errors on the ozone time series on constant pressure levels. Systematic temperature errors should be corrected afterwards. They have normally no influence on ozone times series on constant pressure levels, but on the geopotential altitude. Temperature trend analyses require homogeneous time series.

3.2.1 VIZ sonde (1966 - 1980)

This VIZ sonde is widely used in the world. Its Ni-span-C aneroid pressure cell has a temperature compensated baroswitch linkage. Intercomparison experiments showed rather good results between surface and 10 hPa, e.g. deviations with other sonde types within ± 0.5 hPa at altitudes above 30 hPa (Richner, 1980; Phillips et al., 1981; Hooper, 1984; Nash and Schmidlin, 1987; Huovila, 1987; Schmidlin, 1988).

One third of the Payerne soundings measured pressure with an hypsometer above the 30 hPa level from 1968 to 1980 (usually every Wednesday). The mean ozone concentrations for the 2 samples stay within ± 1% up to 10 hPa. At 7 hPa, the difference is 5%. This corresponds to a pressure difference of - 0.3 hPa (baroswitch at 7.3 hPa while hypsometer measures 7 hPa). The years 1975-76 show a larger deviation than the other ones and are mainly responsible for that mean drift. As 1/3 of the soundings worked with an hypsometer, the effect on the ozone series would be reduced to ~ 3% ozone deviation at 7 hPa. As the international WMO expert group made no recommendation for pressure correction related to the VIZ sonde and as a consequence of the rather good consistence of the above mentioned results, no pressure re-evaluation has been applied to the Payerne ozone soundings for the VIZ period.

The long VIZ rod thermistor is subject to significant measurement errors from heating by solar radiation as well as from cooling by radiative heat exchange. Several correction curves have been proposed (Richner et al., 1981; Luers and Eskridge, 1998; Nash et al., 1987; Nash and Schmidlin, 1987; Nash, 1993; Schmidlin, 1999). Surface temperature, clouds (including cirrus) and vertical temperature profile all contribute to the complex infrared heat exchange, making any average correction difficult to justify. Discrepancies have been found between different measuring campaigns and sites in the middle stratosphere. Correction proposals or recommendations from different research groups differ noticeably. Steinbrecht et al. (1997) applied the consistency adjustments recommended by Nash and Schmidlin (1987) to the Hohenpeissenberg VIZ dataset. The corresponding temperature corrections all grow with decreasing pressure. Contrasting to that, most of the daytime correction curves of Schmidlin (1999) decrease in the middle stratosphere, and so do the curves proposed by Luers and Eskridge (1998). Finally, as none of the observation sites correspond to the climatology of Payerne and as most of the measurement campaigns were restricted to a short time period, we chose the mean of 9 observation sites from the latest preliminary results of Schmidlin (1999) with a rough interpolation scheme between night and day conditions. From 1968 to 1980, the launch time of the Payerne ozone soundings changed significantly and exposed the sonde to high sun as well as to near night conditions. The largest correction on daytime amounts to -0.7 K between 100 and 70 hPa; at night, to +1.2 K at 5hPa.

The mean geopotential heights at 10 hPa of the soundings with and without hypsometer do not differ significantly. The differences to the meteorological operational soundings with the Swiss CH sonde on the same days have also been calculated for each sample (Neuhaus, 1998). These differences lie below 100 m (85 m at 10 hPa, decreasing below this level until 5 m at 100 hPa). The uncertainty on individual soundings indeed remains larger (order of magnitude of the standard deviation: 100 m).

During 1975-76, some meteorological profiles appear as outliers. They could not be corrected and they were up to now not cancelled out. They are the main responsible for the geopotential deviation still appearing in Figure 3.4.

3.2.2 Swiss mechanical CH sonde (1980 - 1990)

On November 19th, 1980, the ozone sounding switched from the VIZ system (1680 MHz) to the operational aerological system (403 MHz). At the same time the Swiss mechanical sonde (Swiss CH) replaced the VIZ sonde and the data acquisition of the ozone Brewer-Mast sonde has been improved.

The Swiss CH sonde differs from the VIZ sonde in having two pressure sensors switching from the first to the second at the first measurement level above 100 hPa. The dedicated data processing had been based on experimental studies performed at the beginning of 1980 and has been fixed for the whole period from May 1980 to March 1990 (Rieker and Joss, 1985; Richner and Phillips, 1982). Later on, special soundings with the old mechanical sonde CH and the new electronic sonde SRS revealed systematic pressure differences (Häberli, 1996; Kegel, 1996). The measurements were 6 hPa too low at 700 hPa and 4.4 hPa too high at 100 hPa (first sensor). For the second sensor, the measurements were always too low (2.6 hPa at the beginning and 1 hPa at 10 hPa). Hence, the data series has been corrected. The switching between both sensors has been approximated by a linear slope between 100 and 90 hPa. Accordingly, some instability in the ozone profile remains in this range (see e.g. consequences in Figure 3.2). The improvement in the ozone time series can best be seen when comparing the 10 hPa levels in Figure 3.3 and 3.4.

The silver coated metallic spiral of the CH sonde thermometer had a quite different behaviour than the VIZ thermometer. It has a slower time response, is subject to a strong heating by the direct sun radiation, but reflects almost totally the incident infrared radiation. It requires therefore a strong daytime correction, but no significant night-time correction. Experimental studies at the beginning of 1980 led to a polynomial correction function depending on pressure, sonde ascent speed, sun elevation and cloud cover under 70 hPa (Rieker and Joss, 1985). The correction was as high as 5.2 K at 10 hPa. Although some residual errors have been found in the following years (Rieker, 1984), the data processing did not change until 1990. After a new evaluation, an additional correction has been defined and applied retroactively to the whole CH time series. It starts at 150 hPa, increases until 40 hPa, where 0.5 K has to be added to the measurements. Afterwards, the correction changes its sign and increases continuously (measurements have to be lowered by 2.5 K at 10 hPa; the total correction amounts to 7.7 K).

3.2.3 Swiss electronic SRS sonde (since 1990)

A water hypsometer utilizing high-precision thermocouples provides pressure data for this sonde (Richner et al., 1996; Hoegger et al., 1999). The achievable accuracy was estimated to be 1 hPa at sea level and 0.1 hPa at the 10 hPa level, assuming that the boiling temperature could be measured with an accuracy better than 0.03 K. After some years of operational soundings and some improvements in the sensor design, the data processing algorithms could be refined and small adjustments introduced at the beginning of 1999. The biggest pressure correction occurs at 100 hPa (0.4 hPa) and tends to 0 at 10 hPa. Simultaneously, a correction for the sun radiation error on the temperature measurements has been introduced (Ruffieux and Joss, 1999, 2002). This correction has been expressed by a second-order polynomial linear-logarithmic function which is pressure-dependent (-1.7 K at 10 hPa). Slight modifications in the thermocouple calibration have also been implemented (< 0.05 K). All soundings between 1990 and 1998 have been reprocessed accordingly.

3.3 Summary

3.3.1 Data processing

Table 3.2 summarizes the instrumentation changes and the data processing.

Table 3.2: Current state of the Payerne sounding time series

	1966 - July 68	Aug. 68 - Oct. 80	Nov. 80 - March 90	Apr. 90 - Dec. 98	Since 99
Ozone Sonde	Brewer-Mast 730-5 and 730-8 since 1976				
Meteorological Sonde	VIZ - 1292 and 1392		CH	SRS	
Interface	BM - VIZ		BM - CH	BM - SRS	
Brewer-Mast box	Original BM		Modified CH	Modified SRS (~ CH)	
BM operating procedure changes	(site: Thalwil)	1977	1983 (+ pre-flight calibration)	1993	
Pump correction	WMO Standard, T_p = 300 K		WMO Standard, T_p = 280 K		
Normalisation (CF)	Homogenised time series of total ozone measured at Arosa				
Ozone processing	All ozone profiles checked and reprocessed in an uniform way				
Black current	Not taken into account for the standard time series				
Interferences with SO_2 and other species	Not taken into account				
Special corrections	Yes (isolated soundings, according to Dütsch (1974))			Yes (ozone 90-93)	
Launch time correction	Yes, but not in database		No more necessary since end of 1981		
Rejection criteria	Soundings not reaching 30 hPa and with CF outside the range of 0.9 - 1.4 (normal dataset)				
Pressure corrections	No (VIZ)		Yes (CH)	Yes (SRS)	
Temperature correction	Yes (VIZ)		Yes (CH)	Yes (SRS	
Data submission to world data centre (WOUDC)	Last update of the full time series: 1999				

3.3.2 Dataset

As an illustration of the re-evaluation results, Figures 3.3 and 3.4 present the smoothed ozone, temperature and geopotential altitude at selected pressure levels without and with re-evaluation. The smoothing over 12 months eliminates the annual cycle and allows an overview of the three time series. The remaining variations are due to atmospheric processes with long time scales (including natural and man-made changes in other trace gases), to the different number of valid soundings, to changes in the operating procedures, as well as to instrumental changes and problems. This smoothing procedure does not allow a break analysis, which follows in the next chapter.

Figure 3.5 shows the applied corrections (differences between Figures 3.4 and 3.3) with a much shorter time smoothing. Consecutive pressure and temperature corrections can enhance or reduce their effect on temperature and geopotential altitude. It should be reminded that only ozone and pressure corrections have an effect on the ozone time series at fix pressure levels, and consequently on the ozone trends studied in this report.

The biggest ozone changes can be seen between 1990 and 1993 at nearly all levels as well as between 1980 and 1990 above the 10 hPa level. As already stated, the corrections due to launch time changes have not been introduced in Figure 3.4. The relative time variations of ozone are most important in the levels just above and below the tropopause.

The temperature corrections reduced the differences between the different sondes. With some exceptions (1975-76, 1989-90), the temperatures variations are rather smooth. It should be pointed out that these temperature datasets are not the best candidates for trend analyses. The operational night-time soundings should be better us for this purpose.

The geopotential curves follow in most cases the temperature curves, illustrating the close relationship between them. As temperature errors propagate vertically their effect on the geopotential, the largest geopotential deviations are found at the highest pressure levels. These deviations have a magnitude of 100 m at 10 hPa, but reach there more than 200 m in 1975-76. The differences between the different meteorological sondes could not be completely eliminated.

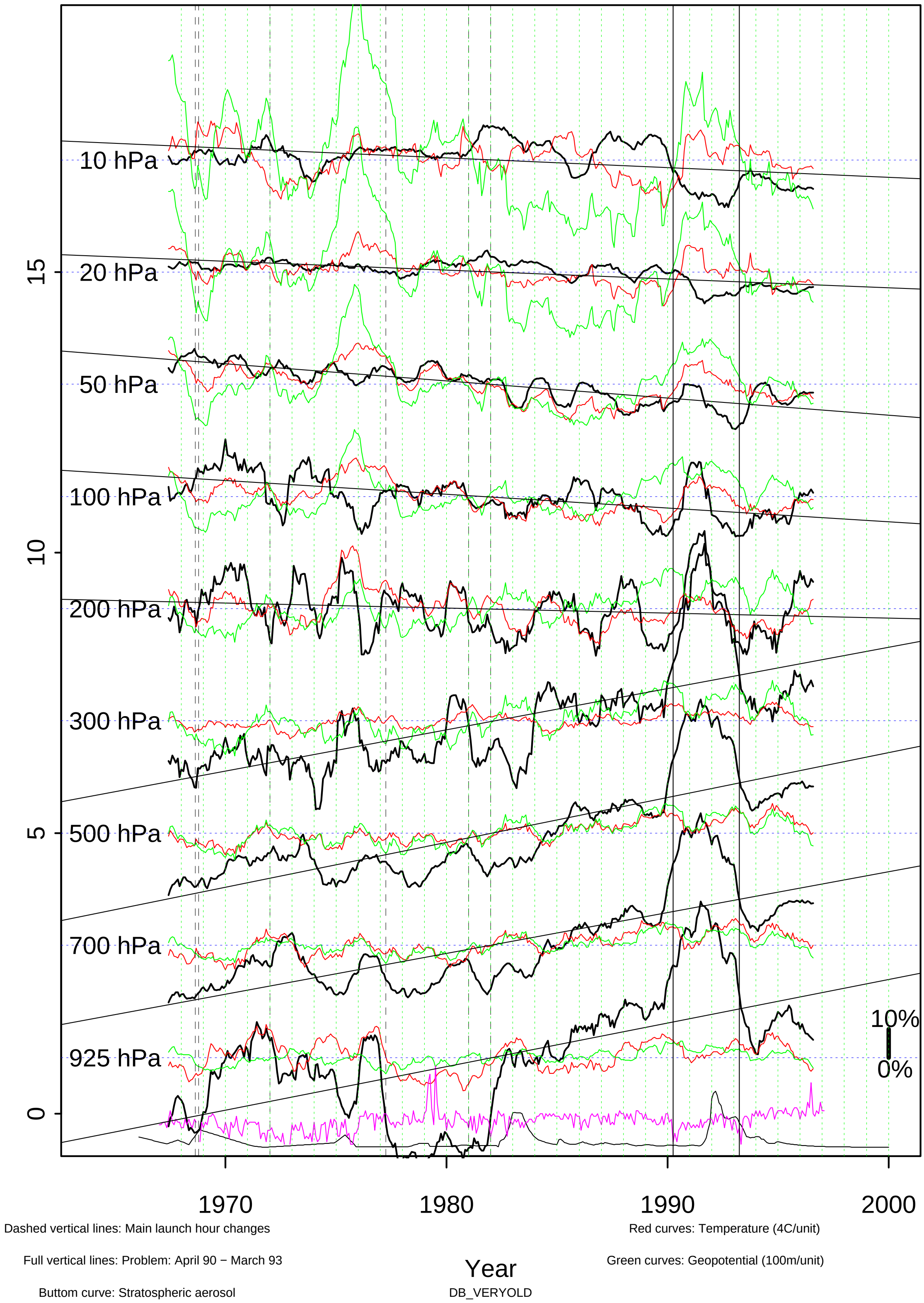


Figure 3.3: Twelve months running means of ozone (black), temperature (red) and geopotential (green) at different levels of the Payerne ozone soundings before re-evaluation (more precisely after the first work in 1996). Ozone values are represented as relative deviations from the mean (scale 10% below right). Temperature and geopotential deviations as absolute values (temperature: ~1 K/cm, geopotential: ~100m/cm). Stratospheric aerosols (index) are represented by the bottom black line, monthly numbers of valid soundings by the bottom pink line (qualitatively). Linear ozone trends are superposed to the time series.The correction for lauch time changes (see Figure 3.2) is not included in the Fig. 3.3 and 3.4.

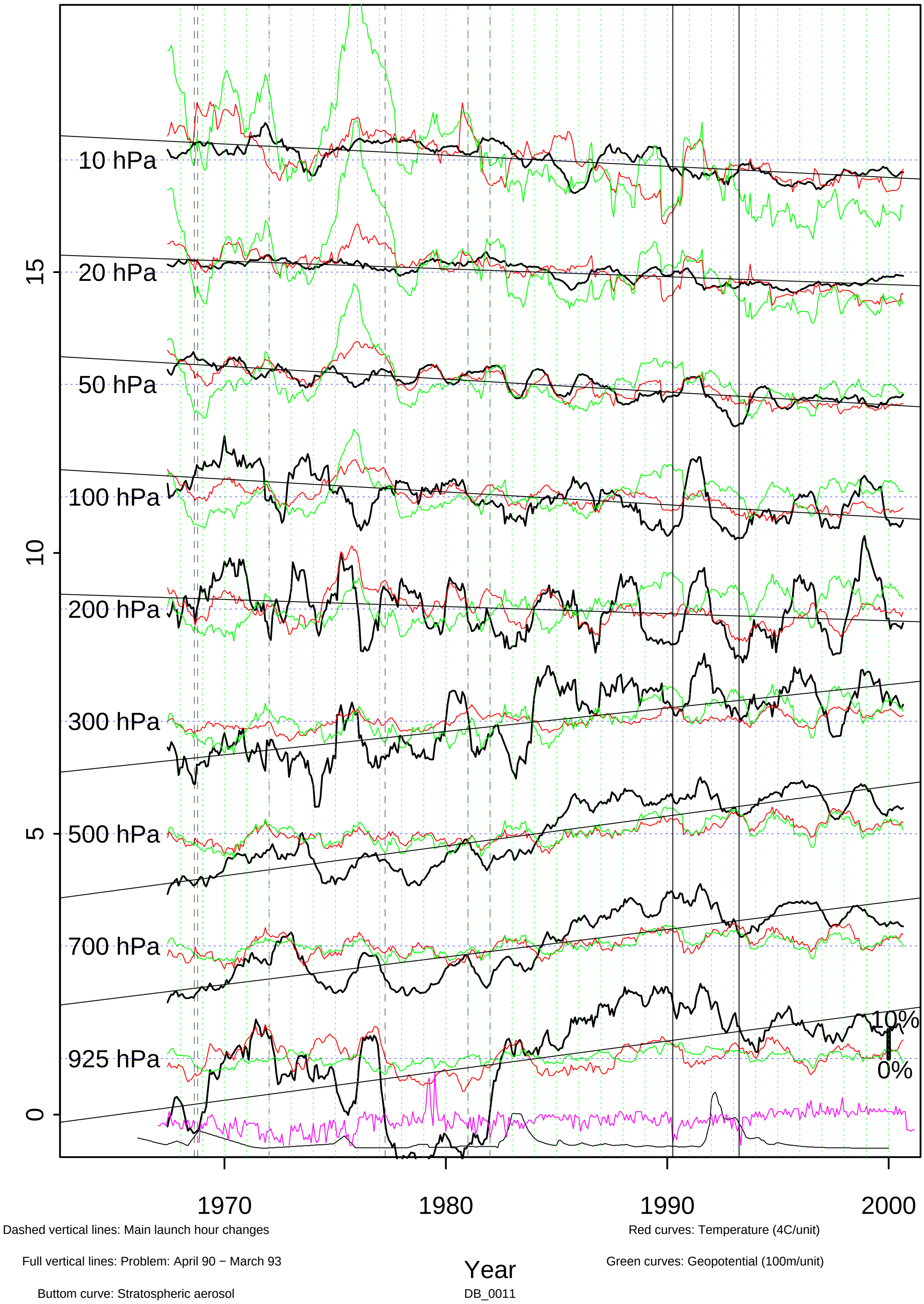


Figure 3.4: Twelve months running means of ozone (black), temperature (red) and geopotential (green) at different levels of the Payerne ozone soundings after re-evaluation. Ozone values are represented as relative deviations from the mean (scale 10% below right). Temperature and geopotential deviations as absolute values (temperature: ~1 K/cm, geopotential: ~100m/cm). Stratospheric aerosols (index) are represented by the bottom black line, monthly numbers of valid soundings by the bottom pink line (qualitatively). Linear ozone trends are superabound to the time series.

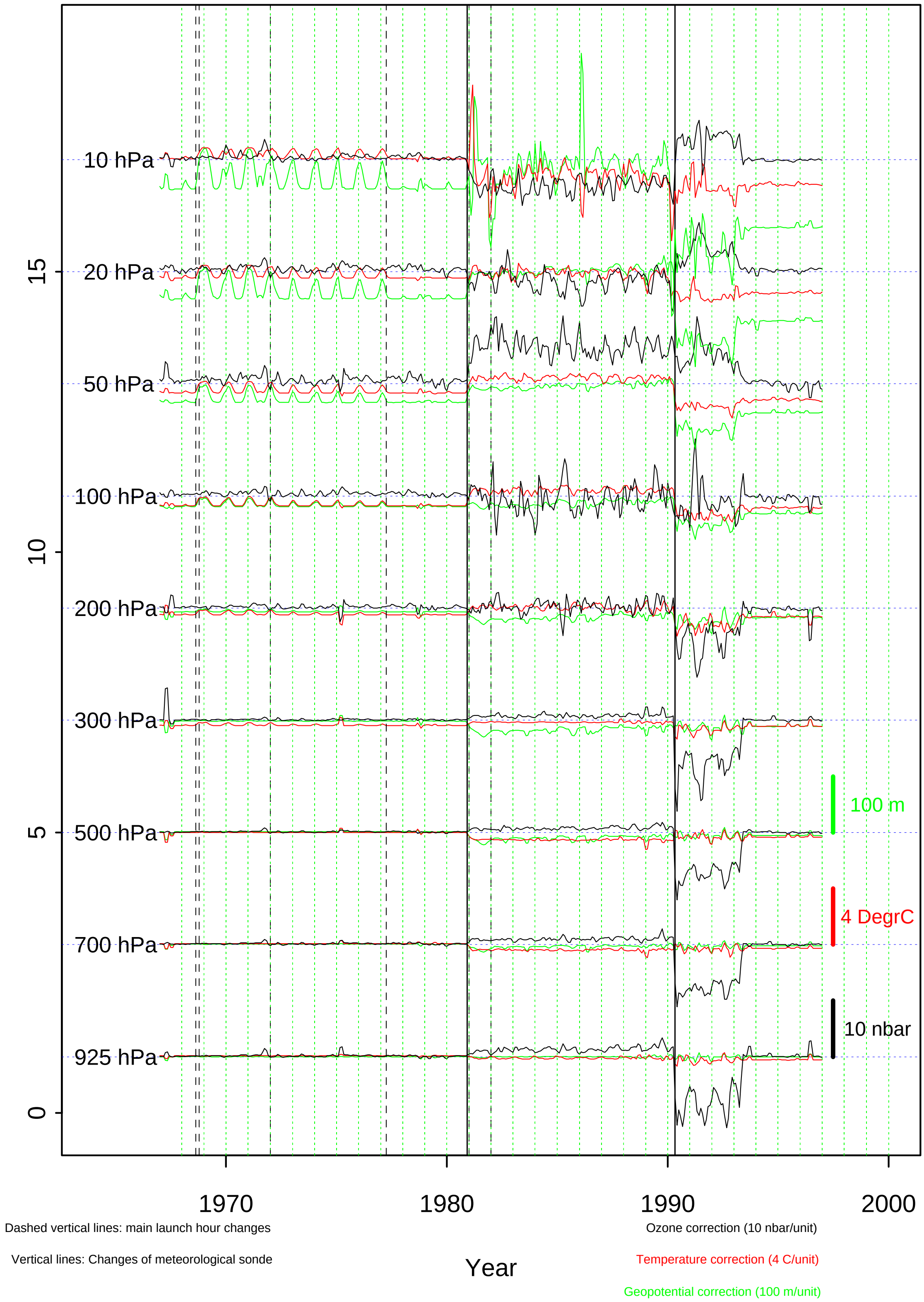


Figure 3.5: Two months running means of the corrections applied to the Payerne time series: ozone (black), temperature (red) and geopotential (green) at different levels of the Payerne ozone soundings during the re-evaluation process. All corrections are represented as absolute values (ozone: ~ 10 nbar/cm, temperature: ~4 K/cm, geopotential: ~100 m/cm).The values represented in this figure correspond to the differences between the corresponding values of Figures 3.4 and 3.3, but with a much shorter moving averaging.

4 HOMOGENEITY ANALYSES

The re-processing and re-evaluation of the ozone time series presented in chapter 3 have removed most of the technical inhomogeneities, but not eliminated all of them. Statistical stationarity (homogeneity of a time series) is an important assumption of most statistical tests and procedures (Lanzante, 1996). In order to get reliable ozone trend results, the homogeneity of the ozone time series has to be checked before trend analyses can be performed. Dedicated statistical tools help finding and qualifying inhomogeneities, which are in most cases abrupt discontinuities (breaks or jumps), but which can also appear as gradual changes (drifts). Such tools are described and used in chapter 4.1. Relationships between ozone and meteorological parameters are then analysed in chapter 4.2, in order to interpret some of the inhomogeneities found in 4.1. Finally, chapter 4.3 is devoted to a final assessment of the Payerne ozone time series.

Inhomogeneities related to technical or operating changes in the ozone soundings should be corrected, but any correction of variations in time that can be related to atmospheric parameters should be avoided.

4.1 Homogeneity analysis of the ozone dataset

4.1.1 Method

In order to separate technical inhomogeneities from time variations of ozone due to atmospheric and climate processes, a reference time series is needed. This one must meet three conditions. First, it must be a comparable time series, second it must be in the same climate region and third it must be free of technical inhomogeneities. Such reference datasets are however not available. In order to partly obviate this problem, the ozone soundings from two European stations that have documented their station history have been used: Hohenpeissenberg in Germany (Köhler, 1995; Claude et al., 2000) and Uccle in Belgium (De Backer, 1999). However, their dataset used in this report could not include the latest corrections applied by their scientists. If a problem is found at the same time in the two comparisons Payerne - Hohenpeisenberg and Payerne - Uccle, it can be attributed with a reasonable confidence to Payerne. This is particularly true for the upper troposphere and the stratosphere, where large-scale atmospheric processes prevail. This approach should hence reduce as far as possible the inhomogeneities of climatic origin. As Hohenpeissenberg is closer to Payerne than Uccle and underwent fewer changes until mid of 1993, priority has been given to the comparison between Payerne and Hohenpeissenberg. The comparisons are performed at specific pressure levels over the entire profile. The altitude range of a break can be used as a criteria in order to discriminate between its different possible origins or as a qualification of its relevance.

For this analysis three different statistical tests and a statistical filter are used simultaneously in order to verify their agreement. With this technique it can be verified if a break is significant for more than one single statistical test.

All three statistical tests identify break points in the monthly time series with a corresponding significance level. They split the time series into a first and a second period separated by an hypothetical break point. Statistical parameters are calculated for the two sub-series and are compared with statistical significance tests. The hypothetical break point is displaced along each point of the whole period excepting some of the first and some of the last points (~ 5 months) and the calculations are repeated each time. The most significant of all hypothetical break points is the final candidate break point. At most one significant break point can be found with these tests. The whole procedure has to be applied to the sub-series in order to find other breaks, or once again to the whole series after having corrected the suspect period. More information on the three tests and on their statistical formulation as well as original references can be found in Bosshard (1996).

Standard Normal Homogeneity Test (SNHT-shift)

The Standard Normal Homogeneity Test checks if there is a significant shift in the mean of the two different sub-series. In this version of the test, only the mean is used to determine the significance of the break point (Alexandersson and Moberg, 1996).

Standard Normal Homogeneity Test (SNHT-var)

This test uses a similar principle to the previous one, but it considers the standard deviation of the two sub-series instead of their mean.

Easterling & Peterson

This test has been specially developed for the detection of trend anomalies. A linear regression is performed on the sub-series and the sum of their quadratic residuals are compared to determine the significance of the break (Easterling and Peterson, 1995).

Kolmogorov-Zurbenko Adaptive (KZA) filter

The KZA is a filter which dynamically adjusts the portion of the time series used for the moving average according to the variation rate of the time series. As the rate increases, the length of data filtered is decreased. The adaptive filter uses an iterative moving average, which separates high-frequency variations from the original data. Contrary to the previous tests, the KZA can detect many breaks without a recursive process (Zurbenko et al., 1996, Hogrefe et al., 1998). This method always finds a principal break, where the filtered curve has the steepest slope. Other breaks with a smoother slope are then identified. The height of the jumps in the filtered data provides a qualitative information on their significance. KZA filters have however two drawbacks: they always identify a break, even in homogeneous series (Ammann, 1999) and they tend to sketch the series by horizontal segments linked with steep ones.

The 3 tests and the KZA filter have been applied to the relative differences of ozone monthly means between the three stations. Only soundings performed on the same days at the 3 stations with a correction factor CF between 0.9 and 1.4 have been taken into account. This reduces their number significantly, but ensures a better meteorological comparability. Linear interpolations for missing monthly means have been then needed, because the tests do not accommodate missing values in the time series.

Test results for periods with interpolated monthly values have to be interpreted with caution. In particular, many interpolations have been carried out from 1972 to 1975 and from 1980 to 1986. From 1983 to 1986, 26 monthly values have been linearly interpolated between the existing values. The time at which the tests assign breaks as well as the breaks themselves are especially uncertain during these periods.

4.1.2 Results

Figures 4.1 and 4.2 summarize over the entire profile from 1969 to 1996 the results for ozone. In order to facilitate the break analysis, the different breaks identified by the SNHT shift test, the SNHT variance test and the Easterling & Peterson test are marked with symbols over the ozone time series filtered with the KZA filter. In order to get a more complete view for the comparison between Payerne and Hohenpeissenberg, secondary breaks have also been computed (not represented on Figure 4.1, only for the longer part of the series cut in two by the first break analysis).

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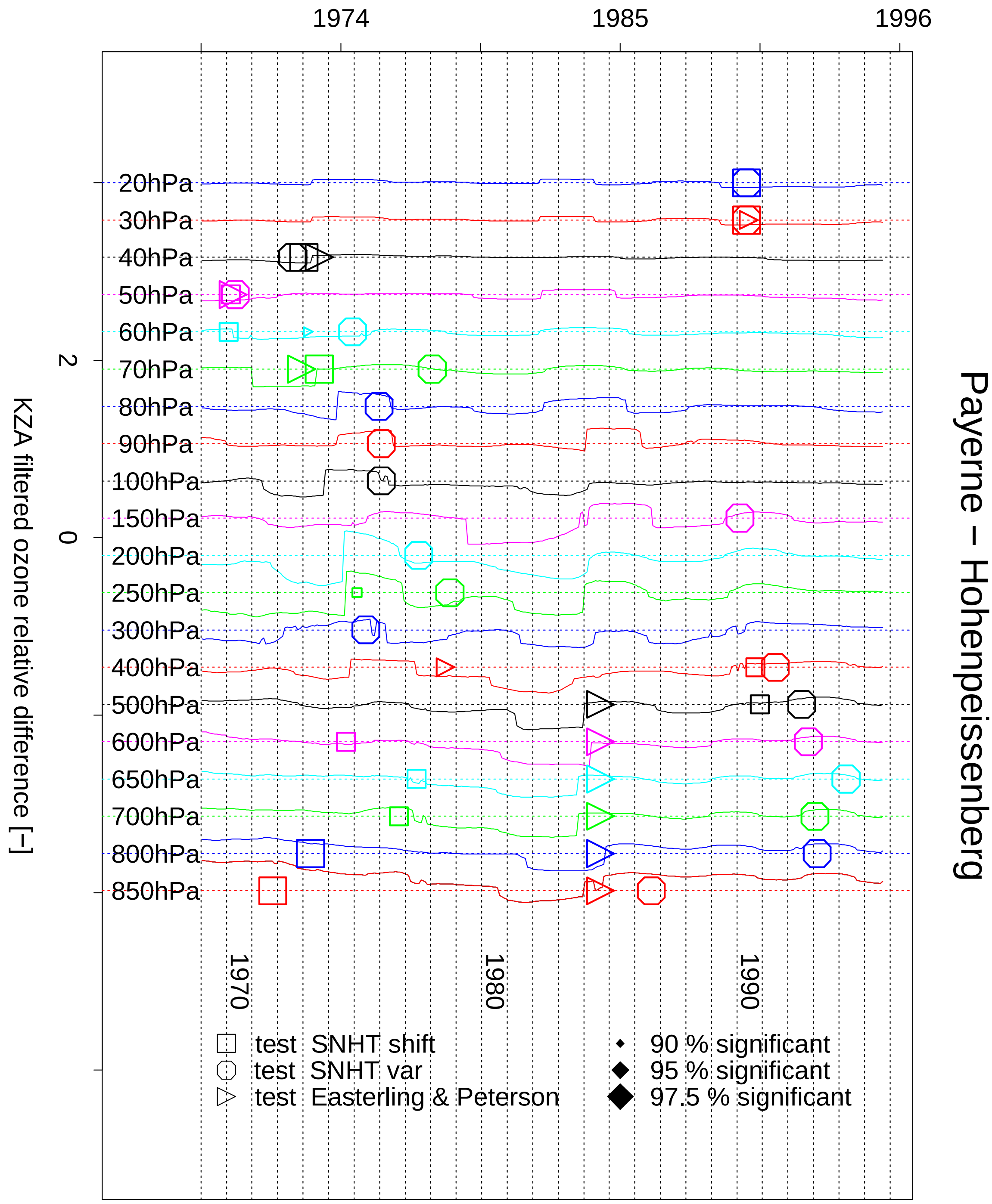

Figure 4.1: Monthly ozone relative differences between Payerne and Hohenpeissenberg at 20 pressure levels for soundings performed on the same days at the 3 stations of Payerne, Hohenpeissenberg and Uccle, filtered with the Kolmogorov-Zurbenko Adaptive method. For each pressure level, the horizontal dotted line crossing the label on the left corresponds to a zero difference between the two time series. Units on the vertical axis are the relative ozone differences and the space between 2 pressure levels corresponds to a relative difference of 40%. Breaks provided by the 3 homogeneity tests are represented by three different symbols, whose size depicts their statistical significance level.

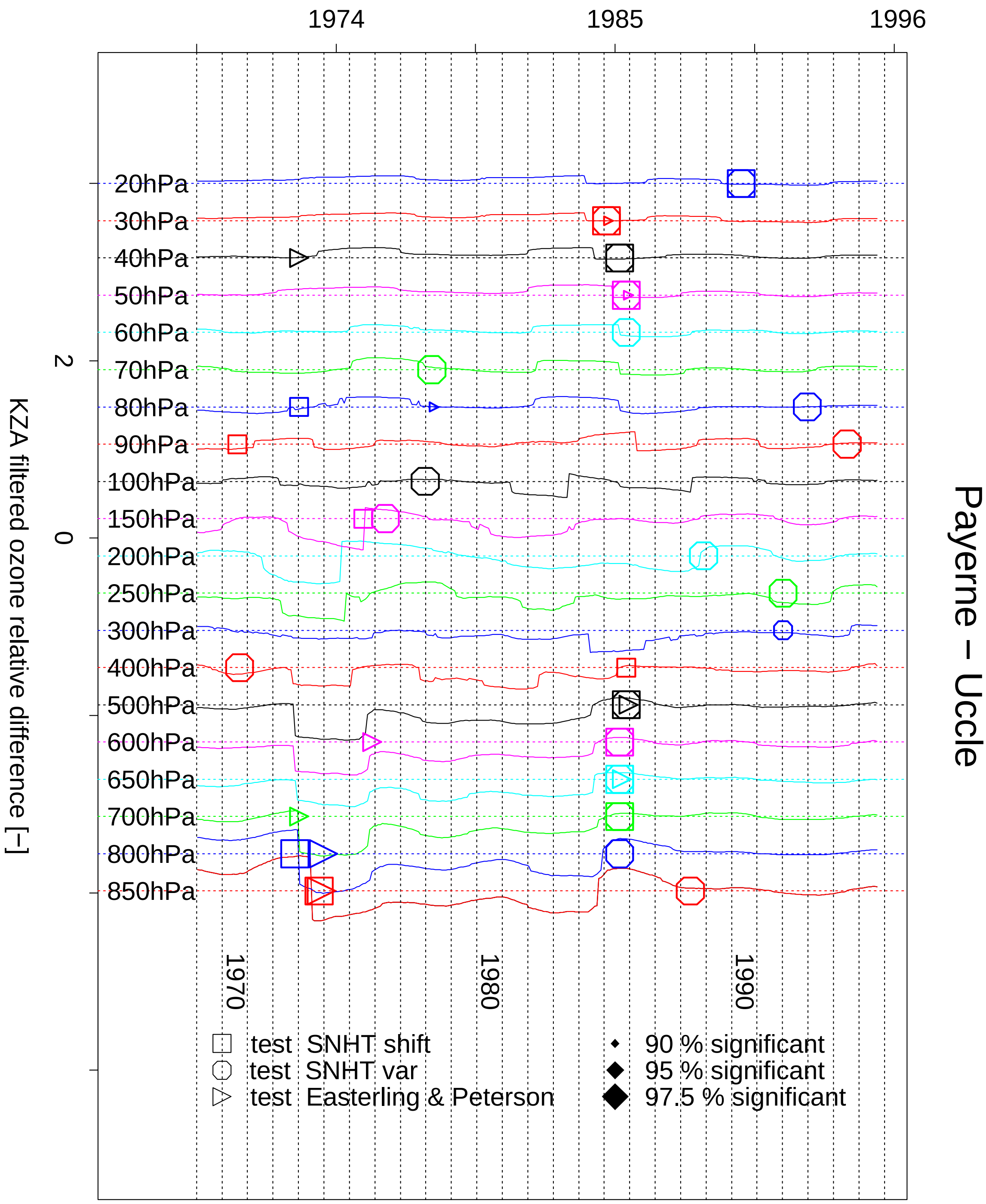

Figure 4.2: Same as Fig. 4.1, but for the differences between Payerne and Uccle.

Comparisons above the 20 hPa level have not been possible (too few cases). The lowest common level considered in Figures 4.1 and 4.2 is 850 hPa, because lower levels are too much influenced by local conditions.

The analysis of Figure 4.1 and 4.2 is first focused on their common features leading to suspect problems at Payerne. Then, an analysis is performed at the different levels as a function of time. Three specific periods with suspect breaks at several levels for the station of Payerne are considered in a third part.

Common features of Figures 4.1 and 4.2

Breaks detected either by the statistical tests and by the KZA filter are analysed in relation with their simultaneous occurrence on the same levels in both Figures 4.1 and 4.1. Breaks detected by the SNHT test are left apart in this first approach, in order to focus on systematic measurement uncertainties at the Payerne soundings. Results of different tests are still considered as simultaneous when they lay within a 1-2 year period, as these methods do not allow a precise time location. Results can be summarized as follows:

- Most tests agree with a high significant break (jump or trend anomaly) occurring in the low and middle troposphere between 1983 and 1985. The KZA filter suggests that this break might extend throughout the tropopause in the lower stratosphere, but without any statistical significance.
- The KZA filter detects a principal break near the tropopause (250 - 200 hPa) in 1974.
- In the middle stratosphere, most tests agree on two points: 1972-73 at the 40 hPa level and 1990 at the 20 hPa level.

Breaks have already been reported for these years in the analyses of previous versions of the Payerne dataset (Logan, 1994; Staehelin and Schmid, 1991; Hogrefe et al., 1998).

Time analysis on different levels

The KZA filter alone provides an interesting continuous information. In most cases, ozone deviations between pairs of stations are represented by a jump lasting for a few years and a return to zero. Nevertheless, slow drifts appear in the lowest levels and near the tropopause.

In the middle stratosphere, from the level of the highest ozone concentrations up to the last compared level (60 - 20 hPa):

- The relative ozone differences between Payerne and Hohenpeissenberg stay under 10% over the entire period.
- These differences are quite the same at the beginning and at the end of this period (1969 and 1995).
- Most deviations last for not more than a few years.
- The differences between Payerne and Uccle are somewhat larger than between the 2 nearest stations of Payerne and Hohenpeissenberg.

In the low stratosphere down to the tropopause, between 90 and 250 hPa, Payerne shows increasing deviations with the 2 other stations in the 2 periods 1973 - 1975 and 1983 - 1986. The largest breaks appear near the mean tropopause level (240 hPa), where the ozone concentrations are low and their natural variability high.

In the lower troposphere (500 - 850 hPa), where the regional meteorological conditions prevail and the different launch times increase the differences:

- The first half of the eighties is characterised by a noticeable deviation of Payerne lasting for a few years.
- Payerne measured more than 20% higher ozone values than the other stations at the lowest levels during the first years of the period.

First period: mid 1970's (1972-1977)

Several breaks are irregularly spread over different parts of the profile, but most of them are related to the variance and are found in only one of the comparisons. During this period only few soundings were performed simultaneously in Payerne, Hohenpeissenberg and Uccle; statistical tests are hence not conclusive. In fact, from 1972 to 1977, 23 monthly means out of 72 had to be interpolated in order to get continuous time series on simultaneous soundings. The deviations between Payerne and Uccle at the levels 800 - 500 hPa during the years 1973 - 1975 (see KZA curves) seem to be caused by the measurements at Uccle. The deviations between Payerne and Hohenpeissenberg at the levels 70 - 100 hPa during the years 1971 - 1975 do not seem to be caused by the Payerne soundings. Nevertheless, breaks at the levels 200 - 250 hPa appear on the KZA curves in both comparisons during this period, as well as at the 400 hPa level somewhat later and at 40 hPa in 1973, showing other discrepancies at the Payerne soundings.

Two launch time changes occurred at Payerne during this period: the first in 1972 (from 15-16:00 UTC to 14-15:00 UTC) and the second in 1977 (from 14-15:00 UTC to 08:30 UTC). Launch time changes are expected to impact in particular the measurements at the lowest tropospheric levels. Breaks appearing at and above the tropopause level are due to other events, such as the changes of the pre-flight procedures in 1976.

The comparison between temperature profiles of the VIZ sonde and of the operational CH sonde shows that the VIZ sonde systematically measured higher stratospheric temperatures compared to the operational CH sonde during 1975 and 1976. A not shown comparison of temperature, respectively geopotential height, at 30 and 100 hPa between these sondes indicates important incoherence in the VIZ measurement quality during these 2 years. Nevertheless this should not automatically infer errors in ozone measurements.

It is worth mentioning that Weiss (2000) found a break in 1975 between the ozone Umkehr (Arosa) and sounding (Payerne) profiles in the layer between 60 and 35 hPa.

Second period: mid 1980's (1983-1986)

Significant breaks have been detected by most statistical tests and by the KZA filter at some time within the years 1983 - 1985. They are shared over most levels of the troposphere as well as over the mid-stratospheric levels in the comparison Payerne-Uccle, but they are only confirmed in the low to middle troposphere in the comparison Payerne-Hohenpeissenberg. The KZA filter detects also some discontinuities in the low stratosphere in the latter comparison. However, these results are based on only a few simultaneous soundings; from 1983 to 1986, 26 monthly values have been interpolated (see 4.1.1).

Since January 1982, the launch time has been set to 11:00 UTC. Therefore discontinuities which appear after 1982 cannot be linked to changes in launch time at Payerne, but Uccle changed the launch time in March 1985 (before: 8-10 UTC, after: 10.30 UTC). Before and during this period the quality of the ozone soundings varies greatly; this is corroborated by the yearly statistics of the correction factor (Figure 2.4). The quality problems peaked from 1981 to 1983, where the correction factor showed high values. The quality were improved significantly end of 1983 due to changes in the preflight procedures. The correction factor slightly worsened again in 1985.

Here too, Weiss (2000) found a break in 1986 in the cumulative differences between ozone Umkehr and sounding profiles (60-35 hPa).

Third period: early 1990's (1990 -1993)

Breaks are detected mainly by the variance tests in the troposphere, but also by the other tests at the uppermost levels in the comparison Payerne-Hohenpeissenberg. In the Payerne-Uccle comparison, breaks appear only in the lower stratosphere and at 20 hPa.

As explained in chapter 3.1.2, the statistical correction applied to the Payerne dataset between 1990 and 1993 left some inhomogeneities in the time series. Furthermore, the important improvement of the pre-flight procedures introduced in 1993 had a noticeable impact on the measurement quality, as can be seen in the correction factor. There are further possible explanations of the breaks found in Fig. 4.1 and 4.2: Uccle changed the meteorological sonde in 1990 (De Backer, 1999), Payerne and Hohenpeissenberg reported a decrease in the quality of the delivered sondes at the end of the eighties, and problems occurred in Hohenpeissenberg at the end of 1993 (Claude et al. 1999). It should also be pointed out that the KZA curves generally show only slight breaks in 1990 and afterwards.

4.1.3 First conclusions

The interpretation of the previous results needs some caution. The dataset selected for the comparisons with statistical tests are far from comprehensive during several years of the 70's and the 80'. Nevertheless, some features can be highlighted for the Payerne ozone soundings:

- A break point is well identified between end of 1983 and 1984 for the low and middle troposphere, with possible extensions into the lowest stratosphere. It marks the end of a period lasting for a few years with noticeable differences with the other 2 European stations.
- Payerne depicts higher values than these 2 stations in the lowest troposphere (800 - 850 hPa) during the first years of the comparison.
- Between 1972 and 1977, the Payerne soundings show some divergences with the other ones, for example in the upper troposphere.
- The highest studied level (20 hPa) still exhibits a residual deviation left by the correction applied to the period 1990 - 1993.

Most of these discontinuities can be linked to operating changes mentioned in the station history, but cannot be quantitatively explained by well defined physico-chemical factors. It is important to note that even small changes in the preflight procedures of the sonde can have a strong influence on the ozone profiles measured by the Brewer-Mast sondes (de Muer, 1984). Due to the complexity of the behaviour of this sonde in the real atmosphere and to the impossibility to reproduce the past measuring conditions, it is hard to propose additional corrections for the periods with suspected deviations mentioned above.

4.2 Ozone and tropopause pressure

The previous section 4.1 has shown some anomalies in the comparison between the ozone soundings at Payerne, Hohenpeissenberg and Uccle and has pointed out on possible problems of technical origin in the Payerne dataset. The possibility that atmospheric processes are involved in these anomalies has now to be explored. The following two atmospheric variables are appropriate candidates for such an analysis:

- Total ozone measured at Arosa, which serves for the scaling of the Payerne ozone soundings and which is linked to the vertical ozone profile through atmospheric processes.
- Tropopause pressure measured by the meteorological sonde of the ozone sounding, which is a good indicator of dynamic processes.

Dynamical changes have been identified as possible explanations for variations in the ozone time series. Dynamical changes in the atmospheric circulation have contributed to the observed ozone trends at Northern mid-latitudes. Total ozone trend has been linked to long-term changes in various dynamical parameters such as, for example, tropopause height

(Steinbrecht et al., 1998). Comparison of ozone data coming from stations far away from one another has also been a matter for research (Weiss, 2000,). Large scale dynamical events (NAO, AO, QBO), but also regional meteorological phenomena that can be expressed by tropopause pressure, have a different influence on distant ozone sounding stations. To properly interpret discontinuities in tropospheric and in lower stratospheric ozone concentrations, one must also consider that natural long-term evolutions in the atmosphere (trends) are not completely linear, but may partly result from step like changes.

Over mid-latitudes, a strong variability in total ozone, and thus in ozone profile, occurs with the displacements of low and high pressure systems. Due to the dynamical constraints on the large-scale flow, surface low and high pressure systems are associated with distinct structures in the upper troposphere and in the lower stratosphere. This includes effects on the potential vorticity, on the potential temperature and thus on the tropopause pressure (or altitude). Low pressure systems are connected with an enhanced potential vorticity anomaly, a warm potential temperature anomaly and a tropopause pressure higher than normal (Hoskins et al. 1985). The correlation between total ozone and tropopause variability has already been well documented (Schubert and Monteanu, 1988; Steinbrecht et al., 1998; Vaughan and Price, 1991). Variability in tropopause pressure is a primary source of total ozone variability, it explains up to 40% of the ozone variability in the layer between 10 and 20 km (Weiss, 2000). Changes in the tropopause pressure reflect changes of the total air mass contained in the above vortex. Based on the dynamical constraints, a simple linear relationship can then be inferred between the variability of the tropopause pressure and the total mass of ozone in the lower stratosphere (Appenzeller et al., 2000). For this reason, anomalies in the ozone series due to meteorological processes would also be present in the tropopause pressure dataset.

In order to improve the interpretation of the breaks revealed by statistical tests, an analysis based on the tropopause pressure measured by the Payerne soundings as well as on total ozone measured at Arosa has been performed. The thermal tropopause has been used in the present study, which is defined as the lowest level at which the lapse rate decreases to 2 C / km or less provided that the lapse rate between this level and all higher levels within 2 km does not exceed 2 C/km. The tropopause pressure has been derived from each temperature profile of the ozone soundings. The value of total ozone measured by the Dobson instrument at Arosa has been extracted for each sounding. When missing, a substitute value according to the description in section 2.3 has been used. The corresponding monthly means are presented in Figure 4.3 (tropopause pressure) and in Figure 4.4 (total ozone). Both variables are linked together with a correlation coefficient of 0.63 (based on monthly means).

In Figure 4.3, the usual annual cycle of tropopause pressure is strongly modified between the end of 1974 and the begin of 1976. Consequently, the KZA filter reacts very strongly to this anomaly and finds a temporary jump of 20 hPa that corresponds to a deviation of the tropopause height of 500 m. A technical problem with the meteorological sonde has been identified during this period (see section 4.1.2), although the very small number of valid ozone soundings on that time limits also the representativity of the monthly means. Even if one of the secondary breaks depicted by the KZA filter occurs in 1990, the 2 changes of the meteorological sondes (1980 and 1990) do not have a noticeable impact on the tropopause time series. The KZA filter finds 2 other breaks, in 1985 and 1993, which occur in periods where breaks have been detected in chapter 4.1. The linear trend of the tropopause between 1970 and 1999 is given in Figure 4.3. Its value of -4.2 hPa/decade (or -1.8%/decade) corresponds to an increase of the tropopause height of approximately 110 m/decade. Steinbrecht et a. (1998) showed that the tropopause over Hohenpeissenberg has moved up by 150 m $\pm$ 70 m (2 standard deviations) per decade over the last 30 years.

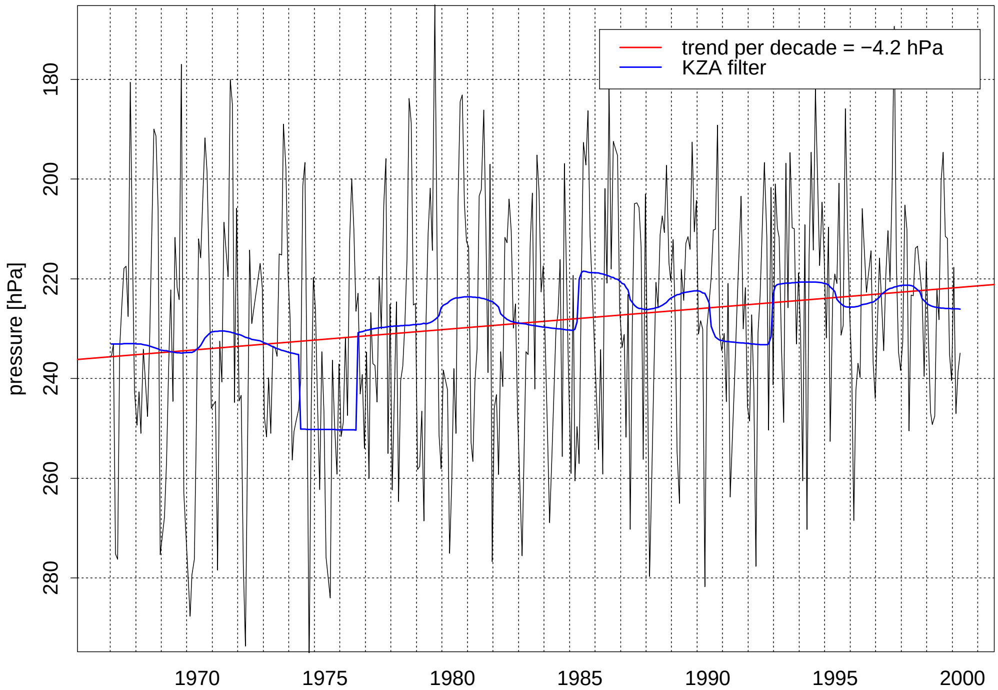

Figure 4.3: Tropopause pressure of the Payerne ozone soundings. Monthly means in black, linear trend in red and KZA filtered values in blue. The pressure scale is linearly decreasing from bottom to top, so that the tropopause height increased from 1967 to 2000. The trend of -4.2 hPa/decade corresponds to -1.8%/decade.

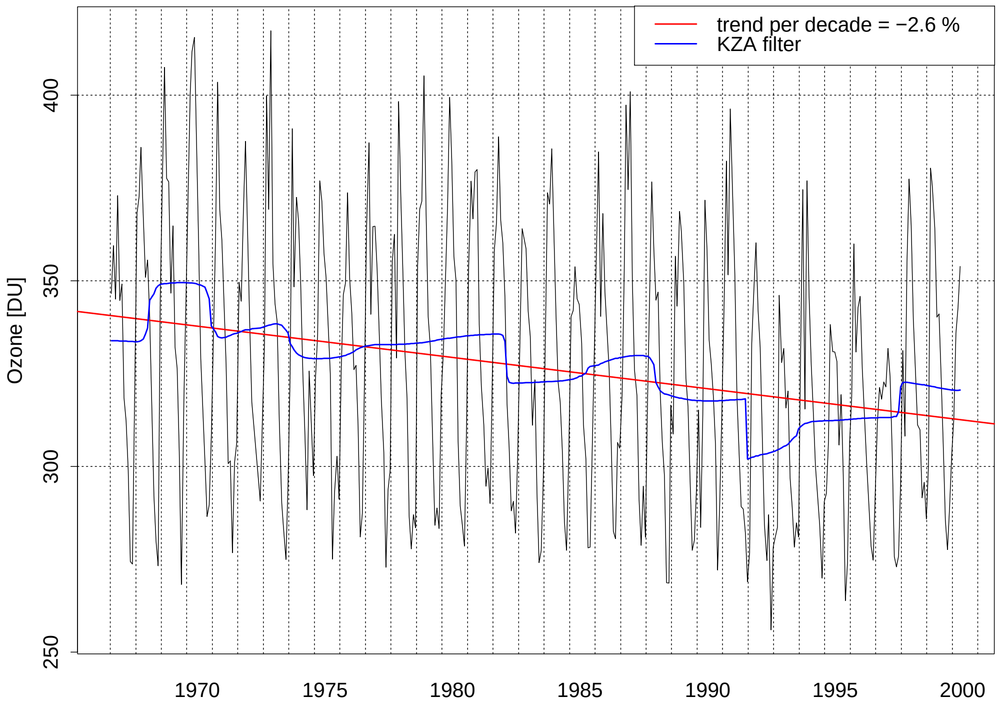

Figure 4.4: Total ozone (Arosa) for the days with ozone sounding at Payerne. Monthly means in black, linear trend in red and KZA filtered values in blue.

The last major volcanic eruptions clearly reduced total ozone for a while. This is not easy to see in the monthly means of Figure 4.4, but the KZA smoothed curve clearly shows a sudden decrease of total ozone after the El Chichon and Pinatubo events (1982 and 1991). The total ozone reduction after the latter amounts to 15 Dobson units.

The period with a possible tropopause anomaly according to Figure 4.3 exhibits only a slightly reduced annual cycle of total ozone, as well as a small deviation in the KZA curve. As only simultaneous values of tropopause pressure and total ozone have been retained in Figures 4.3 and 4.4, this could confirm that the tropopause calculation was then impaired by a measurement problem with the meteorological sonde. According to the KZA curves, none of the noticeable breaks of the tropopause and of total ozone are really simultaneous.

The trend of total ozone for the days with ozone sounding at Payerne given in Fig. 4.4 (-2.6% per decade for the period 1970 - 1999) is similar to the trend for all total ozone measurements at Arosa (-2.3% per decade for the period 1970 - 1996 (MeteoSwiss, 2000)). Hence, this subset is equivalent to the whole set.

In order to obtain more information on the link between total ozone and tropopause pressure, it is interesting to perform a trend analysis for these two parameters. Tropopause pressure and total ozone show similar trends: -1.8% and -2.6% per decade respectively. The trend of the ratio total ozone over tropopause pressure amounts accordingly to -0.8% per decade and is also significantly different form zero.

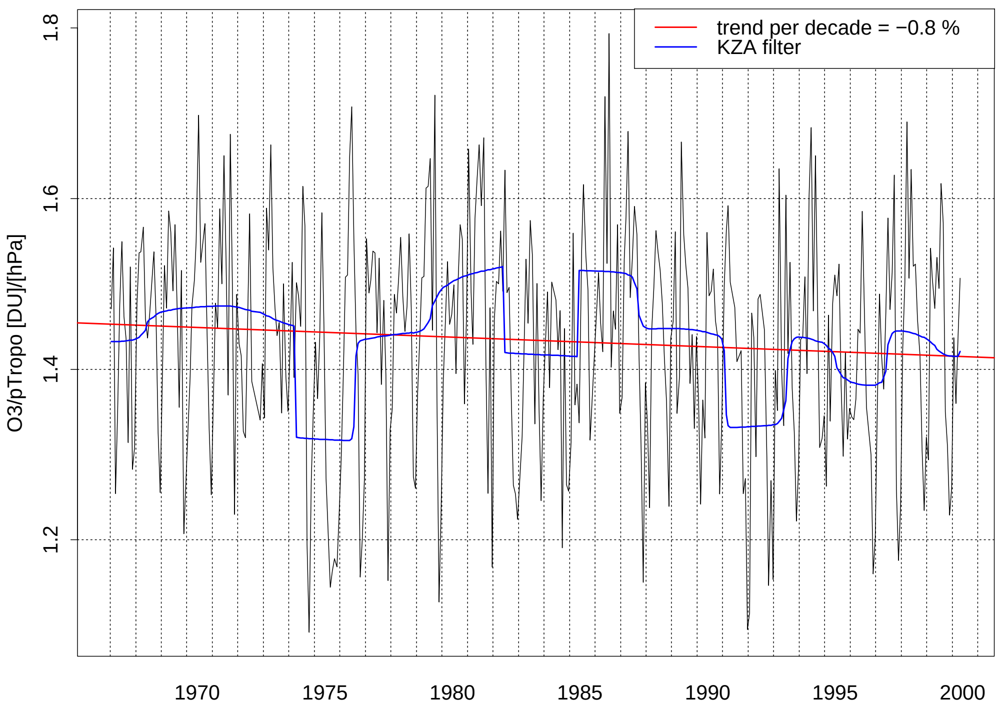

Figure 4.5: Ratio between total ozone and tropopause pressure (according to Fig. 4.3 and 4.4). Monthly means in black, linear trend in red and KZA filtered values in blue.

This ratio (total ozone over tropopause pressure) is presented in Figure 4.5. As suggested by the KZA filtered series that does not deviate very much from the trend line, variability in tropopause pressure is partly reflected in total ozone, according to the well proved correlation between these two parameters. Nevertheless, noticeable discontinuities occur in 74-76, 82-84 and in 91-93. This suggests the possibility that some atmospheric processes have played a role in the divergences found between Payerne and Hohenpeissenberg in the second and at the third period (see Figure 4.1 for comparison).

Another way to study the relationship between total ozone and tropopause pressure is provided by the direct correlation between both parameters, using the values of the single soundings. Hence, correlations have been computed for each year of the whole period 1967-1999 between tropopause pressure and total ozone in order to detect anomalous years. As tropopause pressure better correlates with ozone in the lower stratosphere than with total ozone (Weiss, 2000), similar correlations between tropopause pressure and ozone integrated from the ground up to 60, 50 and 40 hPa have been also computed, the variability in tropopause pressure is the primary source of total ozone variability between 10 and 20 km height (which correspond to 200 - 60 hPa), where it explains up to 40% of the ozone variability). Figure 4.6 shows the variance of total ozone explained by the tropopause pressure. It reaches its lowest value in 1976, but remains under 15% in the years 1973 to 1977 and 1996-1997, where 1997 is characterised by a strongly reduced annual cycle of total ozone (Fig. 4.4). It peaks near 50% in 1978, but remains generally under 40%. With one exception (1976), the explained variance for total ozone is lower than for ozone integrated up to the levels of 40, 50 and 60 hPa and the integrated ozone values up to the 60 hPa level correlate best with the tropopause pressure. This figure exhibits a rather large year to year variation of the relationship between tropopause pressure and atmospheric ozone, which seems to be largest until 1980. The few years 1983 - 1988 show a stable relationship between total ozone and tropopause pressure.

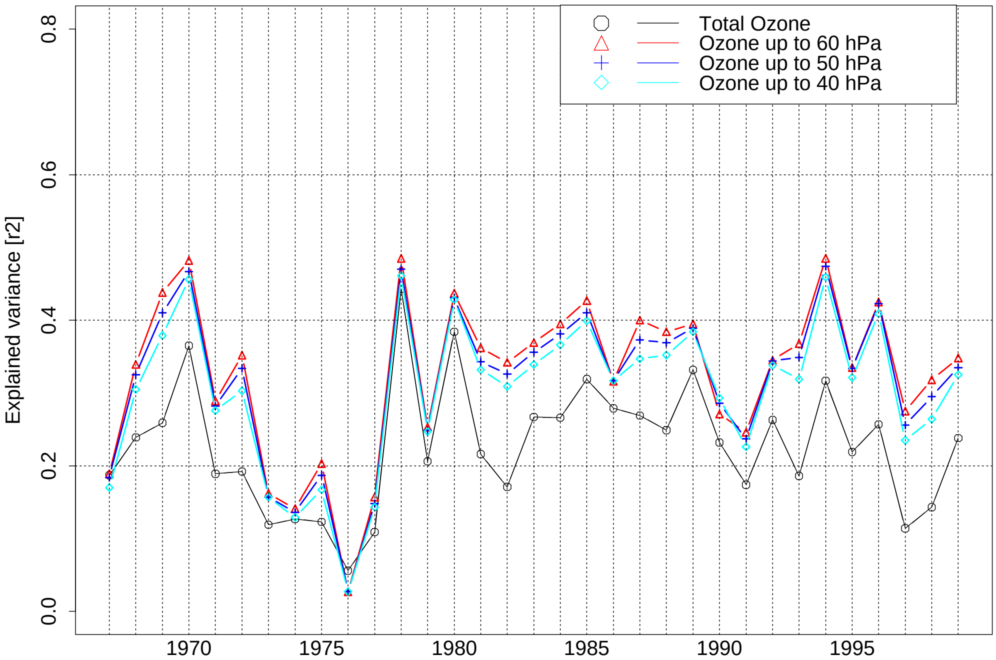

Figure 4.6: Ozone variance explained by tropopause pressure: total ozone in black, integrated ozone values from the ground up to 60, 50 and 40 hPa, in red, blue and green respectively

This comparative analysis of simultaneous values of total ozone measured at Arosa and tropopause pressure measured by the ozone sounding at Payerne can be concluded with the following results:

- Total ozone clearly showed a reduction after the last major volcanic eruptions of 1982 and 1991, but this does not explain deviations between Payerne and Hohenpeissenberg as these eruptions induce large scale processes.

- The tropopause pressure exhibits a deviation from its long-term trend during the years 1974-1976 that is probably due to problems with the meteorological sonde.
- Tropopause pressure and total ozone exhibit somewhat similar trends from 1967 to 1999.
- The variance of total ozone explained by tropopause pressure can vary much from year to year.
- The ratio between total ozone and tropopause pressure exhibits discontinuities during periods when the ozone soundings series, compared to Hohenpeissenberg and Uccle, also exhibits discontinuities.
- This short analysis does not allow results related to a possible atmospheric influence on the differences between the ozone profiles at Payerne and Hohenpeissenberg, because it does not take into account the meteorological differences between these two ozone sounding stations. But, such an influence can yet not be excluded.

4.3 Assessment of the homogeneity of the Payerne ozone dataset

The re-processing and re-evaluation of the ozone time series that have been performed in this project have reduced and partly explained the inhomogeneities found in the earlier versions of this dataset, but have not eliminated all of them.

Homogeneity analyses based on simultaneous data from Hohenpeissenberg and Uccle revealed three periods when the Payerne ozone balloon soundings showed on several pressure levels significant deviations with the two other ozone soundings stations. The three mentioned time periods can be associated to breaks in the time series of the correction factors (scaling to the total ozone measured at Arosa), which are themselves linked to operating changes or problems having occurred at Payerne. Additional analyses of total ozone measured at Arosa and of tropopause pressure measured by the Payerne soundings display some breaks during these periods, which can be related to atmospheric processes. This analysis does not take into account the meteorological differences between the three ozone sounding stations.

The following conclusions can be drawn regarding the homogeneity of the present Payerne ozone profile dataset, as well as its use for further analyses:

- The differences appearing as slowly decreasing drifts during the first measurement years in the lowest tropospheric levels between Payerne and Hohenpeissenberg may well be real. Statistical corrections would be inappropriate, but the measurement interferences by other trace gases have not been accounted for. The high measurement uncertainty of the Brewer-Mast sonde in the low troposphere should be kept in mind and trend results interpreted there with caution.
- The breaks found in the upper troposphere up to the tropopause region between 1972 and 1977 are difficult to interpret because only few soundings remained for the cross comparison. Furthermore, the meteorological parameters showed some inaccuracy between 1974 and 1976. As the statistical breaks are distributed over 5 years with different changes in the operating procedures, the latter ones may explain the breaks but the influence of each change is difficult to evaluate. An influence of the natural variability of the tropopause level can not be excluded. Additional corrections would lack a sufficient technical and statistical basis.
- The well identified break between the end of 1983 and 1984 in the low and middle

troposphere, with a less significant extension up to the lowest stratosphere, ends a period characterised by noticeable differences with the other two European stations. The improved preflight protocol introduced in 1983 after instrumental changes in the previous years (meteorological sonde and Brewer-Mast box) may explain the end of theses differences. But atmospheric particularities (ozone and tropopause level) should also be accounted for. This break - independently of its origin - strongly affects tropospheric trend calculations based on periods beginning or ending in the 80's.

- The Payerne dataset has been statistically corrected for 3 years in the beginning of the 90's due to problems after the change of the meteorological sonde and of the Brewer-Mast interfacing. The 20 hPa level still exhibits a significant residual anomaly during these years and other levels seem also - but less - affected. These years are also marked by a major volcanic eruption with noticeable impacts in the stratosphere and in the tropopause region.
- The major improvements in the preflight procedures of 1993 have significantly lowered the correction factor. Since then, a high quality standard has been ensured and occurring operating problems have been solved with top priority.
- The meteorological parameters measured by the 3 successive meteorological sondes used for the ozone soundings at Payerne have been re-evaluated. Their datasets are still not quite homogeneous. Temperature and geopotential trend calculations should be based not only on the ozone soundings, but also on the operational soundings performed twice a day.
- Any further re-evaluation and/or statistical homogenisation would certainly bring few new improvements in trend calculations over the whole period. Nevertheless, there are still ongoing studies in relation with the pump efficiency and the pump temperature influences, in order to prepare the change of the Brewer-Mast sonde by the ECC sonde in fall 2002. Moreover, a purely statistical homogenisation using the Hohenpeissenberg and Uccle datasets would introduce a statistical dependence to these time series. If trend analysis for the 80's must be improved, the period between 1981 and 1984 should be first re-evaluated and homogenised in more details.

The following Table 4.1 summarises the results of this re-evaluation and homogeneity analysis.

Table 4.1: Suspicious periods of the Payerne time series after the present re-evaluation.

Period	Parameter and altitude range
First years/decade(s)	Ozone: planetary boundary layer
1972 - 1977	Ozone: upper troposphere and tropopause region Meteorological parameters: tropopause and stratosphere between 1974 and 1976
1983 - 1984	Ozone: low and middle troposphere, less significant extension to the lower stratosphere
1990 - 1993	Ozone: residual anomalies in the upper stratospheric levels as well as in the troposphere

5 CLIMATOLOGY

A short climatic overview of the ozone profile and the tropopause measured by the ozone soundings at Payerne is given below. The period analysed starts at the beginning of 1967, including the measurements performed at Thalwil until August 1968. The physical and chemical processes explaining the key features of the ozone distribution in the atmosphere can be found in the review paper of Staehelin et al. (2000).

5.1 Ozone

The ozone profile climatology from 1967 to 2000 is presented in Figure 5.1. The ozone maximum is located in the stratosphere near the 50 hPa level. The concentration at that level varies during the year, reaching its highest value (about 170 nbar) in February and its lowest in late Summer (125 nbar). The ozone maximum is linked to the general circulation pattern in the stratosphere historically referred to as the Brewer-Dobson circulation. The high ozone concentrations just above the tropopause from February to June are explained by the mass transport from the stratosphere to the troposphere, whose maximum occurs in the spring season in the Northern Hemisphere, when the tropopause altitude is low. Conversely, the high tropopause altitude in Autumn is linked to low ozone concentrations just above it. The air exchange between the lower stratosphere and the troposphere occurs by complicated mechanisms such as tropopause folds.

In the troposphere, a weak ozone maximum related to photochemical production is found during summer months. The ozone minima are located in the upper troposphere from 500 to 300 hPa, where ozone concentrations drop to 15-20 nbar from early Autumn to late Winter. When considering this figure, it appears that the 40 - 50 nbar isolines better represent the limit between the ozone poor tropospheric airmass and the ozone rich stratospheric airmass than the tropopause itself.

The ozone climatology of the Belgian station of Uccle published by De Backer (1999) looks similar to that of Figure 5.1, qualitatively as well as quantitatively.

The same ozone statistics are displayed in Figure 5.2 for each of the three periods with a different meteorological sonde at Payerne. These periods roughly correspond to the last three decades. For each of these periods, the maximum is located at the same altitude, near the 50 hPa level. During the VIZ period (1967 - 1980), this ozone maximum reached 177 nbar, 172 nbar during the CH period (1981 -1990) and 156 nbar during the SRS period (1991 - 2000). In September and October, the 30 nbar ozone isoline crossed the tropopause during the VIZ and CH periods, but not during the SRS period where it stayed under the tropopause. In the upper troposphere, the surface within the isoline of 20 nbar shrank noticeably between the 3 decades.

As these graphics do not take into account the correction for the launch time changes before 1981, the significance of the comparisons for the lower troposphere between the first two decades is limited. Some ozone bias are also expected due to the interferences in the Brewer-Mast sonde with other trace gases (SO_2, NO_x), whose concentrations have changed in the planetary boundary layer over the last decades. The climatology of the last decade shows however an increase in ozone concentrations (over 30 nbar) in the winter low troposphere.

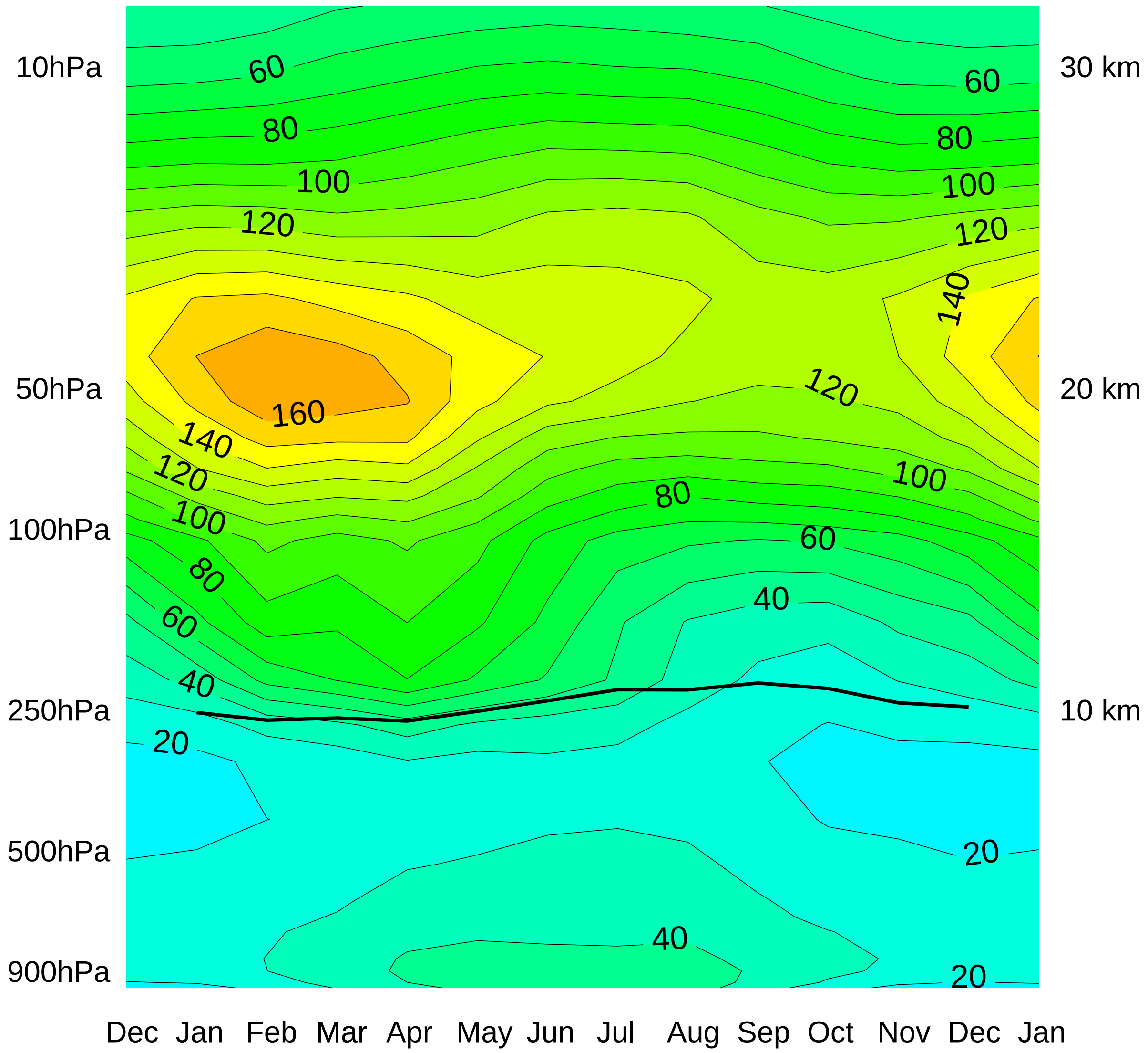

Figure 5.1: Ozone partial pressure (nbar) measured by the ozone soundings at Payerne from 1967 to 2000 as a function of pressure level and season. Isolines each 10 nbar, but labelled each 20 nbar. This figure is based on the monthly means for 25 pressure levels between 925 and 7 hPa. The absolute ozone concentrations are expressed in nanobars. The months of December and January are repeated at the left respectively the right border of the graphic. The climatological tropopause altitude is represented by the black line around 10 km.

Figure 5.3 shows the evolution of the seasonal ozone profiles of the Payerne ozone soundings. They differ significantly from one another and the features discussed on the base of Figure 5.1 appear clearly, such as the high/low ozone concentrations in the spring/autumn lowest stratosphere. The ozone increase over the three decades in the troposphere and the decrease in the stratosphere appear both. However, their significance can only be assessed with a trend model (see the following chapter).

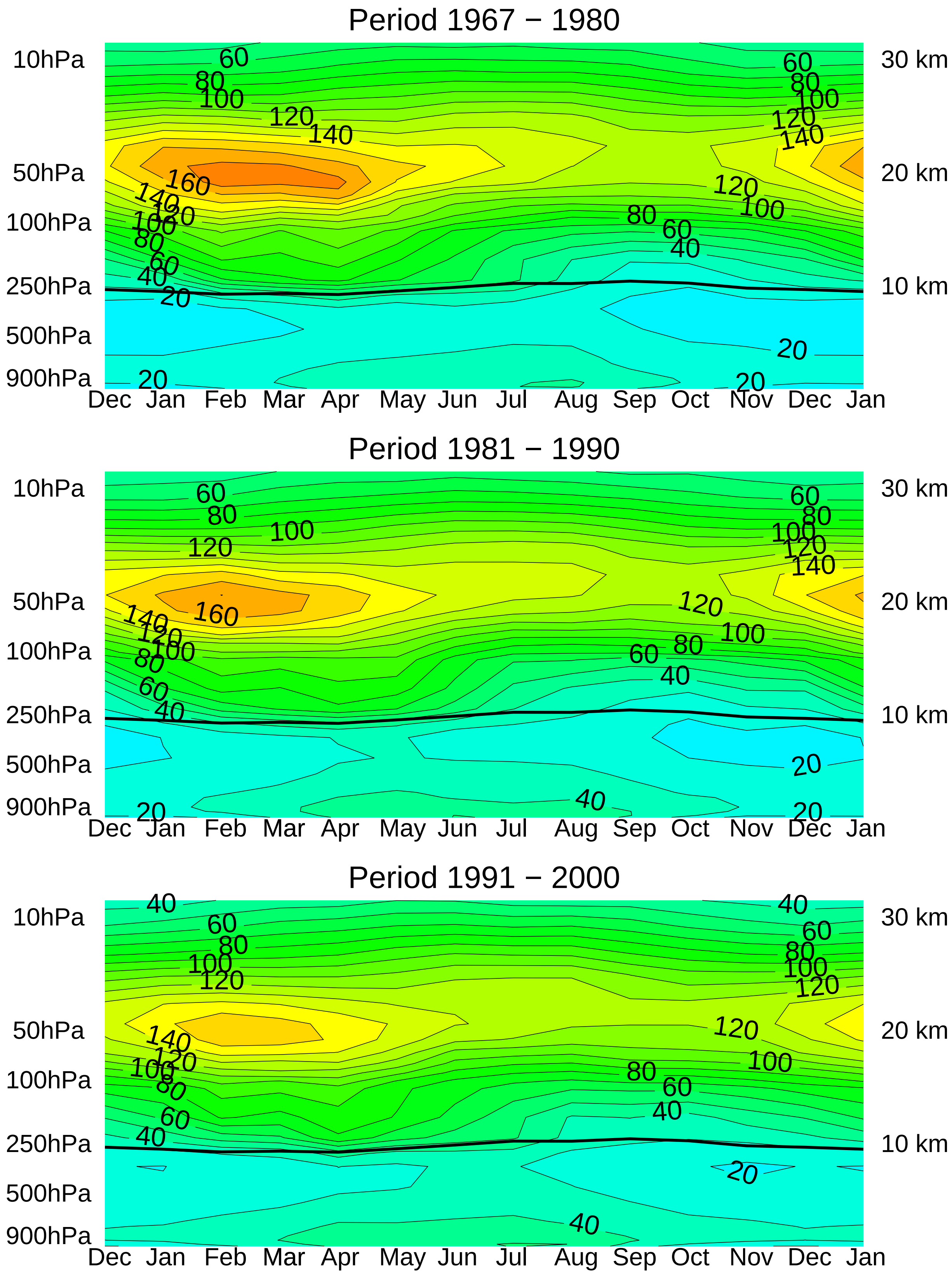

Figure 5.2: Ozone partial pressure (nbar) measured by the ozone soundings at Payerne as a function of pressure level and season for the three periods corresponding to different meteorological sondes: VIZ (1967 - 1980, without correction for launch time changes), CH (1981 - 1990) and SRS (1991 - 2000). Isolines each 10 nbar, but labelled each 20 nbar. See also legend of Figure 5.1.

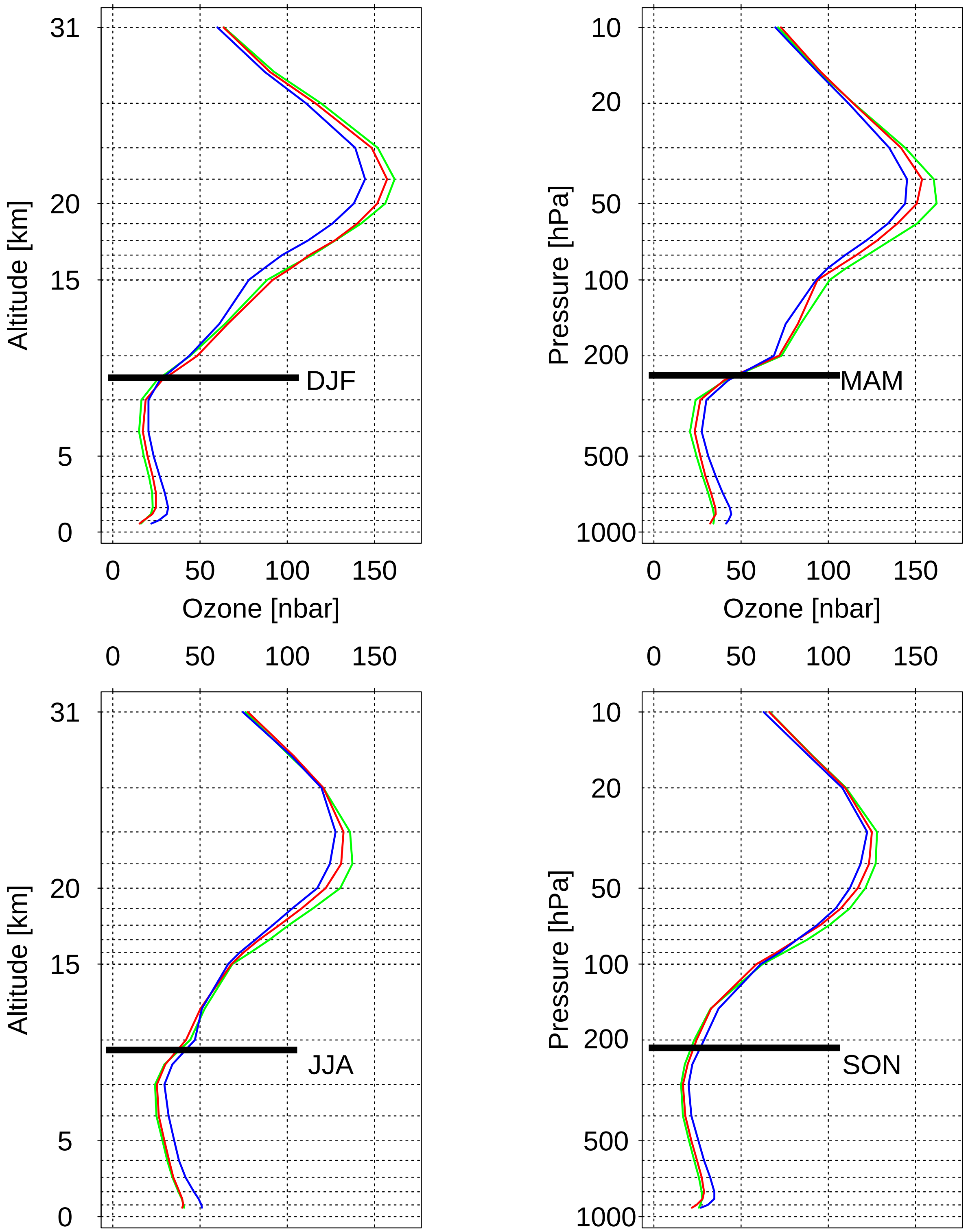

Figure 5.3: Seasonal vertical profiles of ozone partial pressure (nbar) measured by the ozone soundings at Payerne for the last three decades: green (1970 - 1980), red (1981 - 1990), blue (1991 - 2000). Top left: Winter (DJF: December to February), top right: Spring (MAM), bottom left: Summer (JJA), bottom right: Autumn (SON). The thick horizontal lines mark approx. the seasonal tropopause levels.

5.2 Tropopause pressure

In order to correctly interpret the ozone climatology and its trend, especially near the tropopause level, a short description of the tropopause climatology is given below, as well as a preliminary evaluation of its long-term evolution.

The tropopause marks an important vertical transport barrier. The air below this barrier is well mixed due to the almost adiabatic temperature decrease with altitude, while above this barrier, the almost isothermal temperature stratification strongly inhibits vertical mixing. As already stated, the thermal tropopause used in this report corresponds to the WMO definition (see chapter 4.2). Most of the time, the tropopause appears in the temperature profile as a marked temperature minimum, but sometimes the temperature still noticeably decreases in the lower stratosphere and the used definition is no more quite suitable. If a tropopause pressure greater than 500 hPa is found, it is set to 500 hPa.

The tropopause level undergoes a large day to day variation according to the weather situation. Its monthly means still show noticeable variations in addition to the annual cycle (see Figure 4.3).

As it has been done before for ozone, the tropopause climatology has been determined for three periods, according to the meteorological sondes used. The mean annual cycles of Figure 5.4 slightly differ for the 3 periods, but the largest monthly differences remain within 20 hPa (corresponding to a 500 m height difference). These differences can have both atmospheric and technical origins. The tropopause pressure is slightly sensitive to measurement errors (pressure and temperature) and height interval between successive measurements, which were not the same for the three different meteorological sondes. The three meteorological sondes all show the maximum of tropopause pressure (minimum of tropopause altitude) between March and April, when the ozone concentrations reach the highest values above the tropopause level (see Fig. 5.2).

In addition to the annual tropopause pressure trend presented in chapter 4.2 (-4.2 hPa/decade or -1.8%/decade), its long-term evolution has been computed for each month of the year (Figure 5.4). The most important negative tropopause pressure changes appear in late Winter. The March long-term evolution amounts to - 4% per decade, corresponding to a positive altitude change of 250 m per decade. This change is lower than the January value over Central Europe given by Schmitz et al. (2000), but these authors also showed the strong variation of this evolution between the latitudes of 40N and 50N. September and October show a slight - not significant - positive tropopause pressure evolution (Figure 5.4). High tropopause pressure values have been observed in particular during part of the VIZ period (1970 and 1974), that can be partly linked to very low total ozone. A possible autumnal positive trend is also derived by the NCEP re-analysis dataset, which delivers a positive insignificant trend for September (Weiss, 2000).

This brief analysis of the tropopause level shows that the measurements performed with the three different meteorological sondes used for the ozone soundings at Payerne deliver consistent climatological results. Although the trends of Figure 5.4 are statistically significant, they should be confirmed by an analysis of the dataset from the operational soundings performed twice a day, as well as by an enhanced instrumental error analysis. The annual trend calculated for Payerne is similar to the value published for Hohenpeissenberg (Steinbrecht et al., 1998).

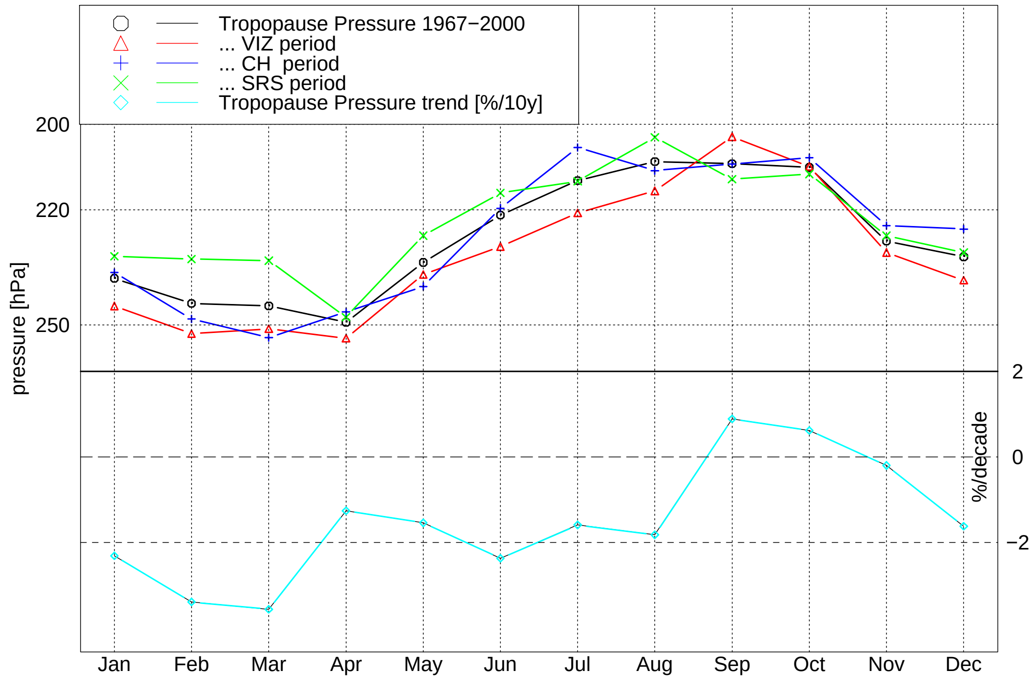

Figure 5.4: Annual cycle of tropopause pressure measured by the Payerne ozone soundings for different periods: black (whole period 1967-2000), red (VIZ period 1967-1981), blue (CH period 1981-1991) and green (SRS period 1991-2000). The pressure scale decreases from the bottom to the top of the graphic.
Below, the monthly tropopause trends are expressed in percent per decade between 1970 and 1999 (see Fig. 4.3 for the tropopause trend on a annual basis). According to this curve, the tropopause pressure has most decreased in March and has slightly increased in September and October.

6 OZONE TREND ANALYSIS

The ozone trend analysis presented here is organised in two steps. The two datasets introduced in chapter 2 are studied in a first step with a simple trend model involving only one trend parameter. The comparison of their trends gives an insight on the influence of the measurement quality and sample selection on trend calculations (chapter 6.1). In the second step, atmospheric explanatory variables are added to the trend model on order to get better estimates of the trend attributed to anthropogenic ozone depletion (chapter 6.2). Some special studies are included in chapter 6.3 and the trend results are summarised in chapter 6.4.

This trend analysis covers the whole period 1967 - 2000 with the - not fully justified, but commonly used - assumption of a linear trend. This report does not tackle the question of possible trend changes during sub-periods of the last three decades (e.g. an acceleration in trends in the 1980’s has been found in the northern mid-latitudes, Staehelin et al., 2001). The question of a possible trend slowering after mid 1990’s will be briefly discussed.

6.1 Observed ozone trends for two different datasets

6.1.1 Datasets and trend model

The datasets used belong to the Swiss ozone soundings performed between January 1967 and December 2000 (34 years), which have been processed as described in chapters 2 and 3, and include the correction for the launch time changes (see Fig. 3.2).

An appropriate balance between measurement quality and temporal representativity is necessary. To cover both aspects, two subsets have been defined and the correction factor has been used as a quality criterion (CF: scaling factor to total ozone measured at Arosa, see chapter 2.5). The comparison of the trends computed with these two datasets should provide useful information on the influence of the measurement quality and sample selection. However, despite several studies (e.g Logan et al., 1999), the choice between larger representativity but lower data quality or restricted representativity but better data quality is still a problem.

The first dataset includes all soundings characterised by a correction factor between 0.9 and 1.4; they are the ones submitted to the World Ozone and UV-radiation Data Centre (WOUDC). This results in at least 65 soundings per year, with one exception in 1972: only 38 soundings qualify for this criterion (Figure 2.4). Altogether this dataset amounts to almost 4000 soundings, and its mean correction factor is 1.18. Nevertheless, several months have no sounding, especially in 1972.

The second dataset satisfies to a more strict quality criterion with a CF between 0.95 and 1.25. The number of such soundings drops to 2800, but the resulting mean CF improves and decreases to 1.13 (Figure 2.5). Several years exhibit less than 50 soundings: 1968, 1970, 1971, 1972 (15 soundings), 1973, 1974, 1975 and 1981. More than hundred months have less than 4 soundings, most of them during the years 1967 - 1977 and 1980 - 1983. These months can be critical, since three to four soundings are considered to be necessary to get a representative monthly value.

Different analyses already used two similar datasets for studying the Payerne soundings (WMO, 1982, 1987; Staehelin et al., 1991; Miller et al., 1995; Bojkov and Fioletov, 1997; Logan, 1985; Logan et al., 1999, WMO, 1998; WMO, 1999). In the following sections, the first dataset is referred to as the normal dataset, the other as the reduced dataset. The model used to perform the trend analysis considers only a linear trend term:

oz' = trend + epsilon = c . ramp + epsilon

Where oz' is the deviation of the measured ozone concentration from the annual, seasonal or monthly mean over the 1967 - 2000 period. *trend* is composed of a variable *c* multiplying a linear *ramp* starting in January 1970 and rising by 1 per decade. This choice is justified by the almost linear increase of ozone depleting substances in the stratosphere from the beginning of the 1970's up to the middle of the 1990's. *epsilon* is the residual error term. The statistical function **lm** of the Splus software package is used (MathSoft,1993).

6.1.2 Annual trends

The two datasets, which noticeably differ in quality and representativity, are first analysed with the same model to determine if they deliver comparable trends. Annual mean values are used to remove the influence of seasonal variations on the trend estimate. Figures 6.1 and 6.2 present the results for the 25 selected pressure levels, first in nbar per decade, second in percent per decade.

The absolute trends in Figure 6.1 differ significantly from zero at the 95% confidence level for the two data samples on most levels of the profile, except in the tropopause region. As it is already well known, the trend is positive in the troposphere and negative in the stratosphere. The agreement between both datasets in the low and middle troposphere up to 400 hPa is within 0.5 nbar per decade, as well as at the level of the ozone maximum (50 hPa) and above 15 hPa. At the other levels, the trends differ within their uncertainty limits of 2σ; on a few levels, they lay outside (e.g on 200 and 250 hPa). In both cases, the maximum positive trends amounts to +4.5 ($\pm$ 1) nbar per decade ($\pm 2\sigma$). The maximum negative trends differ somewhat more. They range from -6 to -7 ($\pm$ 2) nbar per decade, but lay at two different altitudes. A noticeable difference appears between the two trend profiles, with a sign change at the ozone maximum. Both datasets are not equivalent for annual trend calculation in the lower stratosphere.

The relative trends in percent per decade are shown in Figure 6.2. They are more commonly used than the absolute trends. The reference used is the ozone value at the beginning of 1970. In addition to the interpretation of Figure 6.1, it is interesting to notice the almost constant positive trend in the troposphere between 925 and 400 hPa, due to the tropospheric air mixing convection, that amounts to 12-13%/decade with a slight maximum at 900 hPa. Above the tropopause, a first minimum occurs at 90 hPa for both datasets (4.5%, respectively 7% per decade, $\pm$ 2%); their vertical profiles do also not differ very much above the 100 hPa level. The trends in the stratosphere decrease then up to the 20 hPa level, before increasing again above. At the 250 hPa level, both dataset depict opposite trends that are not significantly different from zero.

The annual trends published in the SPARC/IO_3C/GAW assessment report (WMO, 1998) and later on by Logan et al. (1999) are superposed to the results on Figure 6.2. They are based on a partly re-evaluated dataset ending in 1996, submitted to the WOUDC in early 1998 and on more sophisticated trend models with atmospheric explanatory variables. The results of Logan and Megretskaia (LM) are similar to the present results based on the normal dataset. The results of the other group (Tiao) show rather large differences, which are due to an other very strict data screening and to the use of not normalised ozone profiles. The present results based on the two new datasets match much better together than the results of the two WMO groups, excepting for the levels between 300 and 70 hPa.

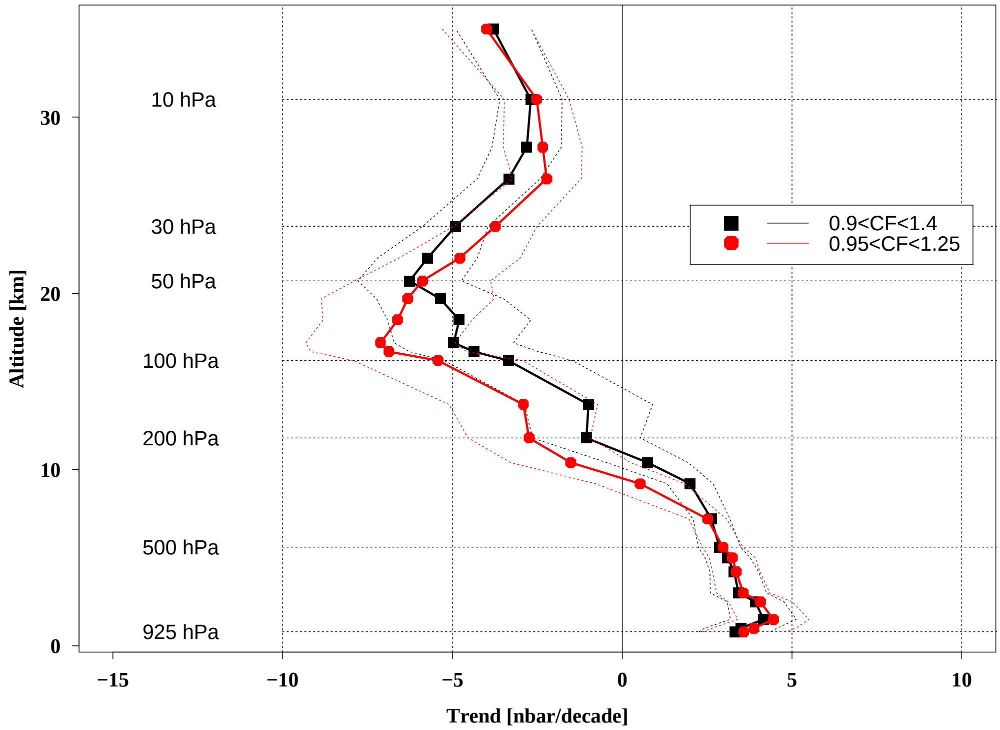

Figure 6.1: Annual ozone trends of the Payerne ozone soundings over the period 1970-2000 in nbar per decade. In black for the normal dataset (0.9 < CF < 1.4), in red for the reduced dataset (0.95 < CF < 1.25) The dotted curves reproduce their uncertainties (two standard errors).

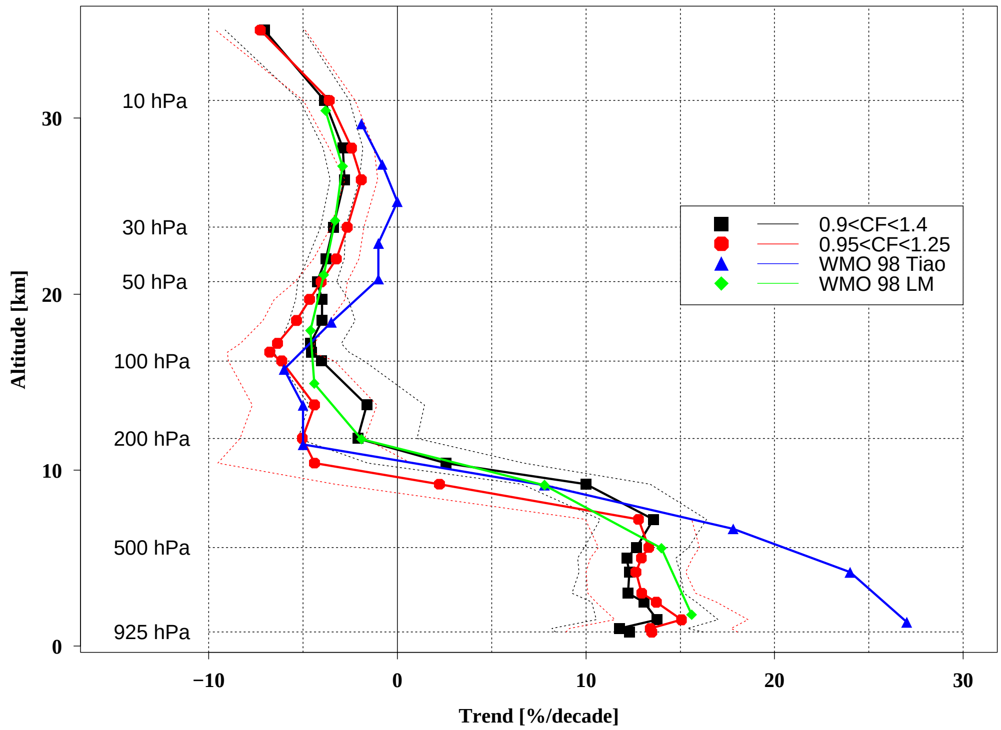

Figure 6.2: Annual ozone trends of the Payerne ozone soundings over the period 1970-2000 in percent per decade. In black for the normal dataset (0.9 < CF < 1.4), in red for the reduced dataset (0.95 < CF < 1.25) The dotted curves reproduce their uncertainties (2σ). The SPARC/IO$_3$C/GAW trend results (WMO, 1998) for the period 1970-1996 are included without their error estimation (slightly larger than the present one): results of the Chicago University (Tiao et al.) in blue, results of the Harvard University (LM: Logan and Megretskaia) in green, both related to Payerne.

The annual trends derived from the two datasets of the present report are significant and fairly consistent outside the atmospheric layer between 300 and 100 hPa. However, they can not be considered as equivalent for trend calculation. Our results show a fair agreement with the results of the Harvard University, described in the SPARC/IO$_3$C/GAW assessment report. Attention should be paid to the fact that different time periods are used.

6.1.3 Seasonal trends

Figures 6.3 and 6.4 provide the seasonal trends that have been computed quite similarly to the annual trends. The meteorological definition of the seasons is applied: Winter begins with December.

In the troposphere, positive trends ranging from 10% to 20% are observed. The largest (relative) positive trends are observed in Winter in the lowest levels (with 30% per decade for the normal data, 25% per decade for the reduced data). The smallest positive trend in the whole troposphere is observed in Summer, when the highest ozone concentrations are due to high photochemical pollution. Several processes could explain theses features:

- Trace gases such as SO_2 and NO_2 interfere with ozone measurements by the Kalium Iodine method (Schenkel and Broder, 1982). NO_2 leads to a slight overestimation of the ozone measurements. SO_2 causes ozone registration to be too low or even zero, because the decrease in the ozone registration is approximately equal to the SO_2 concentration. On the other hand, the SO_2 emissions peaked in Switzerland between 1965 and 1980 and decreased then very sharply between 1980 and 1990 (OFEFP, 1995). The SO_2 concentrations followed the same evolution. The SO_2 concentration reached 50 ppbv during Winter smog episodes at Payerne. The SO_2 annual mean at Payerne decreased from 5 to 0.5 ppbv between 1980 and 2000 (OFEFP, 2001). Consequently, this SO_2 decrease could be responsible for a noticeable artificial positive ozone trend in the planetary boundary layer, especially in the Winter up to 850 hPa (1500 m asl). This layer depth corresponds well to the winter ozone trends over 20% per decade. According to Schenkel and Broder (1982), it is also very likely that the BM sonde yields bad results in the whole troposphere, when it is launched during winter smog episodes. Consequently, it is important to note that the ozone measurement quality is questionable in the boundary layer, especially in the Winter.
- The half-life of tropospheric ozone with respect to chemical destruction is about 12 days in Summer, but is much longer in the Winter. These effects compensate the much slower ozone production rate in Winter, which is only about 10% of the rate in Summer. In addition it is believed that emission of the tropospheric ozone precursors is more strongly dominated by anthropogenic sources in Winter than in Summer (Liu et al. 1987). Staehelin et al. (1990) suggests that the concentration of tropospheric ozone is more heavily influenced by local and regional ozone formation in Summer than in Winter. Because of the much longer half-life of ozone in Winter than in Summer, the influence of air pollution encompasses a larger area during the cold season. This can also partially explain why the strongest relative ozone trend over Payerne occurs in Winter and not in Summer.
- Due to the lower amount of ozone titration, the decrease of NO emissions might be partially responsible for the increase in tropospheric ozone in Winter in the lowest part of the troposphere in the 1990.
- Stratospheric intrusions can also influence trends in the troposphere, mostly in the cold season.
- The variability of UV-B radiation during late Winter to Spring relative to the interrannual variability of monthly or annual mean can also possibly become non negligible (Brönnimann et al., 2000).

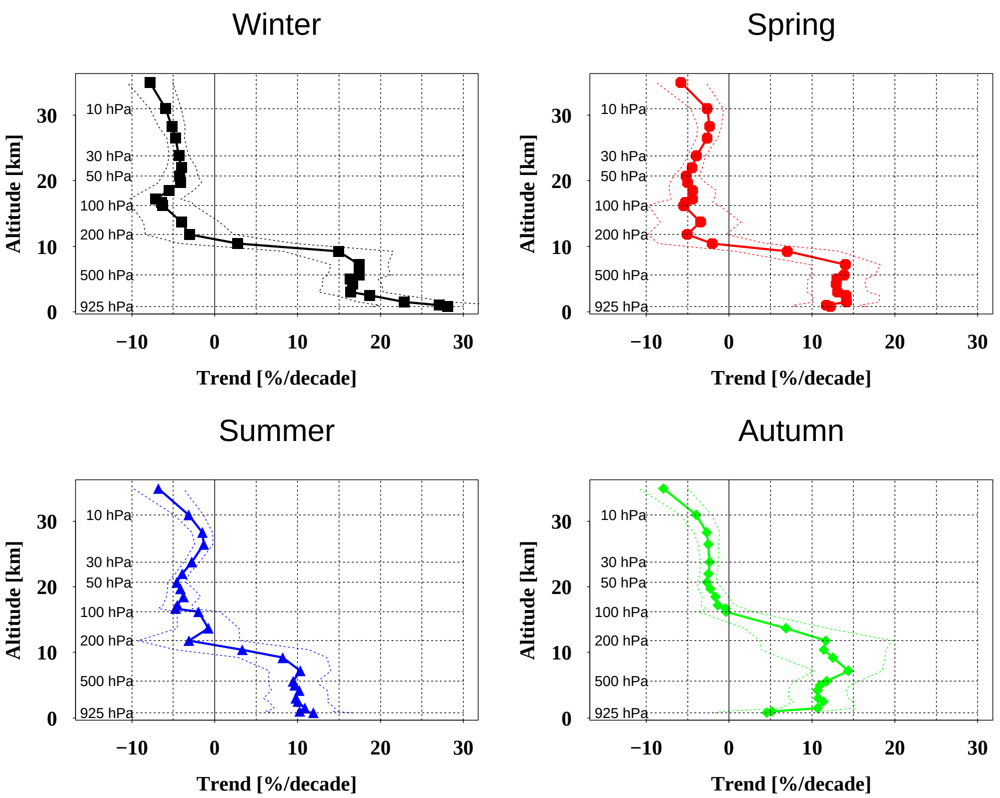

Figure 6.3: Seasonal ozone trends of the Payerne ozone soundings over the period 1970-2000 in percent per decade for the normal dataset (0.9 < CF < 1.4). The dotted curves reproduce their uncertainties (2σ).

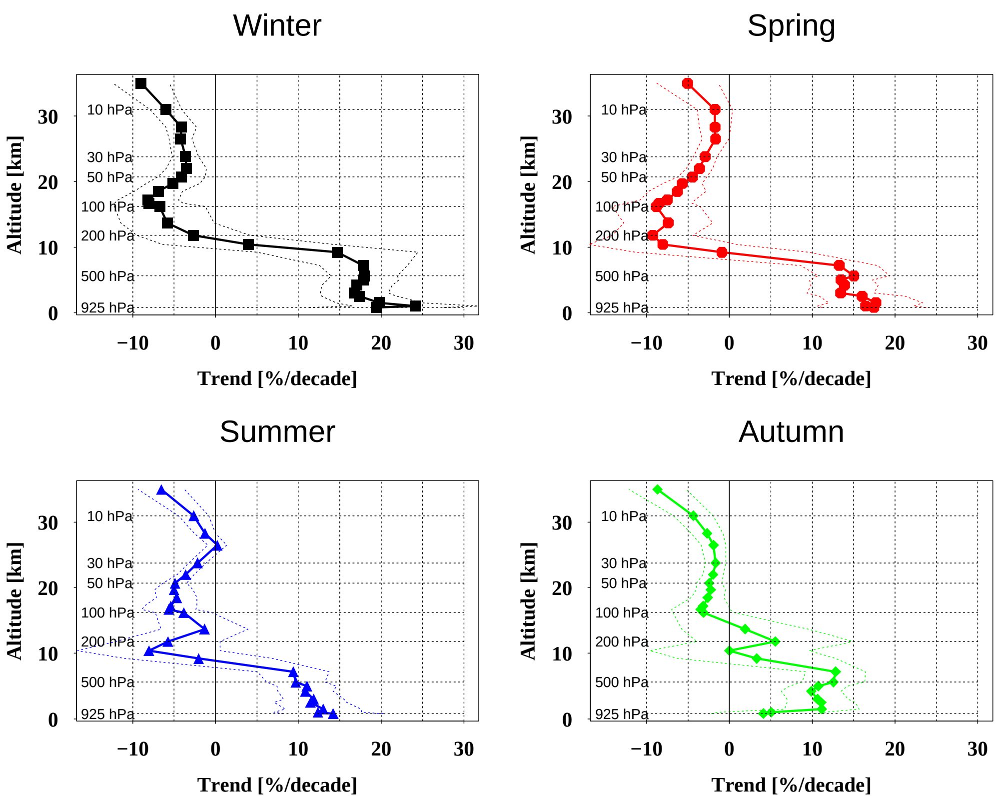

Figure 6.4: Same as Figure 6.3, but for the reduced dataset (0.95 < CF < 1.25).

- Last but not least, the choice of the trend unit, which is very sensitive to the concentration level (low in Winter and high in Summer, in the troposphere). In the case of large trends, the intercept at 1970 is quite smaller than the mean over the 34 years and amplifies the relative trend values. Trends expressed in absolute values [nbar/decade] show a less pronounced annual cycle.
- However, all the phenomena presented above do not explain completely the large positive tropospheric ozone winter trend.

Positive trend values are found up to 150 hPa in the autumn soundings for the normal dataset (Figure 6.3). They can be partly explained by the slight tropopause height decrease during this season (see chapter 5.2). During the other seasons, the positive ozone trends remain below the 200 hPa level (Figure 6.3: normal dataset), period during which the tropopause height increased significantly (see Figure 5.4). As explained in section 4.2, changes in the tropopause height can generate changes in the total air mass contained in the air column above. The tropopause height variation can thus modify the ozone profile near the tropopause. When in particular the tropopause height increases, the low ozone concentrations just above the tropopause can move up, respectively be modified through complex circulation patterns. As ozone concentration increases with altitude above the tropopause, ozone can undergo negative trends near the tropopause when the altitude of the latter increases. The opposite phenomenon is observed in September and October when the tropopause altitude decreases, which means that the higher concentrations found above the tropopause can slightly move down to a lower elevation, too. This could explain the positive ozone trend observed at the tropopause level (Figure 6.3, panel for Autumn).

In the stratosphere, the largest negative ozone trends are observed in Winter with -7.8% per decade around 80 hPa. At 10 hPa and above, large negative trends are also found. In Autumn, the negative trend increases regularly with altitude for both datasets. Nevertheless it is important to note that the measurements quality of the Brewer-Mast degrades above 15 hPa and that the standard errors of the trends increase above this level. According to the WMO recommendations (1998), the sonde data are expected to be reliable for trend determination up to 27 km (15 hPa).

The comparison between the trends of both datasets (Fig. 6.3 and 6.4) concludes to similar features; except for major deviations in the tropopause region between Spring and Autumn. However, the reduced dataset shows more anomalies of the tropopause altitude then the normal dataset (summer cases with doubtful low tropopause altitude). The standard deviation of the trend is furthermore larger for the reduced dataset at the 250 and 300 hPa levels, where the trends strongly differ between the two datasets.

The variances explained by the model are shown in Figure 6.5. The model used explains between 20 and 70% of the ozone variability in a large part of the troposphere and the stratosphere. In a broad region around the tropopause, the model provides however no explanation for the ozone variability in any dataset.

Concluding this seasonal trend analysis, it appears that trends calculated with the normal dataset are more consistent in the upper troposphere and lowest stratosphere than trends computed using the reduced dataset. In the lower and middle troposphere, as well as in the middle stratosphere, both datasets provide rather similar trends and it is not possible to discriminate them. The results show that the main features of long-term trends are stable with respect to the data quality selection. Some differences in the profile shape although remain. Thee results also show some evidence for a better internal consistency if the representativity is given higher weight than the data quality, in particular where the natural variability is large.

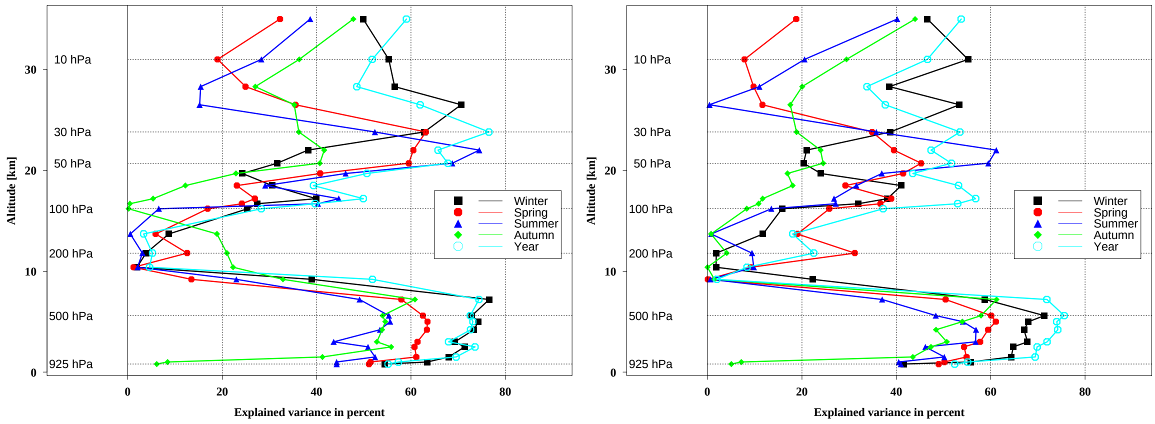

Figure 6.5: Explained variance (R^2) of the observed ozone (simple trend model) for the four seasons as well as for the year, left the normal dataset (0.9 < CF < 1.4), right for the higher quality dataset (0.95 < CF < 1.25).

The key results of this observed trend analysis are summarised in the following table.

Table 6.1: Annual and seasonal values of the observed trends at 25 pressure levels (P [hPa]). T: Trend [% per decade], 2s: statistical uncertainty of the trend (two standard deviations) R2: Explained variance [%])

P	Year			Winter			Spring			Summer			Autumn		
	T	2s	R2	T	2s	R2	T	2s	R2	T	2s	R2	T	2s	R2
925	12.3	3.9	55	28.1	9.1	54	12.2	4.2	51	11.9	4.7	44	12.3	6.3	6
900	11.8	3.6	57	27.1	7.3	63	11.7	4.0	51	10.3	4.1	44	11.8	6.0	8
850	13.8	3.2	70	22.9	5.5	68	14.1	4.0	61	10.9	3.7	52	13.8	4.5	41
800	13.1	2.8	74	18.7	4.2	71	14.1	4.0	61	10.1	3.5	51	13.1	3.6	56
700	12.2	3.0	68	16.4	3.9	69	13.1	3.7	61	9.8	3.9	44	12.2	3.6	53
650	12.3	2.7	73	16.7	3.6	73	12.9	3.5	63	10.2	3.4	53	12.3	3.5	54
600	12.2	2.6	73	16.3	3.4	74	13.0	3.5	64	9.7	3.1	56	12.2	3.5	55
500	12.7	2.8	73	17.4	3.8	73	13.8	3.8	63	9.5	3.0	55	12.7	3.8	54
400	13.6	2.8	74	17.5	3.4	77	14.0	4.2	58	10.3	3.7	49	13.6	4.1	61
300	10.0	3.4	52	14.9	6.6	39	7.0	6.3	13	8.2	5.3	23	10.0	6.3	33
250	2.6	4.1	5	2.8	7.0	2	-2.0	6.5	1	3.3	8.1	2	2.6	7.5	22
200	-2.1	3.2	5	-3.0	5.4	4	-5.1	4.7	13	-3.1	6.1	3	-2.1	8.0	21
150	-1.6	3.1	3	-4.0	4.6	9	-3.5	4.9	6	-0.8	3.7	1	-1.6	5.0	19
100	-4.0	2.3	28	-6.3	3.8	25	-5.5	4.3	17	-2.0	2.6	7	-4.0	3.3	0
90	-4.5	2.0	40	-6.4	3.7	27	-5.3	3.3	24	-4.7	2.0	40	-4.5	2.3	0
80	-4.6	1.6	50	-7.2	3.1	40	-4.4	2.6	27	-4.5	1.8	45	-4.6	2.0	5
70	-4.0	1.7	39	-5.5	2.9	31	-4.4	2.8	23	-3.8	2.1	29	-4.0	1.6	12
60	-4.0	1.4	51	-4.2	2.6	24	-5.0	2.1	41	-4.1	1.6	46	-4.0	1.5	23
50	-4.2	1.0	68	-4.2	2.2	32	-5.2	1.5	60	-4.6	1.1	69	-4.2	1.1	41
40	-3.8	1.0	66	-4.0	1.8	38	-4.5	1.3	61	-3.9	0.8	74	-3.8	1.0	42
30	-3.4	0.7	77	-4.3	1.2	63	-3.9	1.1	63	-2.8	0.9	52	-3.4	1.1	36
20	-2.8	0.8	62	-4.7	1.1	71	-2.6	1.3	36	-1.3	1.1	15	-2.8	1.2	35
15	-2.9	1.0	49	-5.2	1.6	57	-2.4	1.4	25	-1.5	1.2	15	-2.9	1.6	27
10	-3.9	1.3	52	-5.9	1.9	55	-2.7	1.9	19	-3.1	1.8	28	-3.9	1.9	36
7	-7.1	2.1	59	-7.8	2.8	50	-5.8	3.0	32	-6.8	3.0	39	-7.1	2.9	48

6.1.4 Monthly trends

Monthly trends for the normal dataset are presented in Figure 6.6. Some features are easier to see than in the previous analysis.

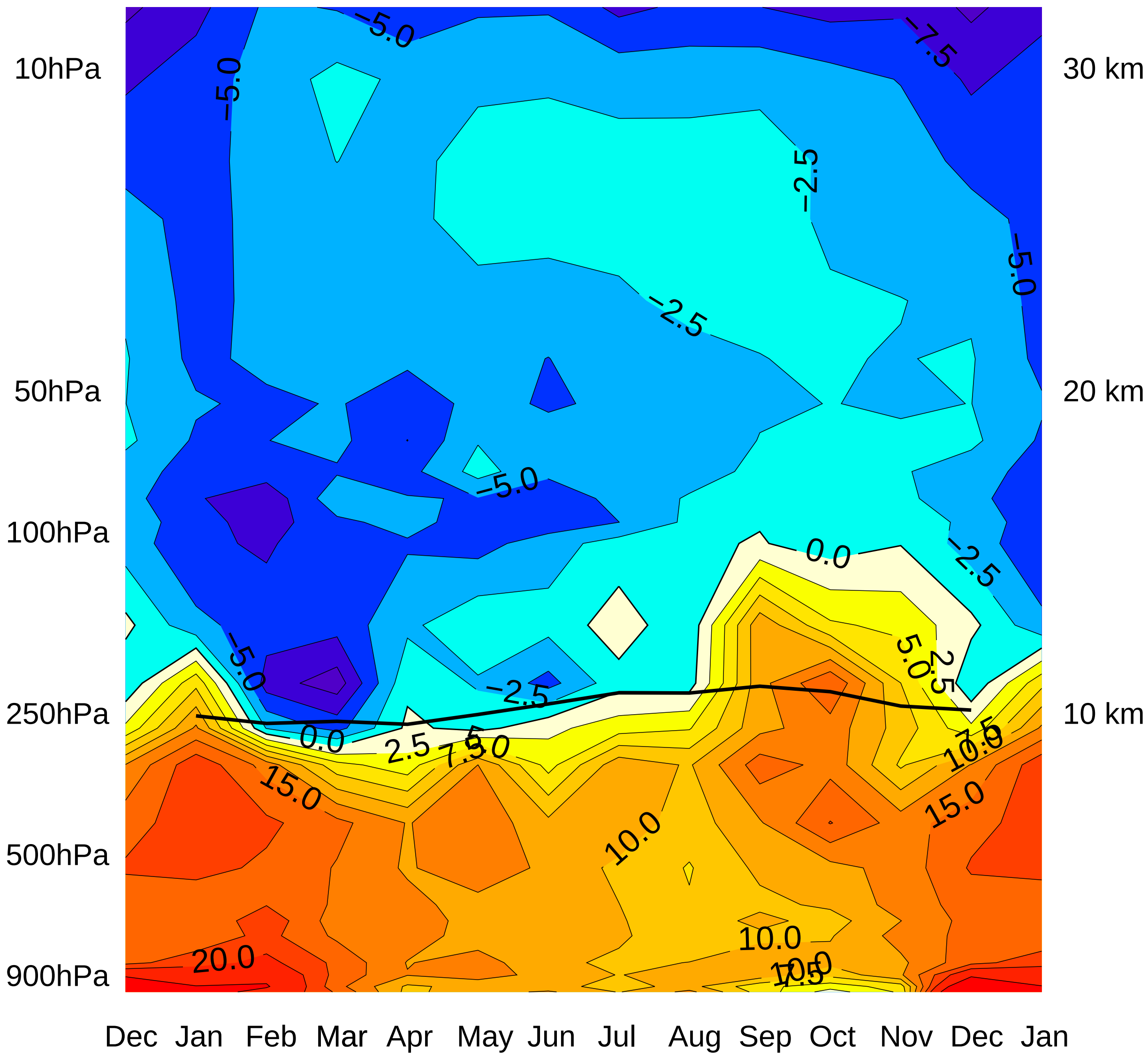

Figure 6.6: Isoplethes of observed ozone monthly trends between 1970 and 2000, calculated for 25 pressure levels between 7 and 925 hPa of the normal dataset (0.9 < CF < 1.4) and expressed in % per decade. The positive values are in warm colours, while the negative values are in cold colours. The climatological tropopause altitude is represented by the black line around 10 km.

Between late Winter and early Spring, the switch from a negative to a positive trend is very sharp and located at the tropopause. But two periods do not exhibit this feature: January and September to October. Comparing this figure to the climatology of the ozone soundings (Figure 5.1), it can be seen that the boundary between the positive and the negative trend can be identified with the 40-50 hPa ozone isolines. The analogy between the annual cycle of the tropopause pressure trend (Figure 5.4) and the similar cycle of the zero ozone trend line in Figure 6.4 can also be highlighted.

The most negative trend is found in March at the 200 hPa pressure level (-11% per decade, with the two standard deviation bound at ± 6% per decade); between 100 and 80 hPa negative trends are almost as important. The maximal positive trend is found in December at the 925 hPa level with 40% per decade. Nevertheless, it must be taken into account that trend results can be affected by large measurement errors in the boundary layer. At 850 hPa, this trend already decreases to 23% per decade. The figure confirms also the large positive trends in the whole troposphere during Autumn and Winter, that even extends to the low stratosphere.

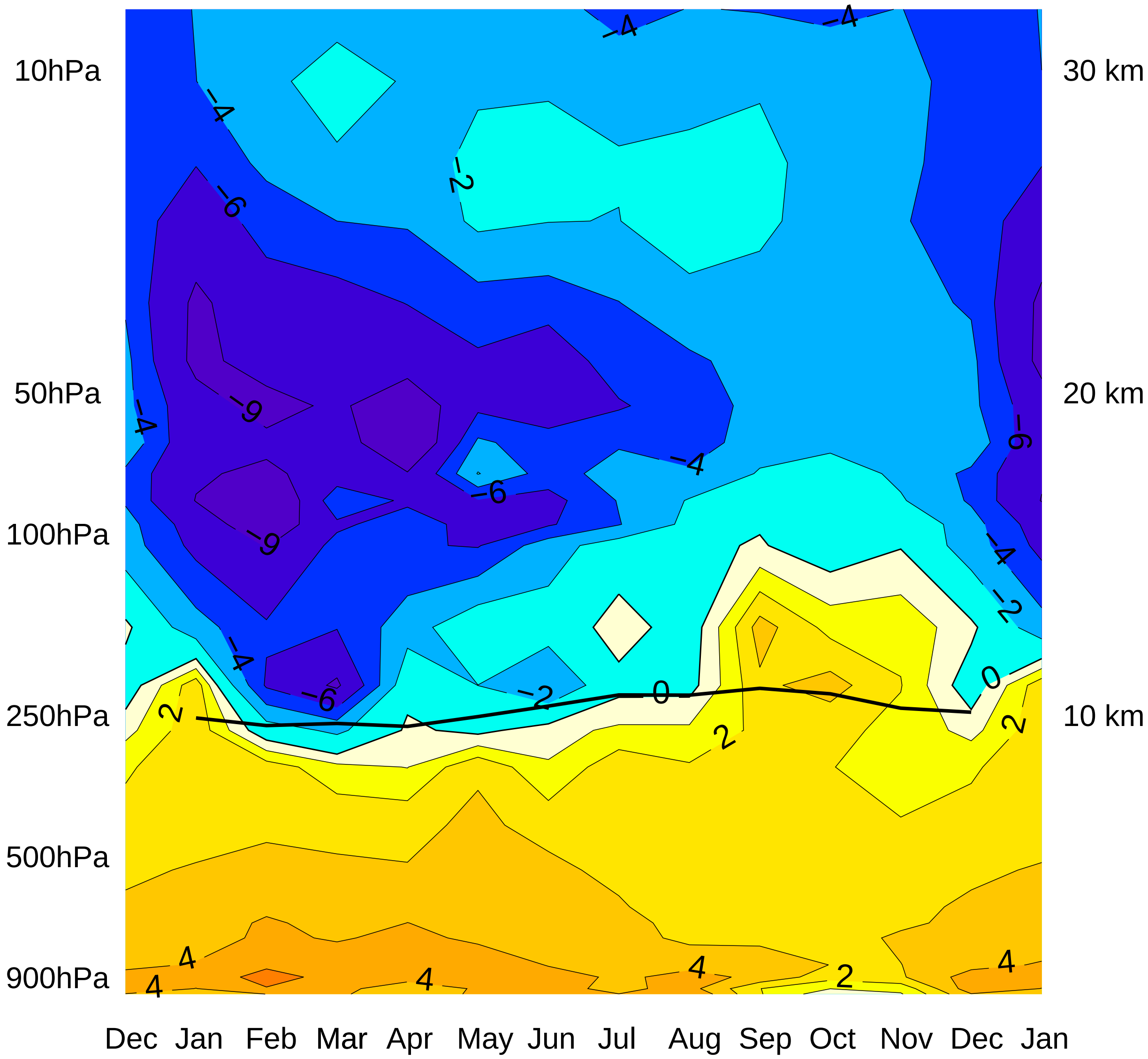

Figure 6.7: Isoplethes of observed ozone monthly trends between 1970 and 2000, calculated for 25 pressure levels between 7 and 925 hPa of the normal dataset (0.9 < CF < 1.4) and expressed in nbar per decade (absolute trends). Positive values in warm colours, negative values in cold colours. The climatological tropopause altitude is represented by the black line around 10 km.

Monthly trends for the normal dataset are again presented in Figure 6.7, but this time as absolute trends expressed in nbar per decade. The comparison between Figures 6.6 and 6.7 shows the same features in the tropopause region, but also illustrates large differences in the troposphere. The absolute trends are smaller in the troposphere than in the stratosphere and their annual cycle is strongly reduced in the troposphere. The strong winter maximum in the troposphere in Figure 6.6 loses its strength in Figure 6.7.

Monthly relative trends for the reduced dataset are presented in Figure 6.8. The features discussed in section 6.1.3 appear again, especially the negative trend under the climactical tropopause in June, where erroneous tropopause altitudes during the 70's have been suspected.

In order to get some insight into the confidence level of this monthly analysis, it is necessary to complete the observed trends with their statistical significance, which is commonly expressed by the p-values. The p-value is the level of significance for which a statistical quantity lies on the boundary between acceptance and rejection of a null hypothesis stating that it is zero. The confidence level is 100% (1 - (p-value)). For example, a p-value < 0.05 means that with at least 95% probability the statistical quantity is different from zero, because the null hypothesis is rejected at that 95% level. The same level of confidence is given to a quantity whose two standard error limits do not encompass the zero point. This is a common choice for calling a quantity statistically significant.

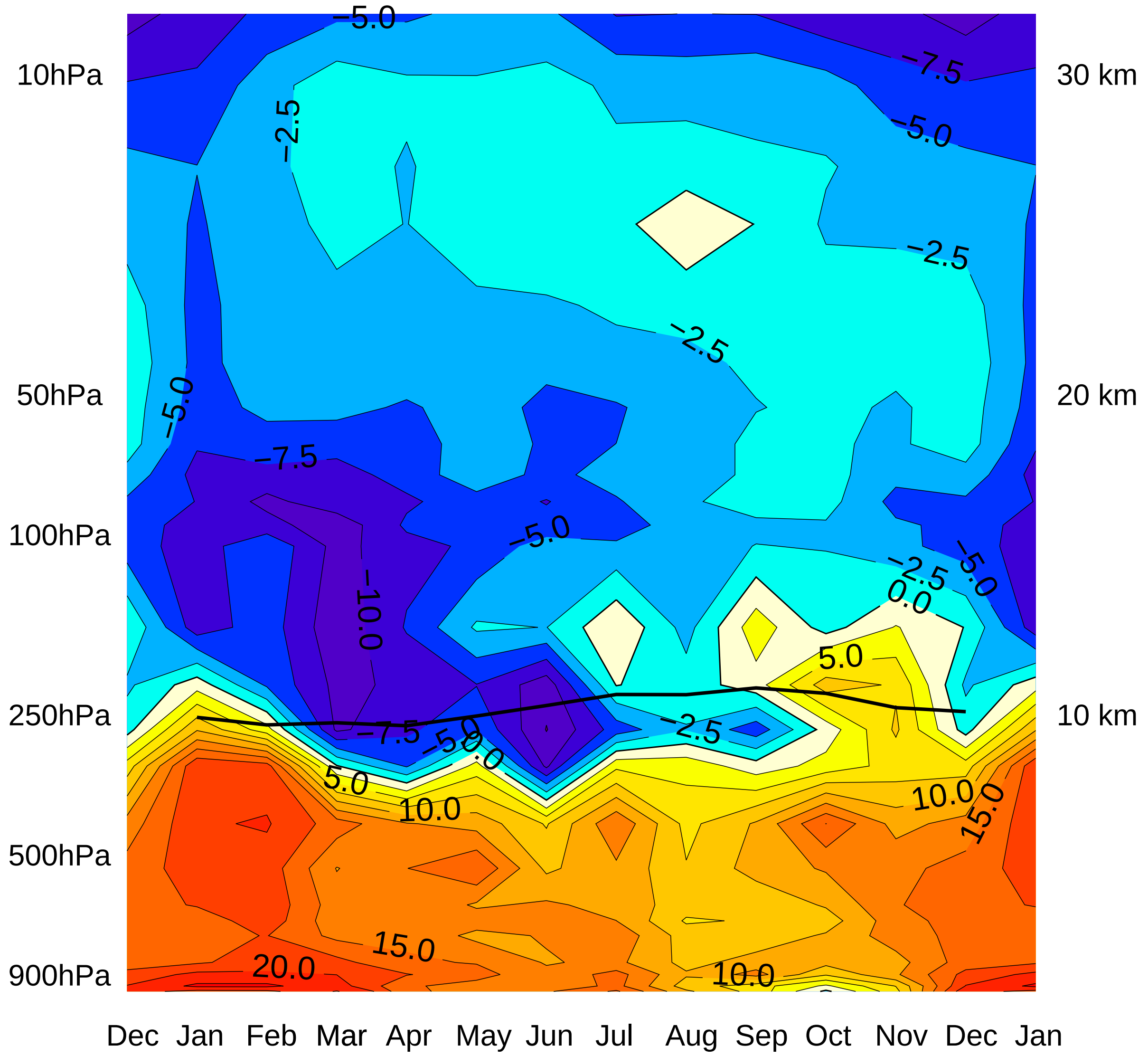

Figure 6.8: Same as Figure 6.6, but for the reduced dataset (0.95 < CF < 1.25). Trends are expressed in % per decade.

For altitudes and months appearing in white in Figure 6.9, the trends of Fig. 6.6 to 6.8 are not significant at a 90% confidence level. The normal dataset provides significant monthly trends at more altitude levels than the reduced dataset, due to the bigger number of soundings. The monthly trends are not significantly different from zero at the 90% level within a deep layer around the tropopause, including the lower stratosphere. This Figure provides similar results with Figures 6.3 to 6.5, but on a monthly basis. Although monthly ozone trends are nearly never highly significant near the tropopause, it does not hinder qualitative interpretations like those presented above, but it prevents attributing them a high confidence level.

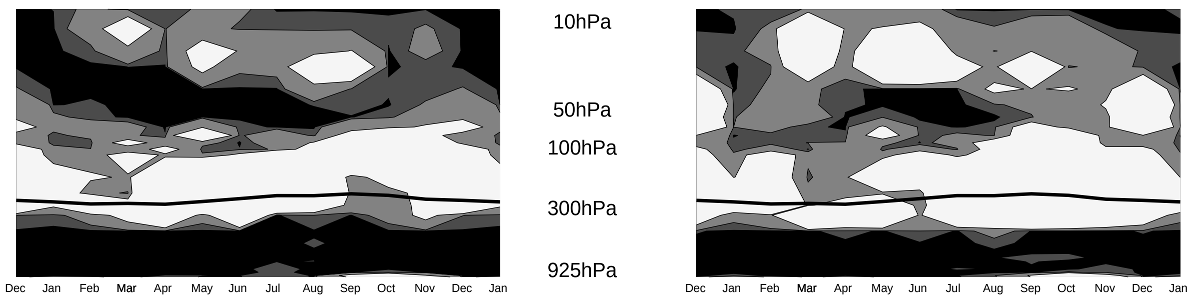

Figure 6.9: Statistical significance of the observed trends, left for the normal dataset (Fig. 6.6-7), right for the reduced dataset (Fig. 6.8). Black indicates a very high significance (p-value < 10^{-3}), dark and light grey high to medium significances (p-value between 10^{-3} and 10^{-2}, respectively between 10^{-2} and 10^{-1}). White (very light grey) areas correspond to not significant trend values at the 90%-level (p-values greater then 0.1). The climatological tropopause in depicted by the thick curve.

6.2 Ozone trend modelling with atmospheric explanatory variables

The analysis of the ozone trend requires a more sophisticated model than the previous, taking into account additional explanatory variables in order to identify the variability due to natural processes (e.g. 11 year solar activity cycle, large scale atmospheric processes) and to analyse the influences of selected processes (e.g. WMO, 1998 and 1999; Staehelin et al., 1998 and 2001; Weiss, 2000; Weiss et al., 2001). Although the Payerne ozone time series is not fully homogeneous, the results presented in the previous section favour the normal dataset for performing a more detailed trend analysis and updating previous studies (e.g. Staehelin et al., 1991; SPARC/IO_3C/GAW, 1998; WMO, 1999; Weiss, 2000).

6.2.1 Model and explanatory variables

The trend model used to estimate long-term ozone trends attributed to anthropogenic causes are often autocorrelated linear models (see WMO, 1999 for a review). The updated model of Weiss (2000) is used here. It does not postulate a constant contribution of the explanatory variables for all periods of the year and replaces the autocorrelation term for the error by a simple error term:

$$oz' = \sum_{i=1}^{k} (c_i \times ev_i) + trend + \varepsilon$$

Where ev_i are the different explanatory variables and c_i their respective weight in the calculation of oz', the deviation of the measured ozone concentration from the monthly (or seasonal, annual) mean. *trend* is the trend not explained by the explanatory variables (constant multiplying a linear ramp starting 1970 and rising 1 per decade) and *epsilon is* the residual error term. A stepwise regression is performed independently for each level and each period (year, season or month). C_p statistics, a measure related to the Alaike's Information Criterion, is used to decide if a term needs to be dropped. The Splus routine **step** is used. It uses itself the Splus routine **lm** (MathSoft,1993). By this procedure, the contributions c_i of the explanatory variables and the trends c are found with their error bounds and significance, i.e. p-values.

The strong day to day variability in the ozone profile at mid latitudes is connected to the weather dynamic, which also depends on the season. The tropopause pressure can be used to interpret these synoptic influences on ozone trends (Weiss et al., 2000; Appenzeller et al., 2000). The tropopause has been derived from the ozone balloon soundings with the WMO algorithm (Figure 6.10). Alternatively, it can be extracted as monthly means from the NCEP-re-analysis data.

Other climate variables that can influence the ozone trend are the North Atlantic Oscillation (NAO, see Hurrel (1995)) and the Arctic Oscillation (AO, see Thompson and Wallace (1998)), both defined using sea surface pressure measurements. They describe the general meteorological situation over Europe and the Atlantic region and are appropriate for the investigation of interannual variations. However, the physical driving of the NAO and AO is unknown at present and it is possible that the observed NAO/AO long-term behaviour is fully natural (Christiansen, 2000) or a response to anthropogenic forcing (Paeth et al., 1999; Shindell et al., 1999). The NAO index is calculated from NCEP re-analysis data (all internet addresses at the end of the report), whereas the AO is provided by Thompson and Wallace.

Another dynamical variable which can influence ozone trend is the seven months lagged Quasi-Biennial Oscillation (QBO) of which only the positive phase is used. The QBO is the most prominent stratospheric phenomenon in the tropics but it also impacts on ozone variations at mid latitudes (Marquardt, 1997).

The solar activity influences the ozone production in the stratosphere, and its 11-year cycle has to be considered in ozone trend determination (Shindell et al. 1999). A commonly used index is the solar radio flux at 10.7 cm that is provided by NOAA.

The dynamical changes described above are considered as reflecting only the atmospheric natural variability. A combination of the anthropogenic and natural influences on the ozone trend is taken into account with the Ozone Depletion Factor (ODF, Solomon et al., 1996), which combines the effects of stratospheric natural aerosols and stratospheric anthropogenic chlorine loading. Large volcanic eruptions have indirect effects on ozone levels. Although Mt. Pinatubo's 1991 eruption did not increase by itself stratospheric chlorine concentrations, it produced large amounts of aerosols. Aerosols increase chlorine's effectiveness at destroying ozone. The aerosols increased therefore the ozone depletion due to the presence of CFC-based chlorine. In fact, the aerosols increased the efficiency of the CFC siphon, lowering ozone levels even more than would have otherwise occurred. Unlike long-term ozone depletion, however, this effect is short-lived. The aerosols from Mt. Pinatubo have disappeared in year 1994, according to satellite, ground-based, and balloon observations, and the strong ozone depletion following this volcanic eruption has returned close to the previous trend line.

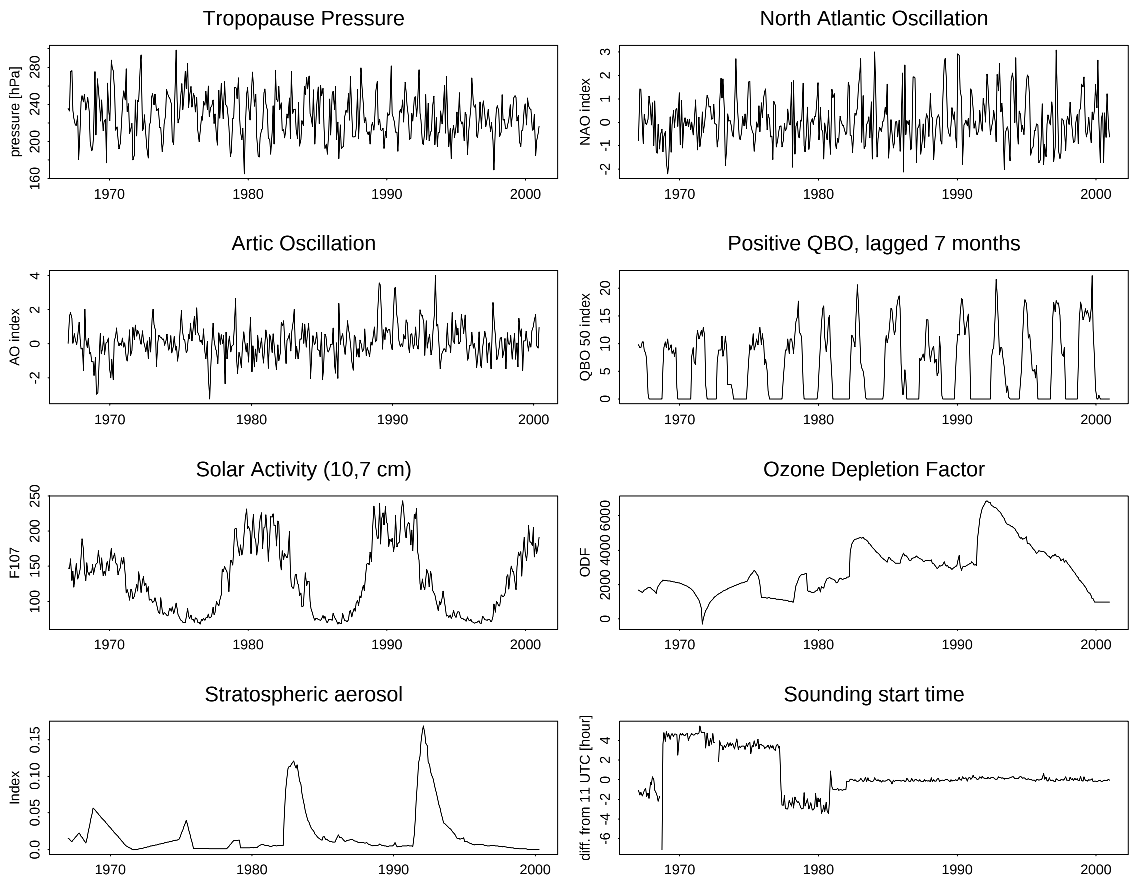

Figure 6.10: Time series of the explanatory variables used in the trend model (from 1967 to 2000, data sources given in the references). Two variables are extracted from the soundings. Top left panel: tropopause pressure of the normal dataset. Bottom right panel: launch times relative to 11 UTC used in chapter 6.3.

Although the stratospheric aerosol load due to volcanic eruptions is a natural factor, Staehelin et al. (1998) introduced it first in a trend model applied to the Payerne dataset in order to take into account anthropogenic influences. Indeed, the aerosols are believed to be important to describe the mid-latitude in-situ heterogeneous ozone depletion by ozone depleting substances. Without a chemically disturbed stratosphere, the chemical mid-latitude ozone depletion would be expected to be smaller (increase of sulfate aerosol levels with significant radiative/chemical impacts and changes in vertical and horizontal transport processes). Consequently, Weiss et al. (2001) replaced the aerosol by the ODF in their newer trend calculations. Nevertheless, the time series of the stratospheric aerosol and of the ODF (Figure 6.10) show very different recovery times after the volcanic eruptions and in other studies, the large volcanic eruptions have been considered as contributing to the ozone natural variability, and their effects separated or not from those of other natural parameters (see references in WMO, 1999). In the present study, the stratospheric aerosol content is introduced in the trend model and considered as a natural factor, reducing probably too much the anthropogenic part of the long-term trend. According to a common practice, the remaining long-term ozone trends not explained by the natural factors introduced in the model are attributed to the anthropogenic release of ozone depleting substances.

6.2.2 Statistical significance of the explanatory variables

In order to illustrate the role of the different explanatory variables in the trend model, a significance analysis has been performed for monthly values following the procedure used by Weiss (2000). The model has been run five different times with the normal dataset (0.9 < CF < 1.4) in order to evaluate the significance of the different explanatory variables:

1- One time with all of the non-dynamical explanatory variables (QBO, solar flux and ODF) and with tropopause pressure as dynamical explanatory variable, for the significance of the QBO, the solar flux, the ODF and the tropopause pressure.

2- One time with all of the non-dynamical explanatory variables (QBO, solar radio flux, but aerosol replacing ODF) and tropopause pressure, for the significance of the stratospheric aerosol load.

3- One time with all of the non-dynamical explanatory variables of run 1 and with NAO as the dynamical explanatory variable, for the significance of the NAO.

4- One time with all of the non-dynamical explanatory variables of run 1 and with AO as the dynamical explanatory variable, for the significance of the AO.

5- One time without any explanatory variable for the significance of the observed trend. This run delivered the results of chapter 6.1.

The three dynamical variables (tropopause, NAO and AO) can not be used at the same time in order to avoid co-linearity problems. Therefore, their study needs the three runs 1, 3 and 4.

The computed p-values for the different explanatory variables are plotted in Figure 6.11, showing where (at which pressure level) and when (in which month) each variable is significant or not for the ozone trend.

The tropopause pressure exhibits a significant influence on the ozone trend throughout the year in the upper troposphere and in the lower stratosphere. There are some other slight tropopause signals in the lowest troposphere which are hard to interpret. Replacing the ODF by the aerosol load has a slight influence on the tropopause significance.

The alternate dynamical variables (NAO, AO) also have an influence on ozone trends, especially in the winter-spring months. NAO and AO contribute to the variability of ozone in the middle stratosphere during late Summer.

At the altitude of the ozone maximum, the QBO exhibits a strong significance during winter and spring months. The solar flux is slightly significant above 100 hPa only during the summer months. The solar flux seems to be significant for the ozone trend at selected pressure levels and seasons where the QBO is not significant.

The ODF shows some influence in the upper domain of the ozone soundings, but only from April to October. During winter months, it exerts a more significant influence in the low stratosphere. The stratospheric aerosol load seems more significantly linked to the ozone trend than the ODF during the winter months, but shows no stratospheric link in the summer months.

This significance analysis has been partly repeated on the reduced dataset, using the tropopause data of the normal dataset and the variables corresponding to run 2. The results related to the tropopause are compared in Figure 6.12. From a pure statistical point of view, the normal dataset delivers better results than the reduced one, confirming the findings of section 6.1.3. For the second dataset, the tropopause pressure exerts a significant influence on the ozone trend in the whole tropopause during June-August.

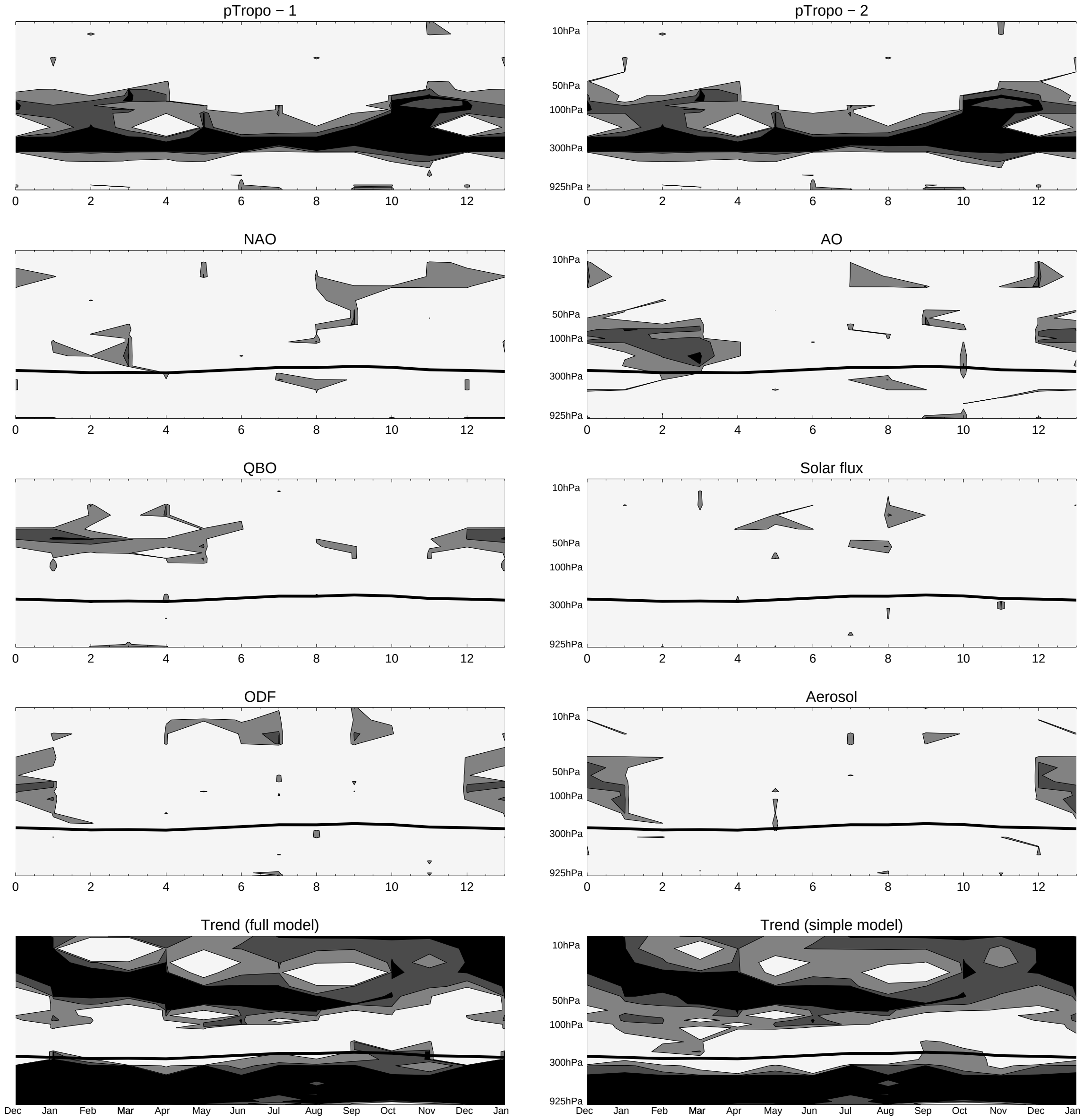

Figure 6.11: Statistical significance of the influence of different explanatory variables on the ozone trend calculation with respect to pressure level and month. The top two panels show the statistical significance of the tropopause pressure (runs 1 and 2). The next row of panels are for the NAO and AO indexes (runs 3 and 4). The third row is for QBO and solar flux (run 1). Fourth row for ODF (run 1) and aerosol load (run 2). The bottom panels are for the unexplained trend (run 1) and for the observed trend without explanatory variable (run 5). Black areas indicate high significance ($p < 0.001$), dark and light grey still good significance ($0.001 < p < 0.01$, $0.01 < p < 0.1$). White (very light grey) indicates low significance ($p > 0.1$), implying that this variable does not contribute with significance to the ozone variability in the trend model.

This significance analysis of explanatory variables based on monthly values shows that the tropopause pressure is the most dominant variable. All the other variables contribute significantly to the trend model only for a few months and a few levels. These results match the findings of Weiss et al. (2001), with some differences that are probably due to the different aggregations of the altitude levels and - possibly - to the last re-evaluation work on the soundings. Weisset al. (2001) performed a more comprehensive sensitivity analysis, with other combinations of explanatory variables and other aggregation periods, so as to maximize the statistical significance of the results. Although the tropopause pressure and the NAO can not be used at the same time, Weiss et al. (2001) used them alternatively for different atmospheric layers: the tropopause for the lowest stratosphere and the NAO for the higher altitudes, which resulted in higher explained variability in the altitude range above ca. 20 km.

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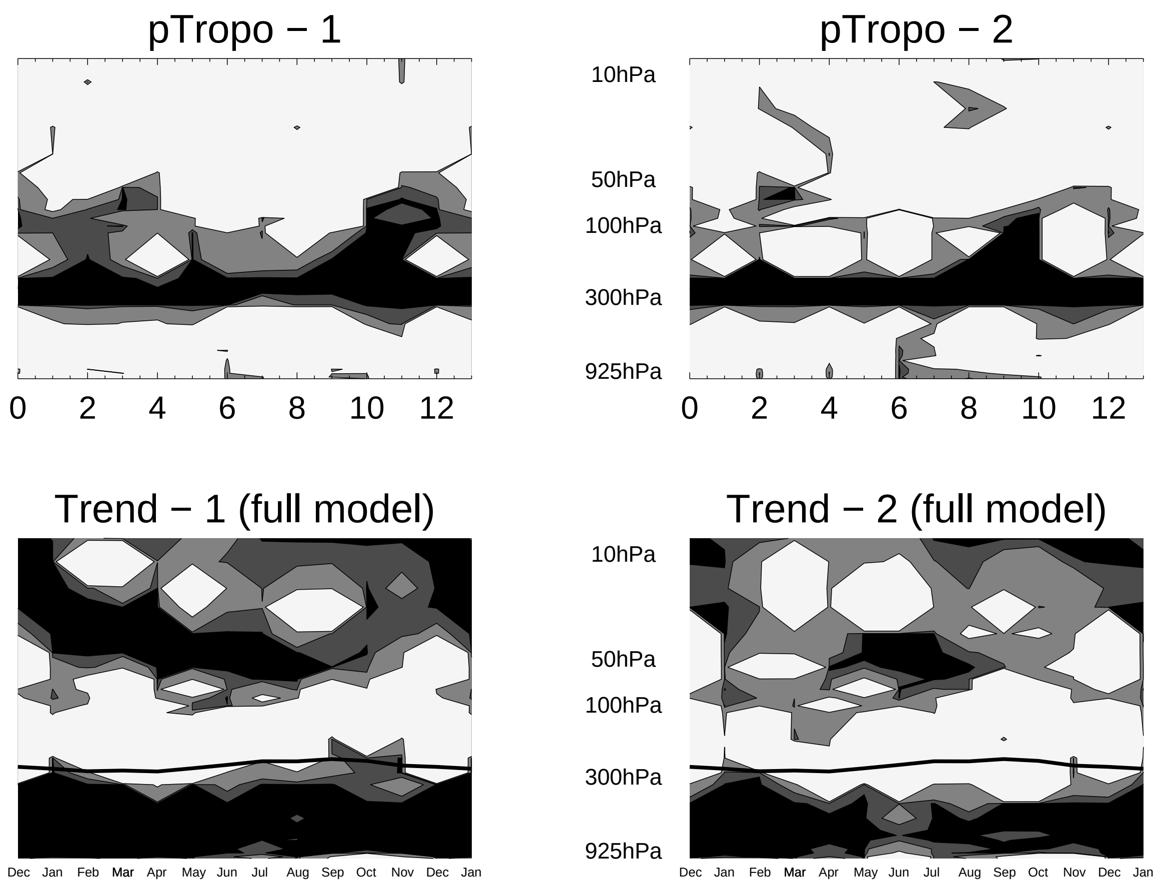

Figure 6.12: Statistical significance of the influence of tropopause pressure (pTropo) on ozone trend calculation with respect to pressure level and month. The left top panel is for the normal dataset (0.9 < CF < 1.4). The right top panels for the reduced dataset (0.95 < CF < 1.25). The bottom panels depict the significance of the related unexplained trends. See Fig. 6.11 for other explanations.

Since results issued from the combination of explanatory variables involving the tropopause pressure, QBO, solar flux and ozone depletion factor - or alternatively stratospheric aerosol - are the most significant and correspond to a wide practice, these two combinations have been retained for the further trend calculations (runs 1 and 2). The choice between ODF and aerosol as an explanatory variable partly depends on the objective. The first set allows the introduction of one clear anthropogenic factor, whereas the second one considers natural factors as well as a - may be partly - anthropogenic factor. In this latter case (aerosol), the calculated trend, which is not explained by these variables, can be considered as a lower limit for the anthropogenic trend. Statistically speaking, the anthropogenic trend is an unexplained trend whose origins have been the subject of intensive research , based on physical and chemical atmospheric simulation models (WMO, 1999) within the international research community.

6.2.3 Annual trends

The calculations of annual trends (runs 1 and 2) are based again on the normal dataset, and the resulting trends are first compared with the observed trends. Figure 6.13 shows the absolute trends in nbar per decade and it must be compared with Figure 6.1. In order to make this comparison easier, the mains results of Figure 6.1 are repeated in Figure 6.13. The full model with stepwise regression delivers only trends significantly different from zero at the 95% confidence level. This explains the absence of trends at the 150 and 200 hPa levels. The model finds significant trends at all other levels of the profile for both runs (ODF or aerosol).

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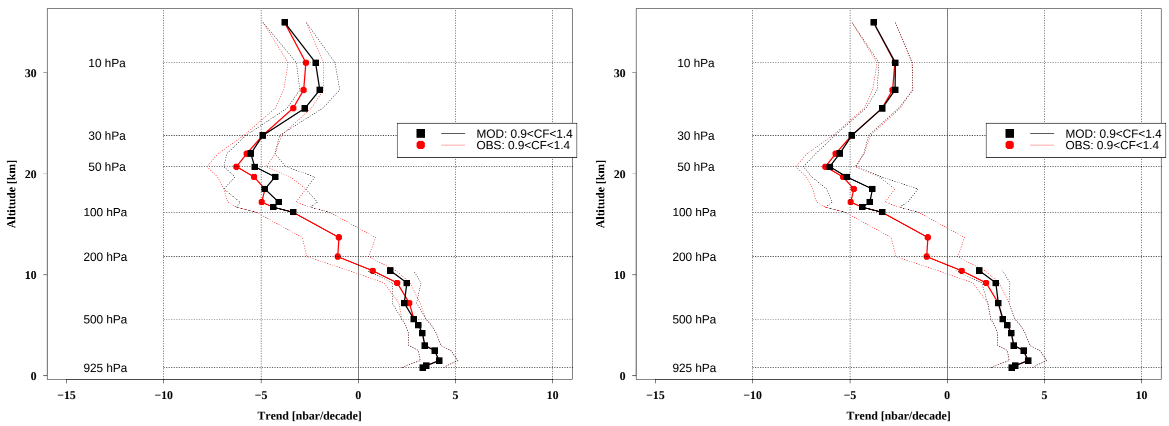

Figure 6.13: Annual ozone trends of the Payerne ozone soundings over the period 1970-2000 in nbar per decade. In black for the trend model with explanatory variables (MODel), in red for the simple model (OBServation). In the left panel, the full model uses ODF, whereas in the right panel ODF is replaced by the stratospheric aerosol load. The dotted curves reproduce the trend uncertainties (two standard errors). The normal dataset is used in all cases (0.9 < CF < 1.4). Only the significant values of the full trend model are shown.

There is a good agreement between all results at all levels. Going more in the details of the profile, it is worth noting that all two trend calculations agree perfectly in the low and middle troposphere up to 500 hPa, because none of the explanatory variables significantly impacts its trend calculation on an annual basis. The introduction of other explanatory variables, more relevant to tropospheric processes, could be helpful (Staehelin, 1991). In the upper troposphere between 400 and 250 hPa, the two new runs provide almost the same trends, which are slightly different from the observed trends. The tropopause influence increases with altitude and the unexplained trends are there slightly larger than the observed trends. At 300 hPa, the full trend model is significant, whereas the simple trend model is not. At the next two pressure levels above the tropopause (200 - 150 hPa), the full trend model is not significant and hence agrees at the 95% significant level with the simple model of section 6.1.2 (uncertainty enclosing the zero trend line). Actually, at the 200 hPa level, the model finds a significant tropopause influence, but the resulting trend is not significant. At the next 100 and 90 hPa levels, all results agree, which means that no explanatory variable exerts a significant influence. According to Figure 6.12, the tropopause pressure is not a significant explanatory variable throughout the year at these two levels, because its seasonal variation is more important than its interannual variation. At the next level, the tropopause is involved again

From the 70 hPa level up to 7 hPa, the tropopause pressure is no more relevant, and the differences between the different calculations are due to QBO, ODF and solar flux. The ODF partly governs the differences between the three calculations in the middle stratosphere. Above 70 hPa, the trends calculated with exclusively natural explanatory variables are nearly the same as the observed trends.

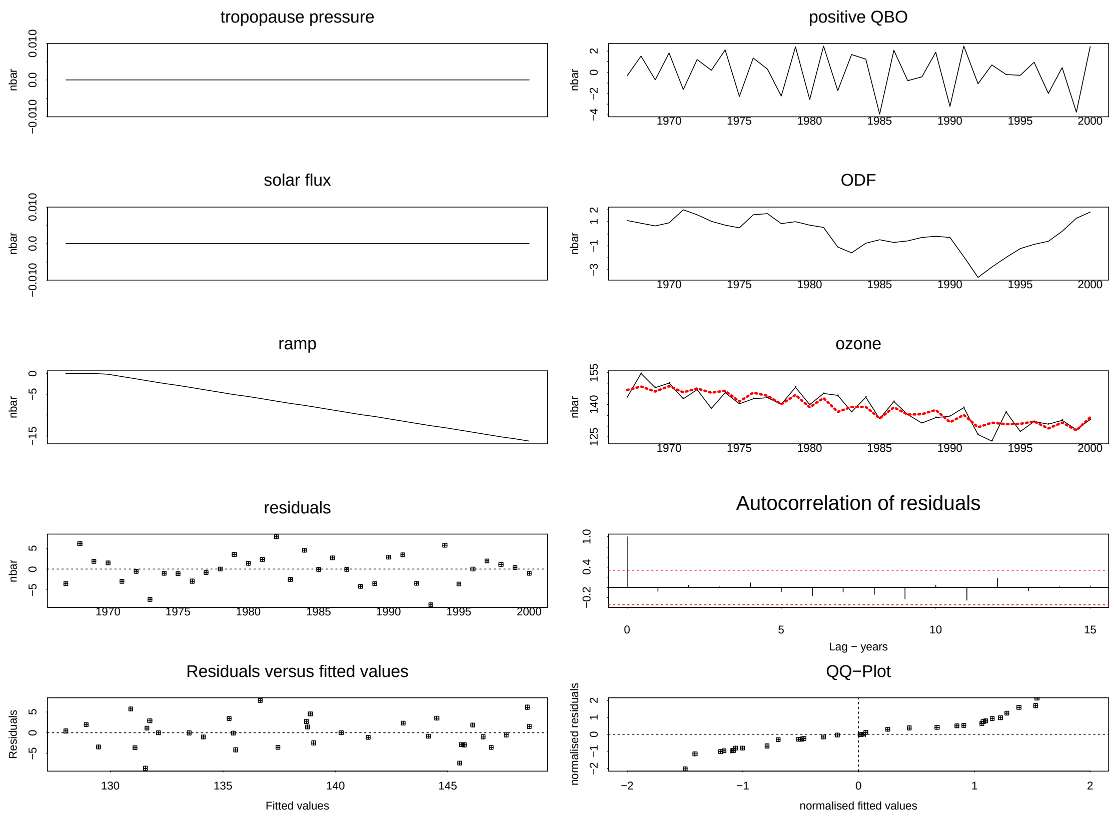

Figure 6.14: Results of the annual ozone trend modelling for the 50 hPa pressure level (run 1). Tropopause pressure and solar flux have no significant influence. The specific influence of the positive lagged QBO and of ODF on ozone are displayed in the corresponding panels. The measured annual ozone time series is represented by the continuous line in the ozone panel and the fitted values are superposed with a thick dotted red line.

The four bottom panels present the main results of the residual analysis: time series of the residuals, scatter plots of the residuals versus the fitted values, autocorrelation of residuals with 95% confidence limits for testing the hypothesis of no cycle component, quantile-quantile plot of the normalised residuals versus the normalised fitted values.

In order to improve the interpretation of the trend results, it is useful to compare the fitted times series with the measurements at different altitudes. Figure 6.14 helps explaining the influence of ODF on the ozone trend in the middle stratosphere, where only very large scale processes like QBO, ODF and solar flux are contributing on an annual basis. The linear trend term, positive QBO and ODF strongly modulate the ozone time series at this 50 hPa level (ozone maximum). The QBO and ODF both explain a variation of 5 nbar of the ozone values. Their combination explain almost all the variance of the measurements. The ODF appears to be the main cause of the trend slowering since 1993 at that altitude. The model results can be considered as valid according to the residual analysis provided by the four bottom panels of Figure 6.14.

Figure 6.15 shows the results of run 2 with stratospheric aerosol replacing the ODF, but is otherwise the same as Figure 6.14. The positive QBO, stratrospheric aerosol and solar flux contribute to the explanation of the variance of the ozone. Solar flux modulates ozone with an amplitude of 4 nbar; the positive QBO contributes to the same variability as in run 1 and the ODF modulates the ozone time series somewhat less as in run 1. The overall action of these three variables brings the model results close to the measurements. The combination of the natural variables (including aerosol, according to 6.2.2) or of the natural variables and ODF can well explain the apparent trend decrease since the mid 1990's at the 50 hPa level, as well as at most stratospheric levels.

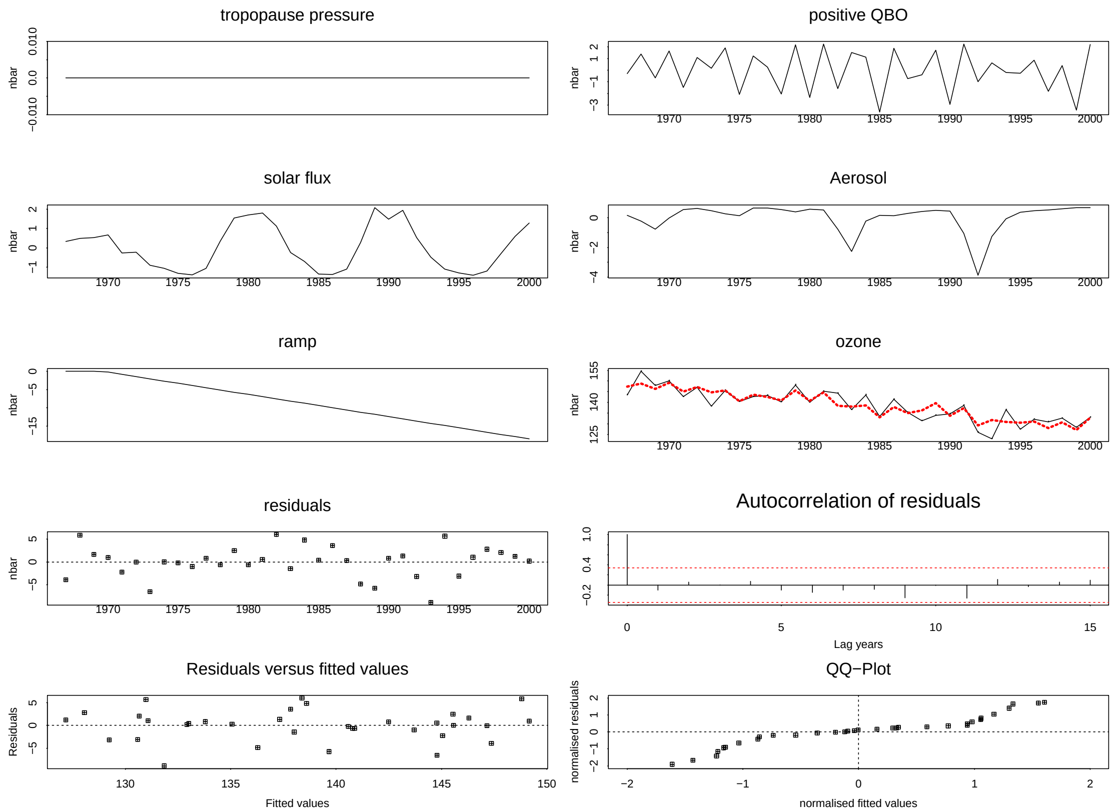

Figure 6.15: Results of the annual ozone trend modelling for the 50 hPa pressure level (run 2). The tropopause pressure has no significant influence. The specific influence of solar flux, positive lagged QBO and stratospheric aerosol on ozone are displayed in the corresponding panels. The measured annual ozone time series is represented by the continuous line in the ozone panel and the fitted values are superposed with a thick dotted red line. The four bottom panels present the main results of the residual analysis.

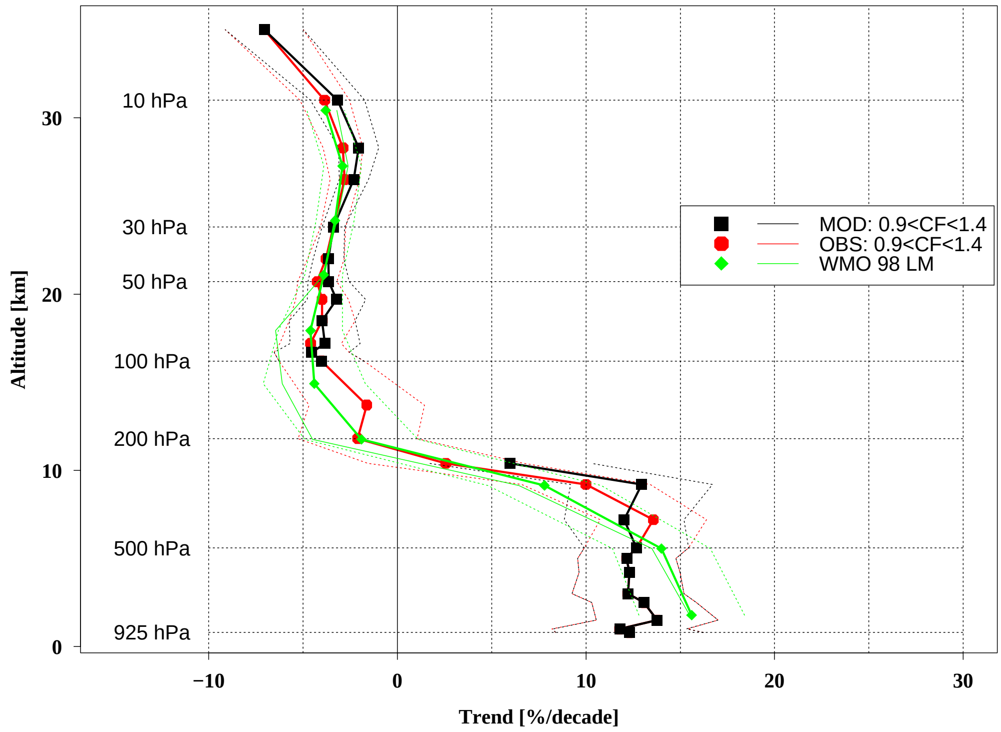

Figure 6.16: Annual ozone trends of the Payerne ozone soundings over the period 1970-2000 in percent per decade. In black for the trend model with explanatory variables (MODel), in red for the simple model (OBServation). The full model uses the ODF. The dotted curves reproduce the trend uncertainties (two standard errors). The normal dataset is used in all cases (0.9 < CF < 1.4). The full trend model delivers trends only in the case they are significant. The SPARC/IO_3C/GAW trend results (WMO, 1998) for the period 1970-1996 are included (LM: Logan and Megretskaia) in green.

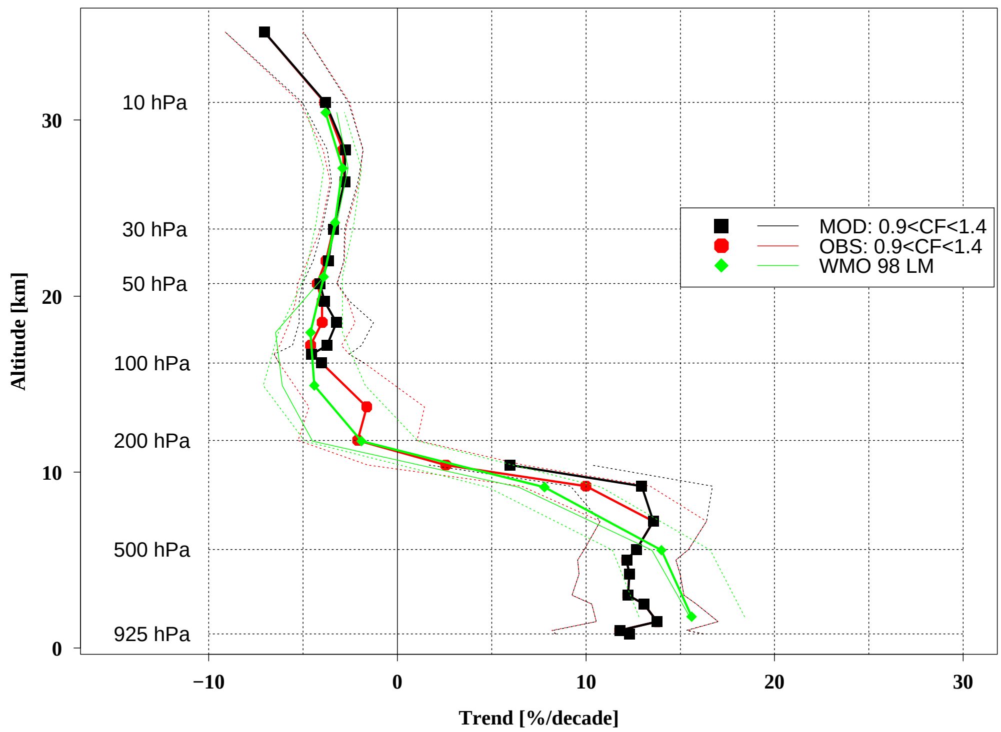

Figure 6.17: same as Fig. 6.16, but the full model uses the stratospheric aerosol instead of the ODF.

Figures 6.16 and 6.17 show the same trends as in Figure 6.13, but in relative units (% per decade), as well as the comparable SPARC/IO$_3$C/GAW trend results. In addition to the previous interpretation, the fair agreement of the present results with those of the last international assessment can be pointed out. The new results show a closer agreement with the tropospheric trends at Hohenpeissenberg.

Not only the statistical uncertainty of the trend model is important, but also the instrumental uncertainty. Both have to be added, assuming for instance that they are independent. Tentative values for the instrumental uncertainties leading to potential drift errors in the data have been introduced in Table 2.3 according to WMO (1999). The resulting combined uncertainty could be as large as twice the statistical model uncertainty given in the Figures 6.16 and 6.17. The WMO experts give no value above 15 hPa, considering the ozone sondes not adequate for reliable trend determination above 27 km. Despite the enlarged combined uncertainty, the trends of Figures 6.16 - 6.17 remain significantly different of zero in the troposphere and in the low - middle stratosphere.

The ozone variance explained by the full statistical model (annual values) can be found in Figure 6.20 of the next section.

As a first conclusion related to the trends computed with annual means, the following points can be highlighted:

- the full model with explanatory variables brings a slight improvement compared to the simple trend model without any explanatory variable.
- The tropopause strongly impacts the results just under and above its mean altitude.
- When considering the ozone depletion factor (ODF) instead of the stratospheric aerosol, the negative linear trend in the middle stratosphere is slightly reduced.
- The natural factors including the stratospheric aerosol load explain the apparent trend slowering since 1993; the use of the ODF instead of the stratospheric aerosol load gives similar results.

6.2.4 Seasonal trends

Seasonal trends are computed in order to better use the set of explanatory variables and hence to obtain a better model performance. Figures 6.18 and 6.19 show the seasonal trends that have been computed similarly to the annual trends of the previous section (runs 1 and 2 based on seasonal values).

In the troposphere, the full model provides significant positive trends up to the tropopause level, somewhat higher than the simple model does (Figure 6.3). This has already been seen for the annual trend; in the stratosphere, the negative trend is smaller and is even not more significant in the Summer at some levels when the ODF is taken into account in the model. Otherwise, the results are the same as for the simple trend model; e.g. in Autumn, significant positive trends appear in the lower stratosphere. As no truly tropospheric explanatory variable has been introduced, only slight differences between the different seasonal models can be expected in the low and middle troposphere.

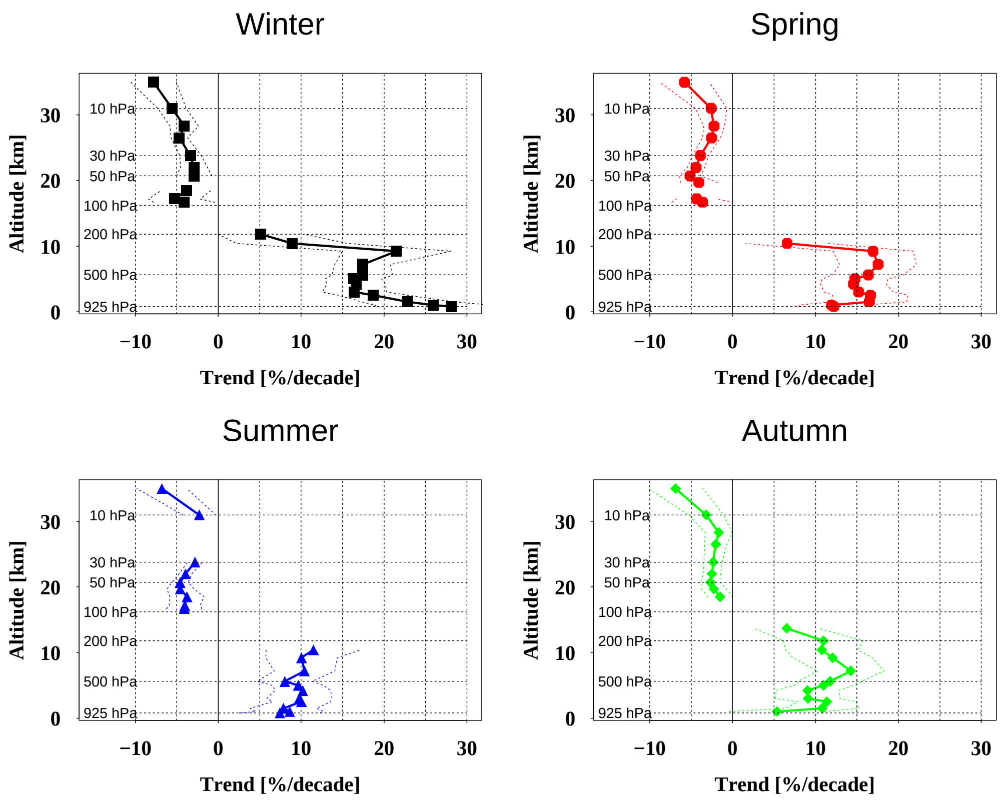

Figure 6.18: Seasonal ozone trends of the Payerne ozone soundings over the period 1970-2000 in percent per decade for the normal dataset (0.9 < CF < 1.4). Calculation with the full model including ODF. The dotted curves reproduce their uncertainties (2σ). The full trend model delivers only significant trends.

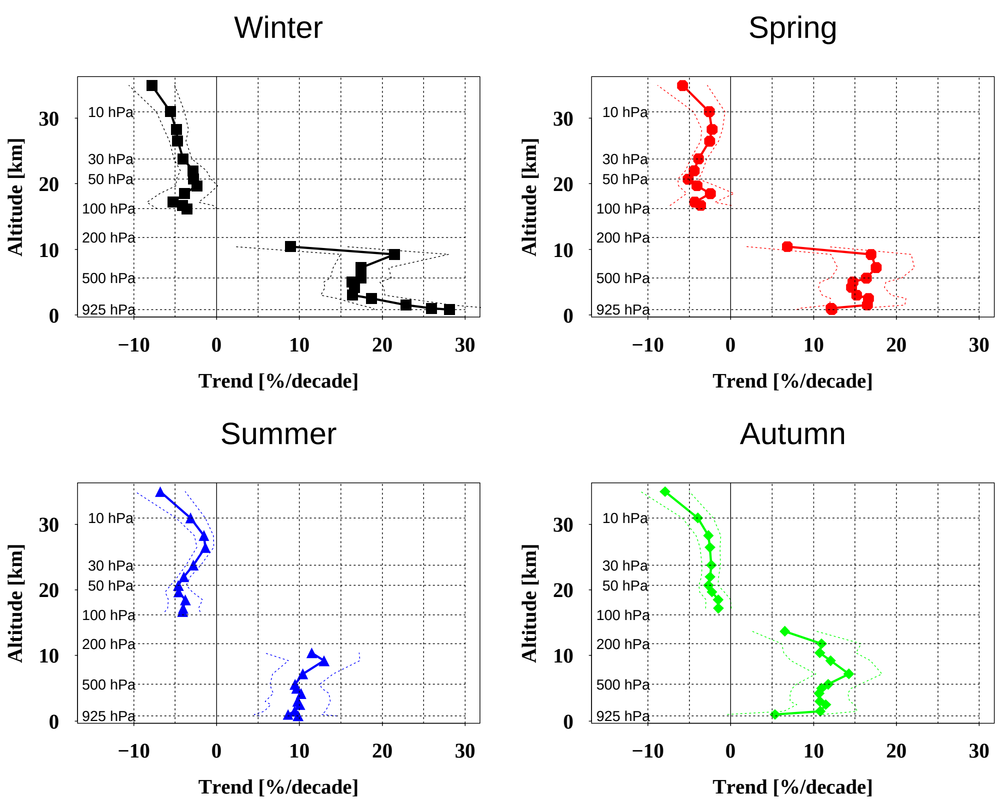

Figure 6.19: Same as Figure 6.18, but calculation with the full model including stratospheric aerosol.

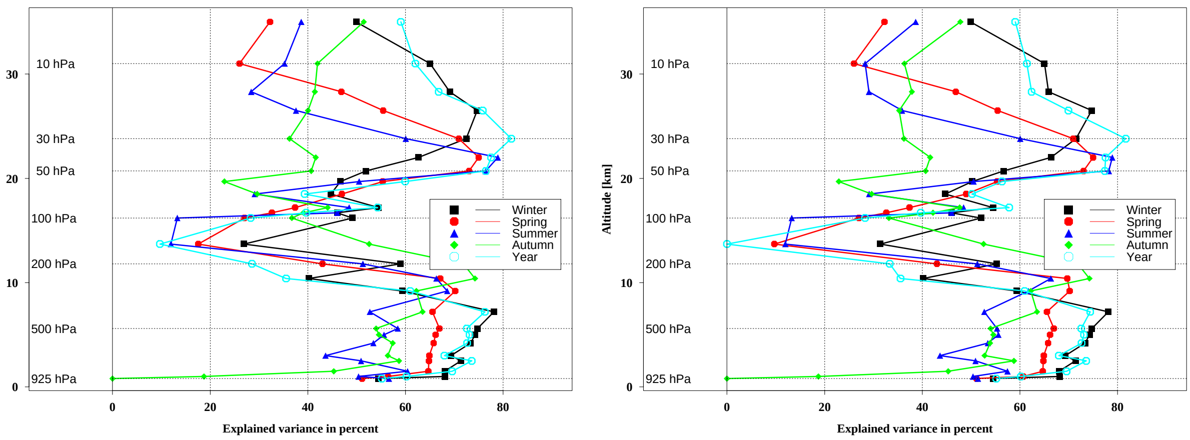

Figure 6.20: Variance (R^2) of ozone explained by the full trend model for the four seasons as well as for the year. Left panel: run 1 (Fig. 6.18). Right panel: run 2 (Fig. 6.19).

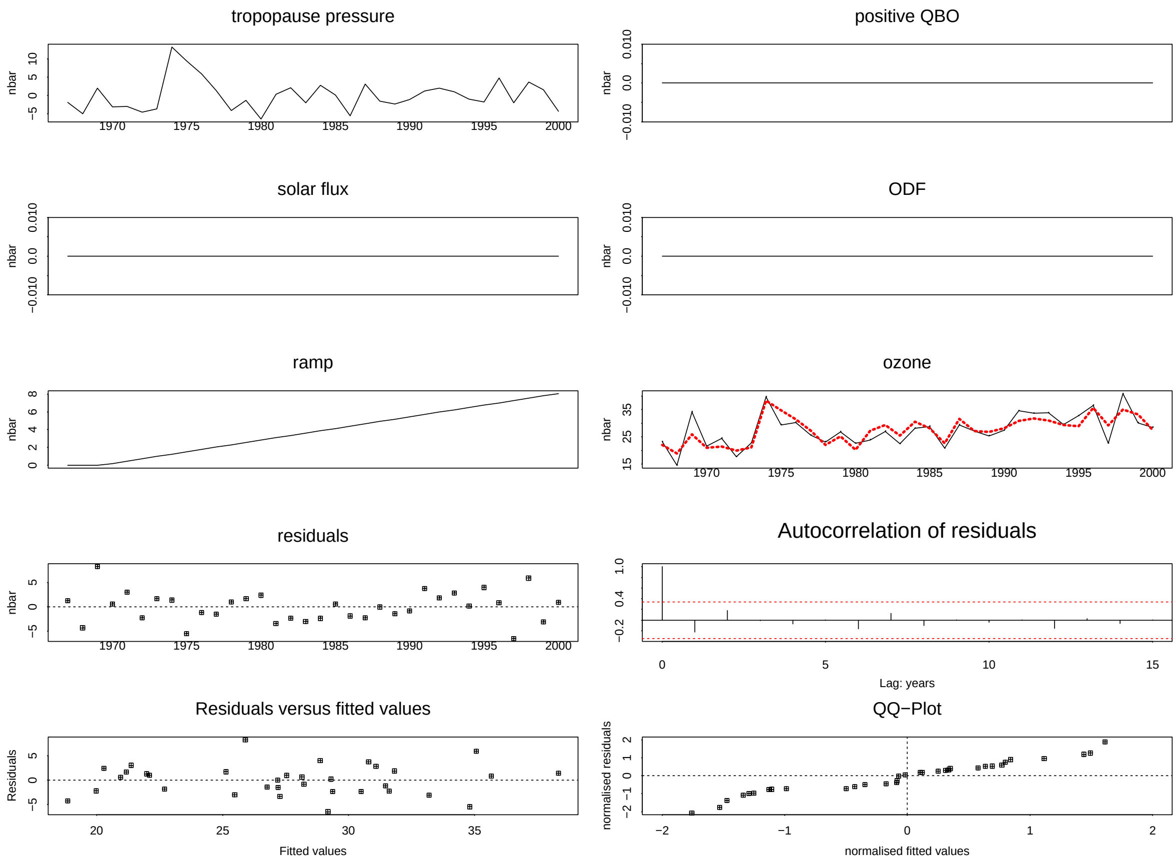

Figure 6.21: Results of the autumnal ozone trend modelling for the 200 hPa pressure level (run 1). Tropopause pressure alone has a significant influence on ozone (top left panel). The measured annual ozone time series is represented by the continuous line in the ozone panel and the fitted values are superposed with a thick dotted red line. The four bottom panels present the main results of the residual analysis (see Figure 6.14 for further explanation).

The analysis of the explained variances allows another qualification of the different models. The comparison between Figures 6.5 (simple model) and 6.20 (full model) shows the improvement brought by the full model. In the lower and middle troposphere, the full model explains roughly between 40 and 80% of the ozone variability, with a gain of a few percents in comparison with the simple model. In the upper troposphere and the lower stratosphere, the mean improvement amounts to 20%; explained variances under 20% are no more the rule, but the exception. In the middle stratosphere between 50 and 10 hPa, the improvement is also noticeable and the maximum explained variance is as high as 80%. The choice of ODF or aerosol in the full model does not alter the explained variances (Figure 6.20).

The efficiency of the full model can be also analysed with the fitted values and the residuals. Figure 6.21 illustrates an example of the previously discussed anomaly in the tropopause time series between 1974 and 1976. This anomaly appears also very well in the ozone time series and the full model reproduces this feature quite well. The model reproduces also very well the measured time evolution in the 80's. The residual analysis in the bottom of the figure confirms the adequacy of the fit. As can be seen in the figure, the contribution of the tropopause pressure to the ozone concentration at this level near the tropopause varies between -5 to and 10 nbar, whereas the ozone concentration varies between 15 and 35 nbar. The correlation between tropopause pressure and ozone is hence very high near the tropopause, as it has been already recognised for the Payerne sounding (Weiss, 2000).

Figures 6.22 to 6.24 displays the comparison between the measurements and the fitted values for the four seasons at 3 pressure levels: 925 hPa (800 m asl or 300 m above ground level), 200 hPa (12 km asl, slightly above the mean tropopause level) and 50 hPa (20.5 km asl, level of the ozone maximum).

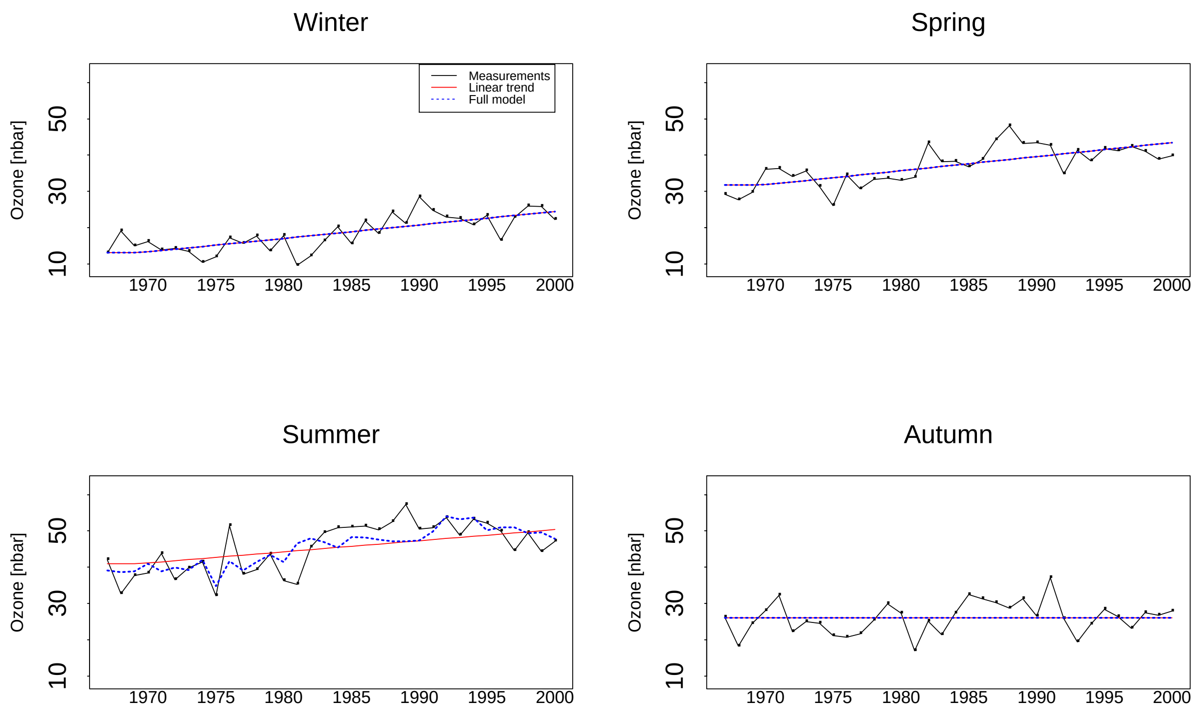

Figure 6.22: Ozone trend results for the four seasons at the 925 hPa level (300 m above ground level at Payerne). The measured ozone is the full black line, the linear trend is the red straight line, and the fitted values are given by the blue dotted line. If no explanatory variable has a significant contribution, the fitted values are assigned to the linear trend line (trend line being then blue). Full model with ODF.

At the 925 hPa level (Figure 6.22), the fit results are not so good. A trend break could be located in the beginning of the 80's, especially for Winter and Summer. It is worth noting that the ozone values are very low in Winter until the beginning of the 80's, when the SO_2 concentrations dropped to lower values (see section 6.1.3). No trend is detected in Autumn. The explained variance is low (see Figure 6.20). Except for Summer, no explanatory variable has a significant influence. A further discussion related to the launch time changes can be found in section 6.3.1.

As mentioned above, in Summer during the 80's, the model can not account for the strong deviation of the measurements compared to the linear trend at the 925 hPa level (Figure 6.22). At the other low to middle tropospheric levels, the model results deviate also strongly from the measurements during these years with deviations to the other European ozone stations. At 200 hPa and 50 hPa, the model has more success during these years and reproduces well the measurements (Figures 6.23 and 6.24).

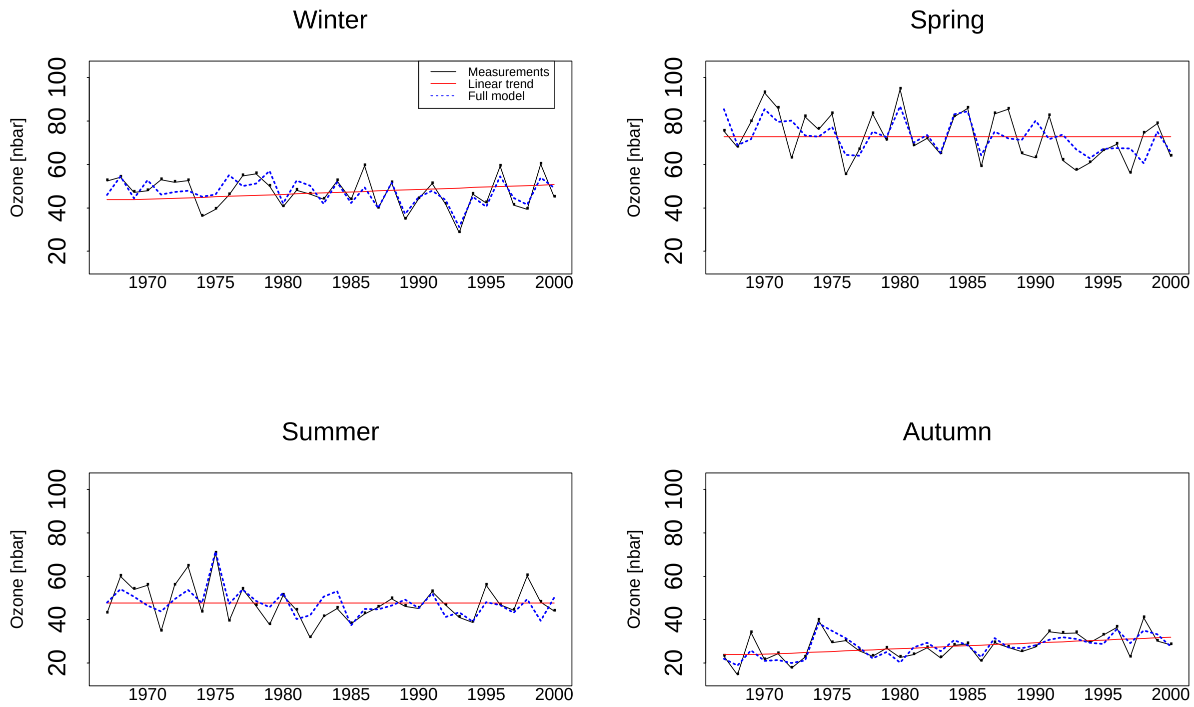

Figure 6.23: Same as Figure 6.22, but at the 200 hPa level (just above the mean tropopause).

During Winter and Spring, the tropopause pressure variations can explain a large part of the low stratospheric and upper tropospheric ozone trends (up to 30% in Winter and 60% in Spring: see Figure 6.25 below). The fitted values approach well the measurements, as can be seen in Figure 6.23 for the 200 hPa level. In Summer and in Autumn, the dynamical contribution is less important (< 20%, respectively < 12%).

A substantial ozone variability (up to 30% in Spring) in the upper stratosphere can be explained by the ODF, which also explains some variability (10-20%) in the upper troposphere during the summer months. This means that the anthropogenic stratospheric ozone trends in mid-latitudes are mainly caused by processes other than the in situ chemistry on aerosols described by the ODF (Weiss, 2000), like for instance the transport of ozone depleted air originating in the polar vortex. However, one has to consider that the major ozone depletion by CFC gas phase chemistry occurs above the altitude reached by the soundings (30-50 km).

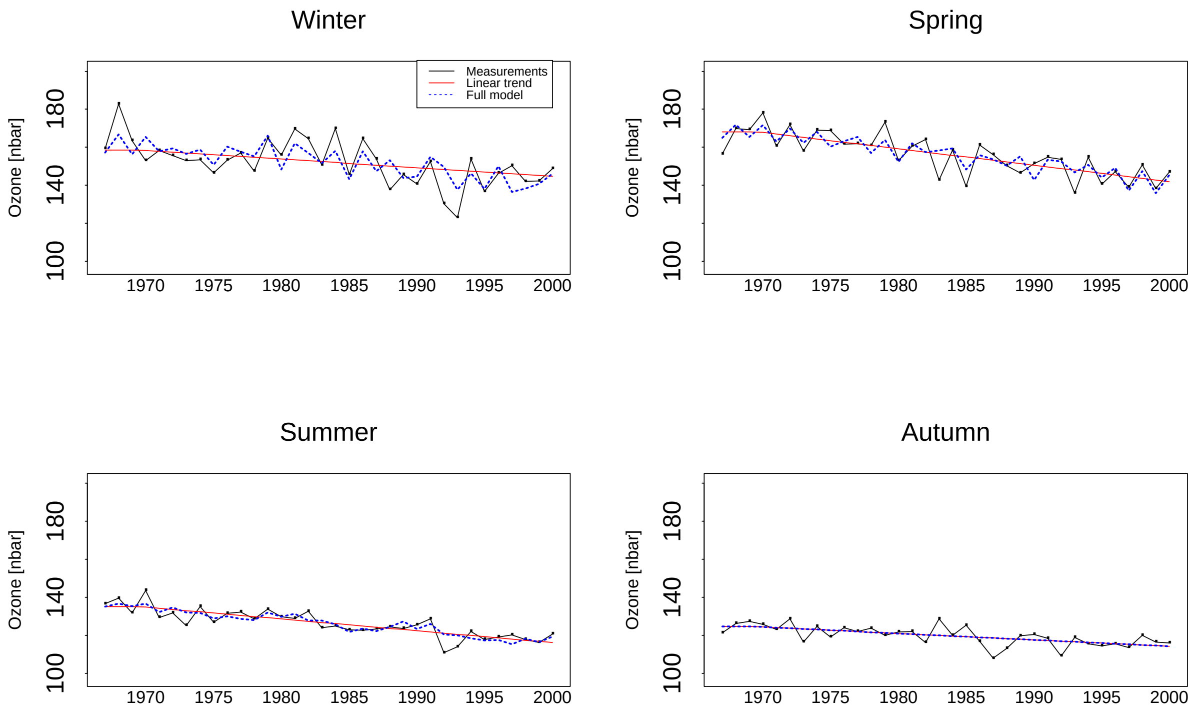

Figure 6.24: Same as Figure 6.22, but at the 50 hPa level (ozone maximum).

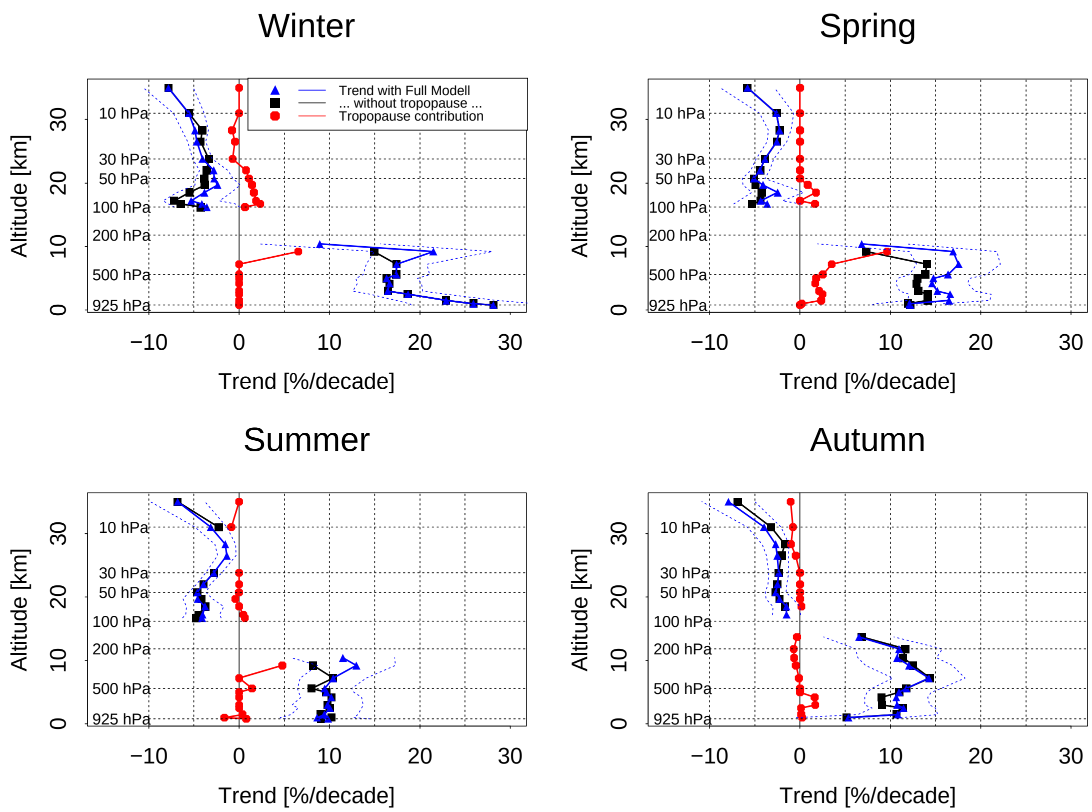

Figure 6.25: Ozone seasonal trends between 1970 and 2000. In blue, ozone trend with explanatory variables. In black, same but without the tropopause pressure contribution. In red, the tropopause pressure contribution alone.

Other explanatory variables, like the QBO and the solar flux, contribute less; the QBO explains less than 15% of the ozone variability in Winter at the height of the ozone maximum, which confirms earlier findings (Soukharev, 1997). The solar flux is significant at selected pressure levels and seasons, where most frequently the QBO is at contrary not significant.

The contribution of the tropopause variability to the seasonal ozone trend is best seen in Figure 6.25. Between 250 and 100 hPa, the tropopause pressure explains a large part of the ozone variability. In the computations performed for this analysis, the tropopause explains up to 30% of the ozone variability in Winter and 60% in Spring. These contributions should even be larger at the levels near and above the tropopause where the model detects no significant unexplained ozone trend. Weiss (2000) attributes about half of the winter and Spring lower stratospheric ozone trends over Switzerland to dynamic contributions.

The key results of this trend analysis with explanatory variables are summarised in the following table (Table 6.2).

Table 6.2: Annual and seasonal trends calculated with explanatory variables (run 2 with the stratospheric aerosol) at 25 pressure levels (P [hPa]).
T: Trend [% per decade], 2s: statistical uncertainty of the trend (two standard deviations)
R2: Explained variance [%]]
The full trend model delivers only significant trends.

P	Year			Winter			Spring			Summer			Autumn		
	T	2s	R2	T	2s	R2	T	2s	R2	T	2s	R2	T	2s	R2
925	12.3	3.9	55	28.1	9.1	54	12.2	4.2	51	9.8	4.7	51	12.3	-	0
900	11.8	3.5	60	25.9	7.0	68	12.1	3.8	61	8.6	4.1	50	11.8	5.8	19
850	13.8	3.2	70	22.9	5.5	68	16.5	4.5	65	9.4	3.7	57	13.8	4.4	45
800	13.1	2.8	74	18.7	4.2	71	16.6	4.5	65	10.1	3.5	51	13.1	3.5	59
700	12.2	3.0	68	16.4	3.9	69	15.2	4.2	65	9.8	3.9	44	12.2	3.6	53
650	12.3	2.7	73	16.7	3.6	73	14.6	4.0	66	10.2	3.4	53	12.3	3.5	54
600	12.2	2.6	73	16.3	3.4	74	14.8	4.0	66	9.7	3.1	56	12.2	3.5	55
500	12.7	2.8	73	17.4	3.7	75	16.4	4.3	67	9.5	3.0	55	12.7	3.8	54
400	13.6	2.8	74	17.4	3.4	78	17.5	4.6	66	10.4	3.6	53	13.6	4.0	64
300	13.0	3.8	61	21.5	6.5	59	16.9	4.8	70	13.0	4.3	62	13.0	4.8	62
250	6.0	4.3	36	8.9	6.7	40	6.8	4.9	70	11.5	5.7	66	6.0	4.4	74
200	-	-	33	-	-	55	-	-	43	-	-	51	-	4.8	72
150	-	-	0	-	-	31	-	-	10	-	-	12	-	3.9	53
100	-4.0	2.3	28	-3.6	3.6	52	-	-	27	-	-	13	-4.0	-	33
90	-4.5	2.0	40	-4.1	3.7	46	-3.6	3.7	33	-4.1	2.1	46	-4.5	-	42
80	-3.7	1.8	58	-5.3	3.1	54	-4.4	2.4	37	-4.0	1.9	48	-3.7	1.6	48
70	-3.2	2.0	50	-3.8	3.0	45	-2.5	2.9	49	-3.8	2.1	29	-3.2	1.5	30
60	-3.9	1.3	56	-2.4	2.5	50	-4.1	2.2	55	-4.6	1.6	51	-3.9	1.5	23
50	-4.1	0.9	77	-2.8	2.1	57	-5.1	1.3	73	-4.6	1.0	78	-4.1	1.1	41
40	-3.7	0.8	78	-2.8	1.6	66	-4.4	1.0	75	-4.0	0.8	79	-3.7	1.0	42
30	-3.4	0.6	82	-4.1	1.1	72	-3.9	1.0	71	-2.8	0.9	60	-3.4	1.1	36
20	-2.8	0.7	70	-4.7	1.0	75	-2.6	1.1	55	-1.4	1.0	36	-2.8	1.2	35
15	-2.8	1.0	62	-4.9	1.5	66	-2.3	1.3	47	-1.5	1.2	29	-2.8	1.5	38
10	-3.8	1.2	61	-5.6	1.7	65	-2.6	1.9	26	-3.1	1.8	28	-3.8	1.9	36
7	-7.1	2.1	59	-7.8	2.8	50	-5.8	3.0	32	-6.8	3.0	39	-7.1	2.9	48

As a conclusion related to the trends computed with seasonal means, the following points can be highlighted:

- Figure 6.26 summarises the profiles of the seasonal ozone trends after removal of the natural influences accounted for by the model. These seasonal trends correspond to the final estimates of the present report related to the long-term consequences of the anthropogenic release of ozone depleting substances. In the troposphere, the positive trends amount to approximately 10% per decade during Summer, ~ 12% during Autumn, whereas they generally reach more than 15% during Spring and Winter. In the stratosphere between 100 and 10 hPa, the trends are significantly negative and vary between -2% and -6% depending to the altitude and the season. Between 100 and 50 hPa, these negative trends are smaller during Autumn than during the other seasons. Between 30 and 10 hPa, the negative trends are larger during Winter.
- The combined or overall uncertainty, which adds instrument-drift uncertainty to the statistical uncertainty derived from the trend model, is indeed larger than the statistical uncertainty. However, most trend results remain statistically different from zero even when the overall uncertainty is considered.

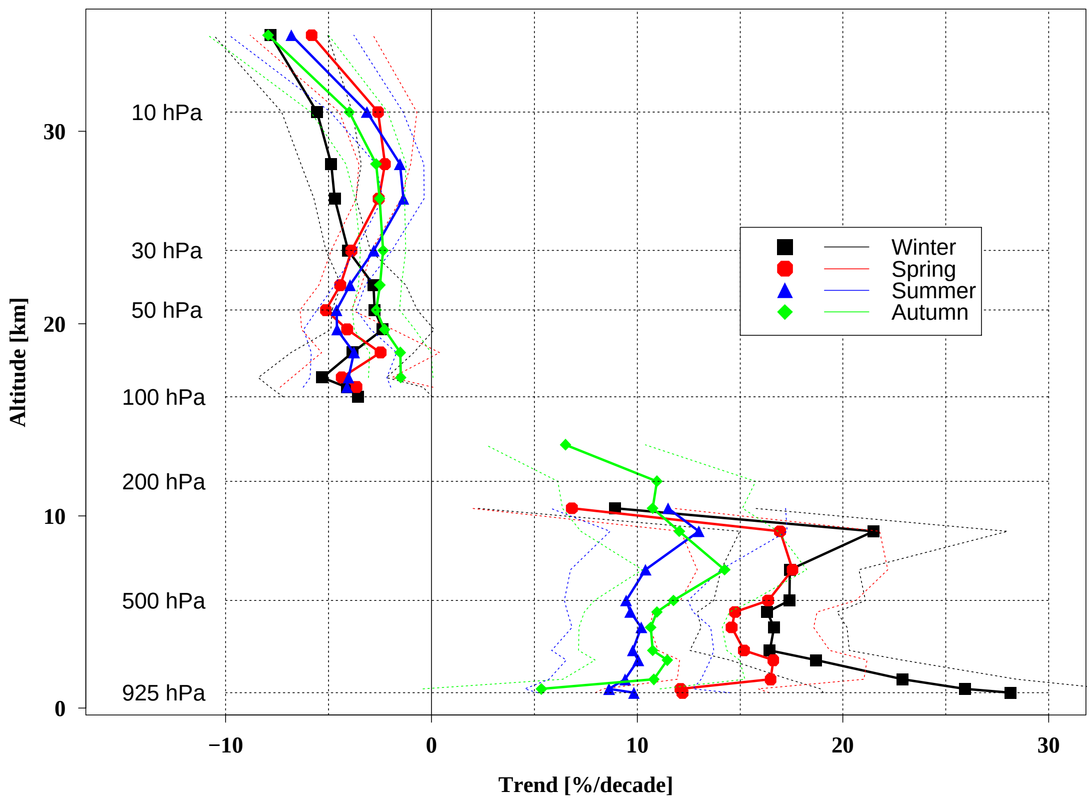

Figure 6.26: Seasonal unexplained ozone trends of the Payerne ozone soundings over the period 1970-2000 in percent per decade, using the normal dataset (0.9 < CF < 1.4). The trend contributions explained by different natural processes have been removed (run 1 of the full model with stratospheric aerosol). The dotted curves reproduce the statistical trend uncertainties (2σ). The full trend model delivers only significant trends. This figure presents the results of the four panels of Figure 6.19.

6.2.5 Monthly trends

The full model presented in chapter 6.2.1 also allows the computation of monthly ozone trends. The trends displayed in Figure 6.27 correspond to the trends observed after removal of the significant influence of different natural processes (tropopause pressure, volcanic stratospheric aerosol, solar flux cycle and Quasi-Biennial Oscillation). They are commonly attributed to the anthropogenic release of ozone depleting substances.

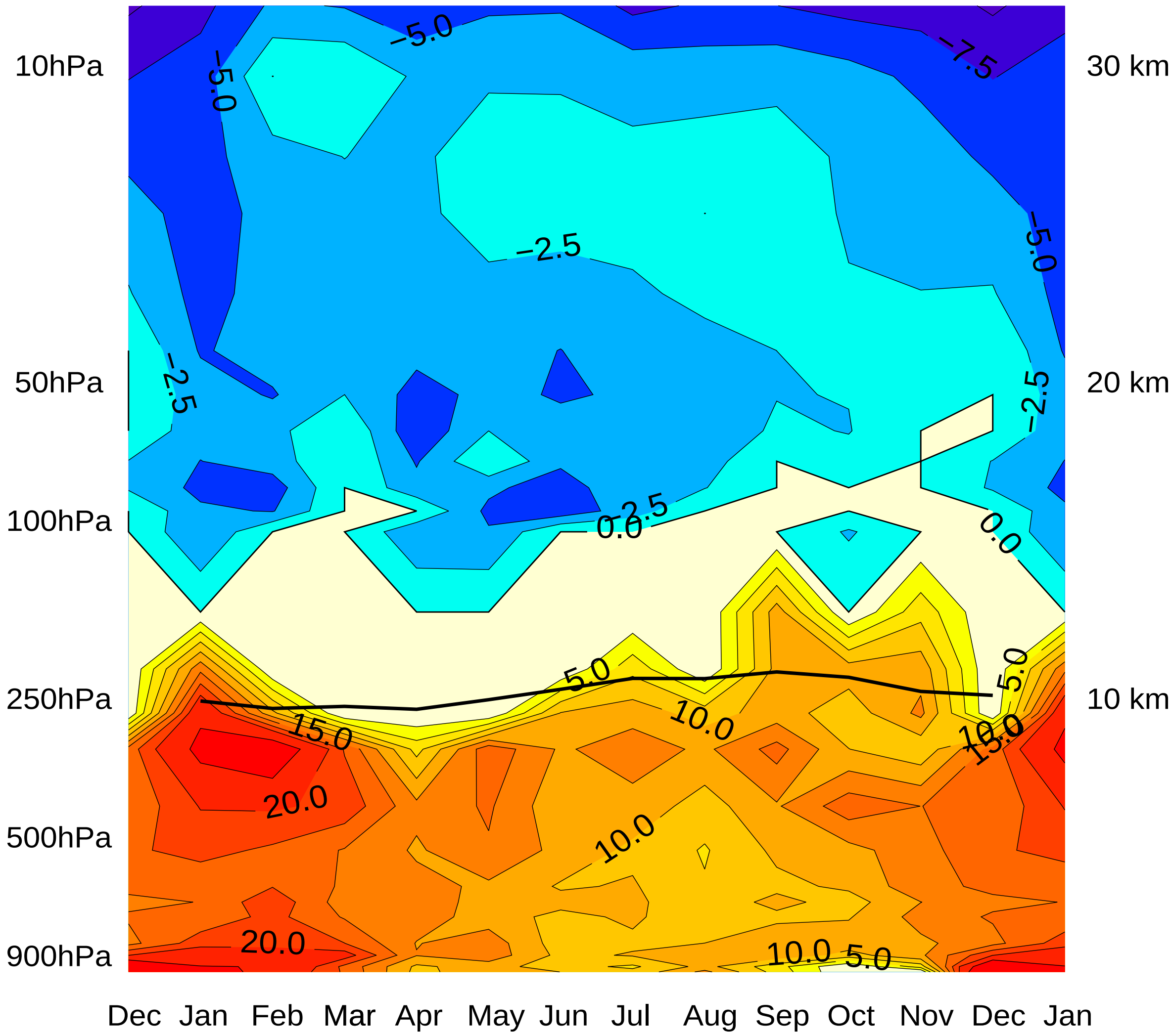

Figure 6.27: Isoplethes of ozone monthly trends between 1970 and 2000, computed for 25 pressure levels between 7 and 925 hPa of the normal dataset (0.9 < CF < 1.4) with the full trend model (run 2 with stratospheric aerosol) and expressed in % per decade. The positive values in warm colours, the negative values are in cold colours. The climatological tropopause altitude is represented by the black line around 10 km. Not significant trends are set to zero by the stepwise regression model and appear as light yellow on the figure (there are no significant trend value between -1.5 and +6% per decade).

As the stepwise regression model sets to zero not significant trend results, large areas of the following figures appear as light yellow. The unexplained trends remain very similar to the observed trends of Figure 6.6 at most levels of the profile, with one noteworthy exception. The variability and trend of the tropopause pressure explains a large part of the ozone variability in a thick region encompassing the lower stratosphere and the upper troposphere. As a consequence, the anthropogenic impact on a thick part of the lower stratosphere is no more statistically significant. The negative trend in the lower stratosphere during late Winter and early Spring in Figure 6.6 can hence be assigned to the tropopause variability. The annual and seasonal aggregations used for the annual and seasonal trend calculations noticeably damp the annual cycle in the tropopause region. Therefore, the monthly averaging helps the process analysis. According to Weiss (2000), another definition of the seasons even improves the seasonal statistical results.

The results of the other run with the ODF replacing the stratospheric aerosol are presented in Figure 6.28. There are minor differences with the previous Figure 6.26 in the lower troposphere during Summer, that could be due to statistical artefacts. The ODF explains however a large part of the ozone in the middle stratosphere. Some trends are less negative between 30 and 10 hPa from April to September.

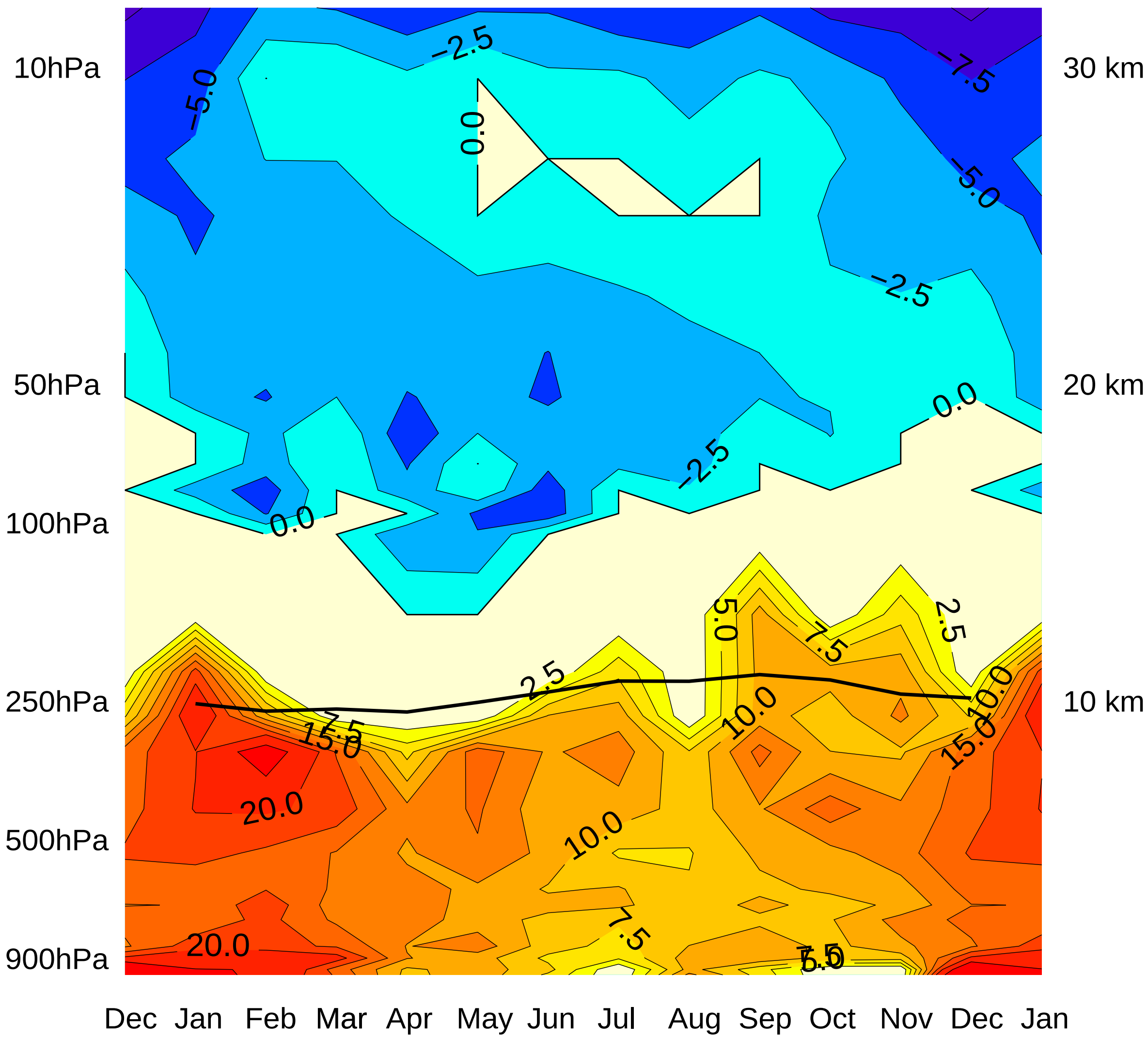

Figure 6.28: Same as Figure 6.27, but for run 1 with ODF (there are no significant trend values between -1.7 and +5% per decade).

Figure 6.29 exhibits the absolute trends corresponding to Figure 6.2.

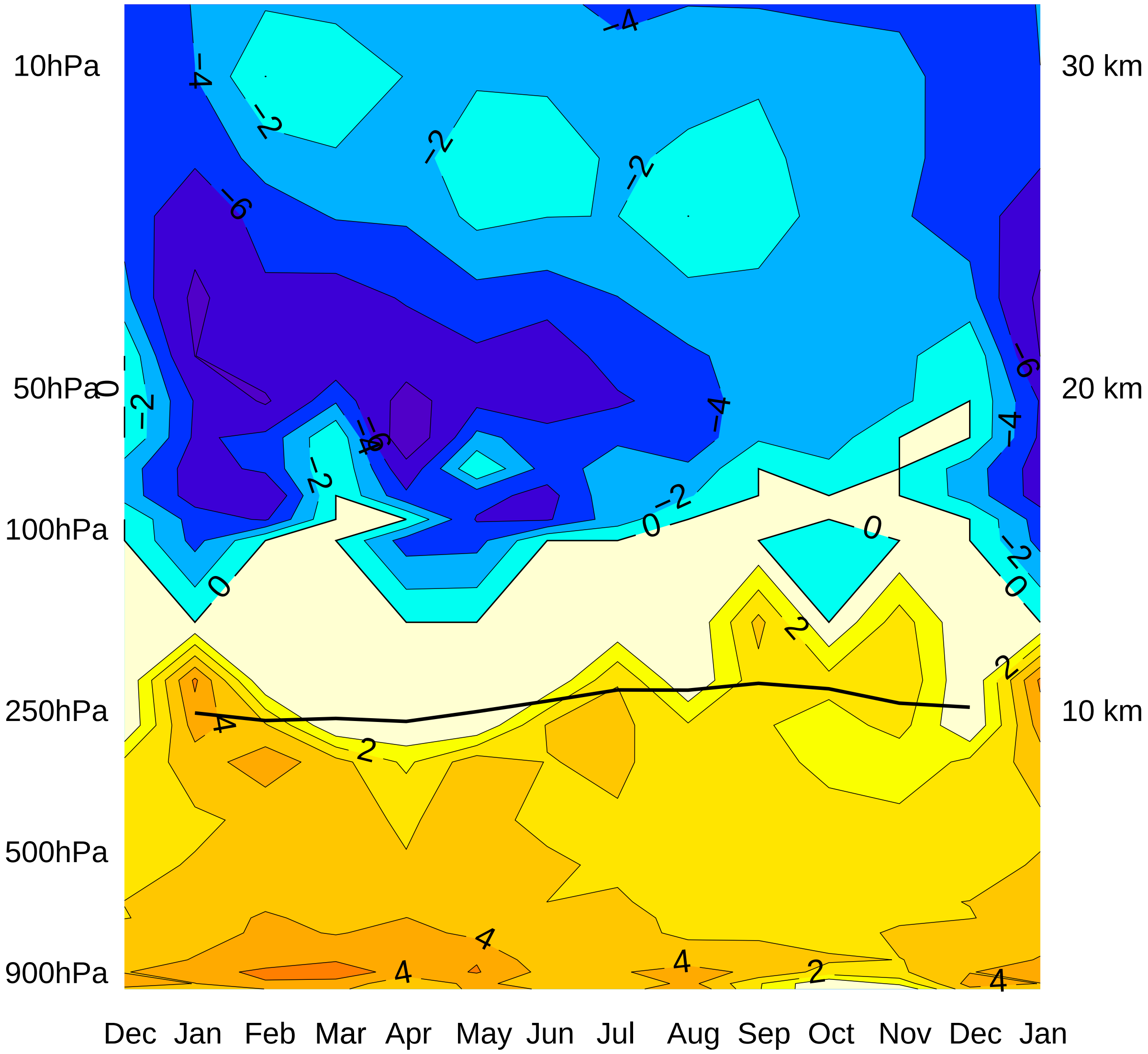

Figure 6.29: Same as Figure 6.27 (run 2 with stratospheric aerosol), but the trends are expressed in nbar per decade (there are no significant trend values between -1 and +1 nbar per decade.

The major result of this monthly analysis is the importance of the annual cycle of natural processes on the ozone trends. This is especially valid for the dynamical influences. Other explanatory variables would be necessary for a better trend (e.g. temperature at the 850 hPa level, SO_2 concentrations).

6.3 Special topic: launch time changes

Many changes affected the Payerne ozone balloon soundings launch time between 1968 and the beginning of 1982, when it was finally fixed to 11 UTC (see chapters 2, 3 and Figure 6.10). Several discontinuities appear in the lowest levels of the ozone series during this period and disturb significantly the trend calculation. Therefore, an additional check of the correction introduced in the Payerne dataset (see section 3.1.3) is presented below.

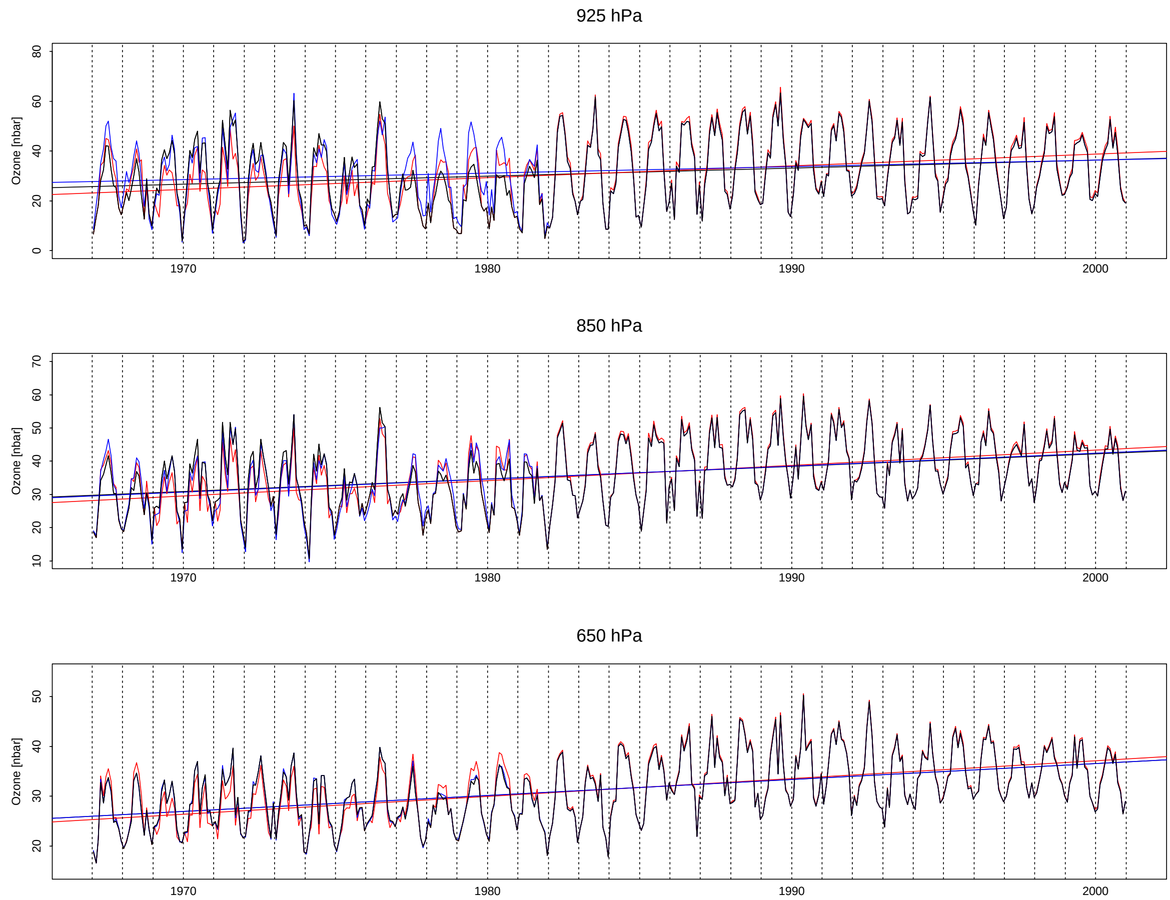

Figure 6.30: Corrections for launch time changes at 925, 850 and 650 hPa. The original series is represented by the black line, the new corrected series based on an additional explanatory variable in the trend model by the red line, the time series used in the present report corrected according to diurnal ozone cycles measured at surface stations between 490 and 3500 m asl in blue. Simple linear trends are also plotted.

An alternative correction for the pressure levels between 925 and 650 hPa can be built in the full trend model by the addition of a new explanatory variable representing the launch time difference to 11 UTC. This method delivers the contribution of launch time changes while simultaneously accounting for other contributions (tropopause pressure,...). The assumption is a linear function between the launch time difference to 11 UTC and the ozone deviation compared to this time. As 11 UTC falls into the morning increase of ozone and as the biggest time differences do not go beyond the diurnal minimum and maximum of ozone, this assumption can be considered as adequate in a first approximation. The effects are expected to decrease from the ground level up to 700 or 650 hPa and be marginal at higher levels. Meteorological situations with deep convections can influence the whole troposphere, but they mostly occur in the late afternoon. In fact, some contributions fit with the model up to the tropopause. As it can be seen in Figure 6.30, corrections are most important between 1977 and 1982 at the time of the biggest launch time changes. For comparison, the correction applied in this report is also presented in the figure.

In fact, between 1968 and 1977, the new correction is smaller for the summertime, except for values between 1977 and 1982. This correction tends to diminish (between 1968 and 1977) or to increase (between 1977 and 1982) the amplitude of the ozone annual variation. The correction reduces the ozone mean value between 1968 and 1977, but increases it between

1977 and 1982, when the biggest launch time changes occurred. This results in a larger general trend, as it can easily be seen in Figure 6.30. It is worth mentioning that the two methods have an opposite impact on the ozone trend at 925 hPa. For the other pressure levels, both methods influence trend in the same direction. At the 650 hPa level, the new method has a grater impact on the time series than the other one.

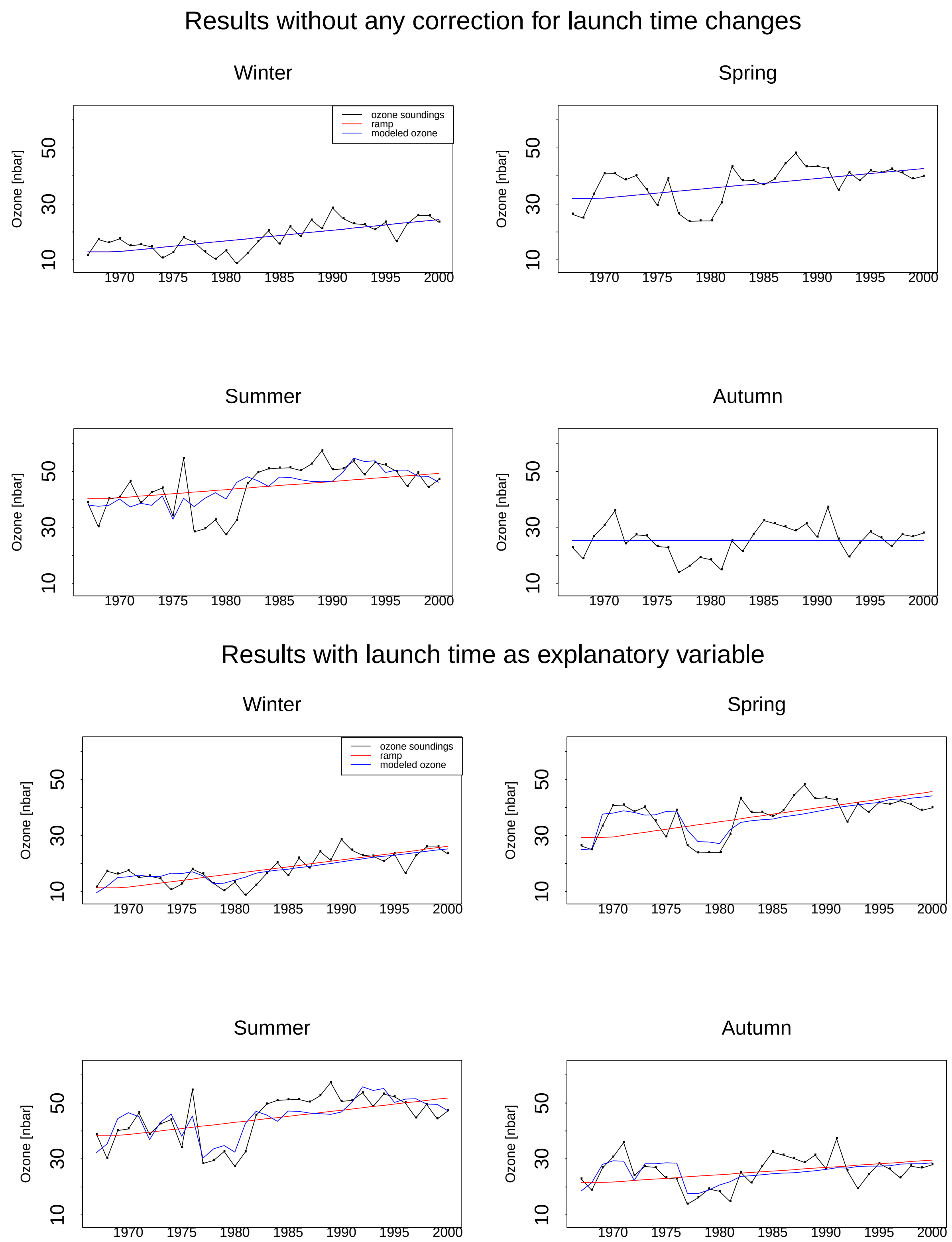

Figure 6.31: Seasonal fitted ozone time series (blue) versus measured ozone (black) and trendline (red) at 925 hPa for two different calculations.

A sensitivity analysis of the new correction shows that it does not affect the significance of other explanatory variables through compensation effects. Launch time changes bring significant contributions to the ozone concentrations in the lowest levels, but also unexpected ones above 500 hPa. In particular from February to May and from July to November significant contributions propagate into the upper troposphere. The correction has been restricted to 600 hPa.

Figure 6.31 confirms that the model is able to simulate the ozone discontinuities introduced by the launch time changes. A clear improvement is observed between 1977 and 1982, but slight corrections are also introduced for the previous years in all seasons. The improvements are most important in Summer when the ozone diurnal cycle is large.

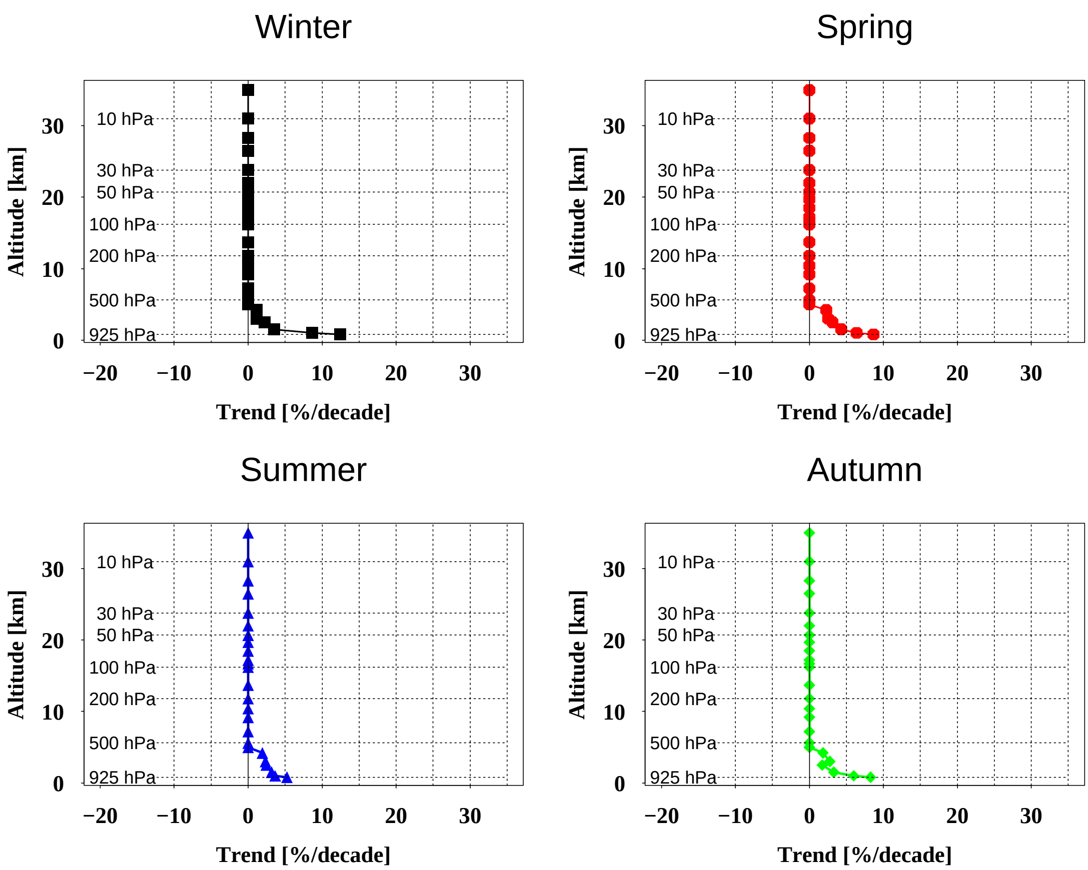

Fig 6.32: Differences of the seasonal ozone trends between calculations with the correction method based on an explanatory variable and without any correction for the launch time changes. Seasonal means calculated for both cases over the 25 pressure levels for the period 1970 - 2000. The trend differences are expressed in % per decade.

A comparison between Figure 6.31 and Figure 6.23 shows that the correction applied to the Payerne dataset before the trend calculations of this chapter 6 is appropriate and delivers a good fit as well. The two types of correction induce different trend values for the lowest levels. The last method (explanatory variable) even increases the trend values, regardless of the month and pressure levels. A sensitivity analysis has been performed by comparing the trends computed with and without a launch time correction. The impact of a launch time change can be seen in Figure 6.32, where differences between seasonal trends are plotted. For all seasons, differences ranging from +2% to +7% are observed for the lowest pressure levels. In Winter, at 925 hPa, the trend calculated with the new correction shows an increase of 13% in comparison with the trend calculated with the original series, and consequently amounts to more than 40% per decade. Considering the measurement uncertainties in the low troposphere, such trend values have to be considered very carefully. The correction issued from surface ozone stations operates in the same direction (reduction of the ozone annual cycle between 1968 and 1977, and increase of the ozone annual cycle between 1977 and 1982), but has a smaller impact on trend calculations than the new one.

It is worth mentioning that the statistical correction introducing the launch time differences as an explanatory variable does not exclude interferences with other parameters.

The following conclusions can be drawn:

- The launch time changes deeply affect trend estimates at the lowest pressure levels.
- Differences up to approximately 10% per decade have been found at 925 hPa (correction - no correction), decreasing rapidly with altitude.
- The correction based on surface ozone stations used in this report provides reasonable results, but it is not possible to judge if it is better than the other one introduced in this section.

6.4 Summary of the trend analysis

Two datasets of the Payerne ozone soundings have been first analysed with a **simple trend model without explanatory variables**. The first dataset includes all soundings with a correction factor between 0.9 and 1.4. The latter requires a higher data quality (correction factor between 0.95 and 1.25), but significantly reduces the number of soundings during the first 15 years of the full period 1967-2000. Main results are as follows:

- The annual trends derived from the two datasets are significantly different from zero and fairly consistent outside a thick atmospheric layer in the low stratosphere.
- This analysis of the seasonal trends shows that trends calculated with the normal dataset are more consistent in the upper troposphere and lowest stratosphere than trends derived from the reduced dataset. In the lower and middle troposphere, as well as in the middle stratosphere, both datasets provide rather similar trends and it is difficult to discriminate them.
- The analysis of monthly trends shows that the change from a negative to a positive trend is sharpest in late Winter, and located at the tropopause. This change does not follow the tropopause for the whole year, but can be better identified using the climatological 40-50 hPa ozone isolines.

A **full trend model with atmospheric explanatory variables** has then been used, in order to exclude the influences of the variability of natural processes or in order to analyse the influence of specific processes. In the present study, the stratospheric aerosol content has been included in the natural factors, and according to a common practice, the trends calculated with the model involving only natural explanatory variables are considered as long-term consequences of the anthropogenic release of ozone depleting substances. Due to some drawbacks of the reduced dataset (higher quality, but reduced statistical representativity), the full trend model has been run mainly with the normal dataset.

Figure 6.17 presents the annual trends and Figure 6.20 the ozone variance explained by the full model. The main results related to these trends are as follows:

- The full model with explanatory variables delivers only slightly changed annual ozone trends compared to the simple trend model without any explanatory variable.

- The tropopause pressure is the explanatory variable that most significantly influences the trend estimates. It strongly modulates the trend results under and above its mean altitude.
- When considering the ozone depletion factor (ODF) in replacement of the stratospheric aerosol, the negative trends in the middle stratosphere are slightly reduced.
- The combination of the natural variables, including the stratospheric aerosol load, or of the natural variables and ODF can explain the apparent trend decrease since the mid 1990’s at the 50 hPa level, as well as at most stratospheric levels.
- The comparison with the results of the SPARC/IO$_3$C/GAW assessment group shows a fair agreement with the results of the Harvard university (different time periods).
- The agreement with the tropospheric trends of Hohenpeissenberg is better than with the previous dataset submitted to the SPARC/IO$_3$C/GAW assessment group.

Figure 6.26 and Table 6.2 summarise the profiles of the seasonal ozone trends after removal of the natural influences accounted for by the model. The main results related to these trends are as follows:

- In the stratosphere between 100 and 10 hPa, the trends are significantly negative and vary between -2% and -6% depending to the altitude and the season. Between 100 and 50 hPa, these negative trends are smaller during Autumn than during the other seasons. Between 30 and 10 hPa, the negative trends are larger during Winter.
- In the troposphere, the positive trends amount to approximately 10% per decade during Summer, ~ 12% during Autumn, whereas they generally reach more than 15% during Spring and Winter.
- The positive tropospheric ozone trend remains somewhat larger than at other European stations (WMO, 1998), in particular for Winter in the lowest troposphere (925 hPa - 800 hPa). Results have to be interpreted there with caution, especially due to measurement interferences with SO_2.
- This report does not handle trend changes during sub-periods of the 34 year period (1967 - 2000). However, the linear trend model with atmospheric explanatory variables still accounts for the stratospheric ozone evolution between 1995 and 2000.
- The variability of the Payerne ozone sounding’s time series can be largely attributed to dynamic processes. In particular, the tropopause pressure is the main natural cause of ozone variability in the lower stratosphere and in the upper troposphere during Winter and Spring. Furthermore, the annual cycle of the tropopause pressure (or altitude) long-term trend seems to explain some part of the annual cycle of the ozone trend.
- The ozone depletion factor (ODF) and, to a smaller extent, the solar flux and the QBO have less importance in the ozone variability and trend.
- The model does not reproduce well the tropospheric ozone measurements during the 80’s, where measurement anomalies have been detected in the Payerne dataset.

The monthly analysis performed stresses the importance of the annual cycle of natural processes on ozone trends, especially of the dynamical influences. Other explanatory variables would be helpful for a better trend analysis in the troposphere.

Launch time changes deeply affect trend estimates in the planetary boundary layer. Related results can be found in the previous section.

7. CONCLUSION

The present report concludes a project started in 1995 which has been devoted to the historical documentation, the re-evaluation, the homogeneity check as well as the trend analysis of the Payerne ozone soundings. The Payerne time series is one of the longest in the world and this report covers its whole period from the beginning of the measurements (end of 1966) up to the end of 2000.

The historical documentation of the ozone balloon soundings, the different reprocessing and re-evaluation procedures, as well as the homogeneity analysis have permitted an appropriate assessment of the homogeneity of the Payerne ozone dataset summarised in chapter 4.3. Clearly justified corrections have been made in order to improve the quality and homogeneity of the dataset. Periods, in which the data quality is still doubtful, have been identified.

Because of the lack of information on the many important details in the pre-launch procedures applied in the past at Payerne, further efforts in the re-evaluation are expected to provide only small improvements in the quality of the series. Indeed, the Brewer-Mast ozone sonde is very sensitive to many details of the operating procedure and new experiments are not able to reproduce the past conditions.

Since the beginning of this project, a noteworthy effort has also been devoted to the improvement of the operating procedure of the Brewer-Mast sonde at Payerne. As a result, the measurement quality has improved to become as high as technically possible since the end of 1993. In addition, there are still ongoing experimental studies in order to assess the comparability of this sonde with the ECC sonde prior to the replacement of the Brewer-Mast sonde by the more precise ECC sonde in Autumn 2002 (Stübi et al., 2002).

The results of this new trend analysis are summarized in the previous chapter 6.4. This trend evaluation represents an update of different analyses performed in the past years, especially by the SPARC/IO$_3$C/GAW ozone assessment group (WMO, 1998; Logan et al., 1999). It relies also on the methods and results of a doctoral thesis at the IACETH (Weiss, 2000). Not only trends have been computed, but their uncertainty has been also taken into account.

Stratospheric chlorine concentrations are no longer increasing in the stratosphere due to the international process of the Montreal Protocol and its amendments. The expected ozone recovery depends upon the correctness of scientific assumptions about ozone depletion, and upon the full implementation of the Protocol and its amendments (Wardle et al., 1997). Because of the very long atmospheric lifetimes of the ozone depleting substances, the ozone recovery is expected to proceed very slowly. Furthermore, ozone is a strong greenhouse gas, in particular in the tropopause region, which requires careful long-term monitoring also in the future.

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- http://www.wmo.ch
- http://msc-smc.ec.gc.ca/woudc
- http://www.meteosuisse.ch
- intro : http://www.epa.gov/ozone/science/process.htm

9 INTERNET DATA SOURCES

Tropopause pressure from NCEP-reanalysis:
- http://www.cdc.noaa.gov/ncep_reanalysis

Solar radio flux at 10.7 cm flux:
- ftp://ftp.ngdc.noaa.gov/STP/SOLAR_DATA/

Quasi-Biennial Oscillation (QBO):
- http://nic.fb4.noaa.gov/data/cddb

North Atlantic Oscillation index (NAO):
- from NCEP-reanalysis : http://www.cdc.noaa.gov/ncep_reanalysis
- measured : http://www.cgd.ucar.edu/cas/climind/nao_monthly.html

Artic Oscillation index (AO):
- http://tao.atmos.washington.edu/data/ao

Ozone Depletion Factor (ODF):
- http://www.giss.nasa.gov/data/

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