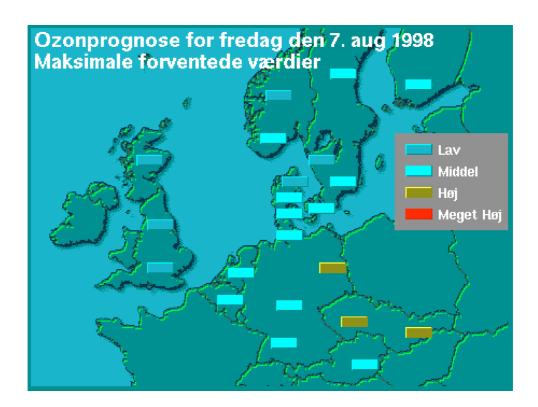
DANISH METEOROLOGICAL INSTITUTE

— TECHNICAL REPORT ———

00-05

Validation of DACFOS Surface Ozone Forecasts 1996-98 - Description of the new Verification System and Model Improvement

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1 INTRODUCTION

1 INTRODUCTION

DMI has developed an operational forecasting system for ozone concentrations in Europe. The system is based on the coupling of a chemical model (EMEP MSC-W's oxidant model) [Simpson, D.(1993)] and DMI's 3D lagrangian transport model. The forecast system utilizes analysis and forecast data from the numerical weather prediction model, Danish Meteorological Institute's HIgh Resolution Limited Area Model (DMI-HIRLAM). The whole system is called Danish Atmospheric Chemistry FOrecasting System (DAC-FOS) [Jensen, M.H.(1996)].

This report is a status report for DACFOS (Danish Atmospheric Chemistry FOrecast System) ver 2.0 and will highlight some of the changes from the first version until now, such as

- implementation of the system as an operational system
- collection of ozone data from all over Europe from the internet for the verification and from the Kalman filtering
- creation of a public information system on the DMI internet web-page
- implementation of some modifications concerning the handling of the trajectories, emissions, mixing height and land-sea mask
- semi-operational verification of the ozone forecasts for the period April 1996 to December 1998

DACFOS has been running operationally since June 1997, most of the ozone data collection from the internet were initiated in spring 1998 and the public information system was on the internet from July 1998. The verification is running semi-operational every month for 9 stations in Denmark, Sweden and England.

Verification of ozone forecasting systems has become an even more important subject in connection with the real-time information of the ozone forecasts to the public. False or no alarms of ozone threshold exceedances should be avoided.

The European Environment Agency (EEA) has initiated a work for harmonizing validation of ozone forecasts and ozone data exchange. In a report from a Technical Working Group on Ozone Forecasting and Data Exchange (TWG-DFO) [Van Aalst, R.M.(1998)] several statistical parameters are taken into account, mostly those concerning the skill to forecast exceedances of the EU public information ozone threshold at 180 $\mu g/m^3$.

This work will also deal with the verification suggested by EEA, except for a modification caused by the few numbers of exceedances of the EU public information threshold in Denmark.

2 THE DACFOS SETUP

2 THE DACFOS SETUP

DACFOS is a photochemical backtrajectory model system. DMI's 3-D transport model can utilize meteorological data from the different versions of DMI-HIRLAM and data from ECMWF's global model.

2.1 THE OPERATIONAL SETUP FOR DACFOS ver. 2.0

The system is set up to make 48 hours forecasts, automatically four times a day, for selected receptor points within the EMEP-grid covering all of Europe. For all receptor points, five backward trajectories arriving at equidistantly distributed heights within the atmospheric boundary layer [Sørensen, J.H.(1996)] are calculated hourly, meteorological parameters needed for the chemistry model are also provided. For each trajectory the concentrations of the chemical species are calculated independent of the other trajectories, assuming total mixing in the boundary layer. Calculations are made for 70 chemical species and 130 chemical reactions using a quasi-steady state approximation (QSSA) numerical method [Simpson, D.(1992)].

The output from the chemical module are Kalman filtered [Chenevez, J. (1998)] for the stations with real-time ozone observations available. The forecast will not be able to describe local variations, partly because the emission data are defined on a coarse grid (at present 50 km by 50 km).

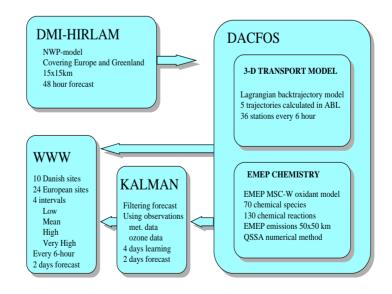


Figure 1: The DACFOS system summer 1998.

2 THE DACFOS SETUP

2.2 FORECAST STATIONS

The number of forecasted stations has increased during the development period. The forecast system was launched in June 1995 with Jægersborg as the only station. 24 EMEP stations were added in 1996, and in 1997 seven Danish stations and two German/Austrian stations completed an overall coverage of Denmark and Europe. Later on in 1998 Paris was added as a result of several ozone exceedances in France in the summer 1998. At the same time Warszawa was included.

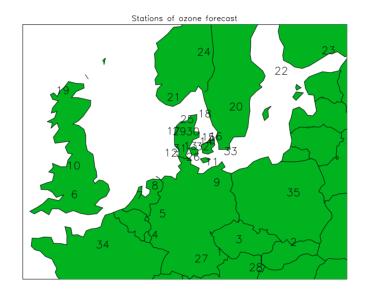


Figure 2: DACFOS stations in Europe.

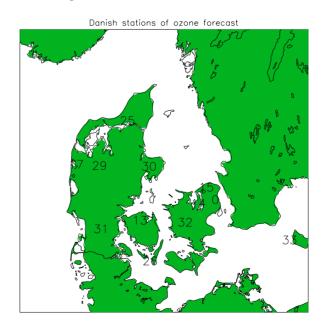


Figure 3: DACFOS stations in Denmark.

2 THE DACFOS SETUP

# at map	lat	lon	height	EMEP code	Name	Start of forecast
0	55.760	12.530	40		Jægersborg	23 June 1995
1	55.760	13.219	1016	DE05	Brotjacklriegel	30 May 1996
2	55.760	20.283	-1	SK04	Stara-Lesna	-
3	55.760	15.083	633	CS03	Kosetice	_
4	55.760	7.052	480	DE04	Deuselbach	_
5	51.117	7.633	510	DE14	Meinerzhagen	_
6	51.567	-1.317	137	GB36	Harwell	_
7	52.117	5.200	420	NL08	Bilthoven	-
8	52.817	6.667	16	NL02	Witteveen	_
9	53.167	13.033	65	DE07	Neuglobsow	-
10	53.333	-1.750	420	GB37	Ladybower	-
11	54.433	12.733	1	DE09	Zingst	-
12	54.926	8.310	12	DE01	Westerland	-
13	55.400	10.390	17		Odense	-
14	55.700	12.100			Lille Valby	-
15	55.967	12.333	10	DK32	Frederiksborg	-
16	56.017	13.150	175	SE11	Vavihill	-
17	56.283	8.433	10	DK31	Ulborg	-
18	57.417	11.933	10	SE02	Rorvik	-
19	57.733	-4.783	270	GB15	Strath Vaich	-
20	57.817	15.567	261	SE32	Norra Kvill	-
21	58.383	8.2500	190	NO01	Birkenes	-
22	59.783	21.383	2	FI09	Uto	-
23	60.517	27.683	8	FI17	Virolathi	-
24	61.250	11.783	440	NO41	Osen	-
25	57.050	9.920	3		Aalborg	28 Aug. 1997
26	54.730	10.720	2		Keldsnor	-
27	48.410	11.730	453		Weihenstephan	-
28	47.770	16.770	183		Illmitz	-
29	56.283	9.1330	52		Karup	-
30	56.300	10.617	23		Tirstrup	-
31	55.233	9.267	43		Skrydstrup	-
32	55.383	11.717	35		Tyvelse	-
33	55.067	14.750	16		Rønne	-
34	48.800	2.400	89		Paris	10 Aug.1998
35	52.200	21.000	106		Warszawa	-

Table 1: Stations of DACFOS ozone forecast.

3 REAL-TIME OZONE OBSERVATIONS

3 REAL-TIME OZONE OBSERVATIONS

Available ozone measurements on hourly real-time basis are collected from Denmark, England, Sweden, Austria, Belgium and Germany from the internet. The stations are all non-urban sites, see table 2.

DMI has been measuring surface ozone in Jægersborg since 1995. An ultraviolet absorption photometric analyzer placed at the WMO station number 06181, Jægersborg $(12^{\circ}32' \text{ E and } 55^{\circ}46' \text{ N})$ located about 7 km north of Copenhagen, is used for continuous measurements of ozone. The sampler is placed 2 meters over ground level and the data are averaged over 10 minutes. Observations and modelled concentrations can be seen in the Appendix.

NERI (the National Environmental Research Institute) is providing hourly measurements of surface ozone from two of their stations, Lille Valby $(12^{\circ}06' \text{ E } 55^{\circ}42' \text{ N})$ and Keldsnor $(10^{\circ}43'\text{E} \text{ and } 57^{\circ}44' \text{ N})$.

The British ozone measurements are provided by AEA-Technology for many years back for the three stations Harwell (1°19′ E 51°34′ N) and Ladybower (1°45′ E 53°20′N) and Strath Vaich (4°47′W and 57°44′ N), for which DACFOS has been forecasting ozone from June 1997.

The Swedish ozone measurements are provided by IVL on hourly basis in the day-hours. These data have been collected for the three stations forecasted by DACFOS, Rorvik $(11^{\circ}56' \text{ E and } 57^{\circ}25' \text{ N})$, Vavihill $(13^{\circ}09' \text{ E and } 56^{\circ}01' \text{ N})$ and Norra Kvill $(15^{\circ}34' \text{ E and } 57^{\circ}49' \text{ N})$, since 12 June 1997.

Measurements from the Austrian station, Illmitz (16°46′ E and 47°46′ N), have been collected since August 1997.

No measurements are available at the internet for the stations forecasted by DACFOS in Germany, but other measurements situated nearby have been collected since the start of 1998.

The method of collecting data, by downloading data every hour from the internet, is a very time consuming and uncertain method, because changes of the layout of ozone web pages and periods of breakdowns in the providings occurs as well as a constant open window of netscape at a workstation is required.

A difficult quality check of the data is also required as most of the data collected directly from the internet have not been quality checked before publication. In some years ahead, a European ozone data exchange project will be initiated but probably only dealing with exchange of the daily maxima of ozone, which does not give the opportunity to make a thorough verification.

lat	lon	height	Name	Match DACFOS st.	Start of retrieval	Inst.
55.760	12.530	40	Jægersborg	Jægersborg	23 June 1995	DMI
55.700	12.100	~0	Lille Valby	Lille Valby	Jan. 1998	NERI
54.730	10.720	2	Keldsnor	Keldsnor	-	-
51.567	-1.317	137	Harwell	Harwell	Jan. 1997	AEA
53.333	-1.750	420	Ladybower	Ladybower	-	-
57.733	-4.783	270	Strath Vaich	Strath Vaich	-	-
56.017	13.150	175	Vavihill	Vavihill	12 June 1997	IVL
57.417	11.933	10	Rorvik	Rorvik	-	-
57.817	15.567	261	Norra Kvill	Norra Kvill	-	-
58.540	17.350		Aspvreten		-	-
64.110	19.450		Vindeln		-	-
55.350	13.000		Malmœ		-	-
52.817	06.667		Emsland	Witteveen	Feb. 1998	NRW
51.117	07.633		Finnentrop	Meinerzhagen	May 1998	NLO
48.410	11.730			Weihenstephan		
47.770	16.770	183	Illmitz	Illmitz	Aug. 1997	UBAVIE

Table 2: List over stations where surface-ozone observations are retrieved from the internet.



Stations of observed ozone 1997

Figure 4: Stations in Europe where hourly ozone observations are retrieved at DMI for DACFOS verification.

4 THE PUBLIC INFORMATION SYSTEM

In the summer 1998 an information system was set up for the internet (WWW.dmi.dk) at DMI. Ozone forecasts are shown for areas in the Northern Europe every 6 hour, with a window for the levels of ozone today and one for the levels of ozone tomorrow. The levels are provided as in table 3.

The "Vis graf" buttom gives the opportunity to see a graphical presentation of the time series of ozone at any of the areas shown on the map 48 hours ahead with hourly values. The areas presented as blocks on the map do not represent the single stations forecasted in DACFOS.

ozone interval ($\mu g/m^3$)	Danish	English
0-60	Lav	Low
60-120	Middel	Mean
120-180	Høj	High
180-∞	Meget Høj	Very High

Table 3: Ozone intervals used in the public information system.

Ozonprognoser Prognose for atmosfærens ozonindhold ved jordoverfladen Ozonprognose for fredag den 7. aug 1998 Finland Midttyskland Maksimale forventede værdier Østtyskland Nordtyskland Sydtyskland Tjekkiet Slovakiet Lav Middel Høj Meget Høj Vis graf Nordeuropa -7. aug 1998 —

Figure 5: A presentation of the DACFOS forecast from 6 August 1998 for the next day. The Danish text on the map says 'Ozone prognoses for Friday 7 Aug 1998. Expected maximum values.'

5 MODIFICATION OF DACFOS 1.0 TO 2.0

A description of DACFOS ver. 1.0 is given in [Jensen, M.H.(1996)].

A few essential modifications have been implemented in ver. 2.0; a revised number of trajectories for each receptor point, a new method of uptake of the emissions and use of a minimum height of the mixing height are applied. In addition a new land-sea mask has been tested.

5.1 NUMBER AND WEIGHT OF TRAJECTORIES

In DACFOS several backward trajectories are calculated, for each receptor point, arriving at equidistantly distributed heights within the atmospheric boundary layer, the chemical species are calculated independent of the other trajectories, assuming total mixing in the boundary layer. In the early semi-operational mode of DACFOS the number of trajectories was 10, and the mean was used as the final concentrations of the species.

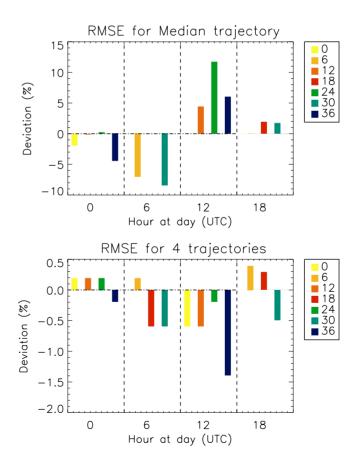


Figure 6: Sensitivity test of number and weight of trajectories. The upper diagram shows the use of a median trajectory and the lower the use of averaging over trajectory 1,4,7 and 10. The Y-axis shows the deviation of the RMSE compared with an average over 10 trajectories, the x-axis the forecast hour and the legend the forecast lengths. Positive deviation means lower RMSE, i.e. improvement.

5 MODIFICATION OF DACFOS 1.0 TO 2.0

In a sensitivity test a lower number of trajectories and use of the median was tested. The sensitivity test was run over a 5 months period from 1/4-1996 with Jægersborg as a receptor point with different use of the results from each of the trajectories. In the test runs $150x150 \text{ km}^2$ emissions from EMEP 1991 without any smoothing are used. The blockdiagram presentations (fig. 6) show the root mean square error (RMSE) in percent of deviation relative to the use of 10 vertical equidistant distributed trajectories equally weighted.

The top figure of RMSE are shown for tests with the median trajectory results used. The results varies quite substantial for the different start hours, with an improvement of the forecasts with origo at 12 hour, but showing less good results at 0 and 6 hour.

The lowest graph shows the results when using an average of 4 trajectories (1, 4, 7 and 10) instead of using 10 trajectories. The impact of this change did not show any substantial difference as the changes on the RMSE are very low.

The use of the median showed varying and not predominant good results, so it is not used in the operational forecasts. As it is possible to save a lot of computertime and space using fewer trajectories without getting less good results, only 5 trajectories have been used in the operational runs.

5.2 EMISSIONS

The emissions used are from EMEP/MSC-W (Co-operative program for monitoring and evaluation of the long range transmission of air pollutants in Europe. Meteorological Synthesizing Centre - West) [Tuovinen, J.(1994)], the work with the inventories are time consuming so the emissions are always several years old when they are available for the operational forecast of ozone. In the semi-operational runs emissions were defined on a grid at 150x150 km which is rather coarse so a refined emission inventory at 50x50 km was sensitivity tested together with some smoothing methods.

The sensitivity tests was run for a 5 months period from 1/4-1996 with Jægersborg as a receptor point. In the test runs 10 vertical equidistant distributed trajectories weighted equally were used. The blockdiagram presentations (fig. 7) show the root mean square error (RMSE) in percent of deviation relative to the 150x150 km emissions from EMEP 1991 without any smoothing.

In the first test a 50x50 km emission inventory from EMEP 1994 was used without any

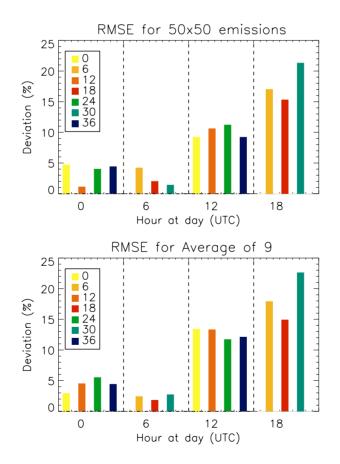


Figure 7: Sensitivity test of different emission. The upper diagram shows the use of 50x50 km emissions and the lower the use of averaging over 9 neighboring grids. The Y-axis shows the deviation of the RMSE compared with the 150x150 km emissions from EMEP 1991 without any smoothing, the x-axis the forecast hour and the legend the forecast lengths. Positive deviation means lower RMSE, i.e. improvement.

smoothing. The results, RMSE (top figure in fig. 7) and bias (not shown)), showed a general improvement for all forecast lengths by using the refined emissions.

In the second test a smoothing was performed on a 50x50 km grid. Every grid was averaged with the 8 neighboring grids. In this test an even better improvement was seen in most cases especially for the long forecasts. Other smoothing methods like cubic splines have been tested without success. The 9 grid averaging method has been implemented in the operational DACFOS in May 1997.

5.3 MIXING HIGHT

A study of the development of mixing height, temperature and the chemical species along a trajectory are shown for two cases.

In the first case, without a lower limit for the mixing height fig. 8, an explosive development of NO and SO2 is seen when the mixing heights are low, around step 330-370. This is followed by a high ozone development with a maximum at 155 ppb around step 400. This explosive development can be due to the fact that DACFOS distributes the emission uptake in the mixing layer defined by the mixing height. When the mixing height is very low, heigh concentrations can occur.

In the second case, with a lower limit for the mixing height, fig. 9, NO and SO2 did not develop as explosive as in the first case and the following ozone maximum is around 115 ppb, which is 33 % lower and much closer to the observed value at 60 ppb.

In fig. 11 compared to fig. 10, the impact of the use of a lower limit at the mixing height are seen for a specific point and period, Harwell in August 4-22 1997, where a lot of unrealistic high ozone peaks showed up. By implementing the mixing height limit this problem was partly eliminated, especially in the period August 16-18 1997. This correction is implemented in the calculation of the trajectories and used in the operational DACFOS since September 1997.

5 MODIFICATION OF DACFOS 1.0 TO 2.0

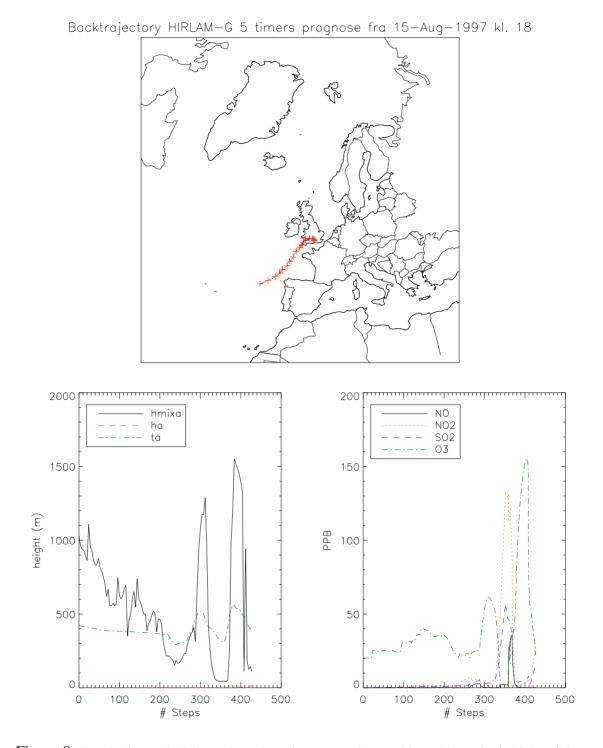


Figure 8: Results from calculations along the trajectory are shown without a lower limit of the mixing height. *hmixa* is the mixing height, *ha* is the actual height of the trajectory and *ta* are the temperature at the actual height of the trajectory (not in scale). Every step is 15 min. The trajectories shown in these cases are the lowest possible and are following the surface close in these examples.

5 MODIFICATION OF DACFOS 1.0 TO 2.0

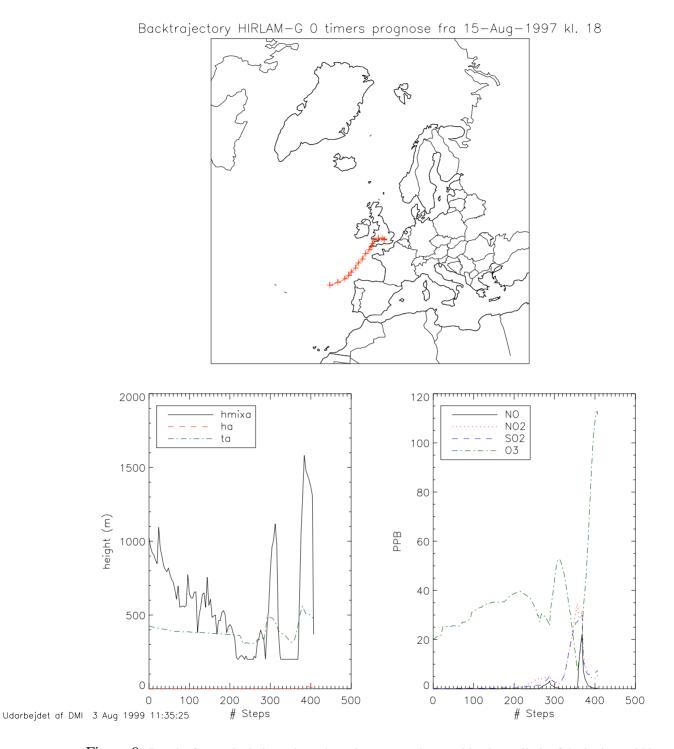
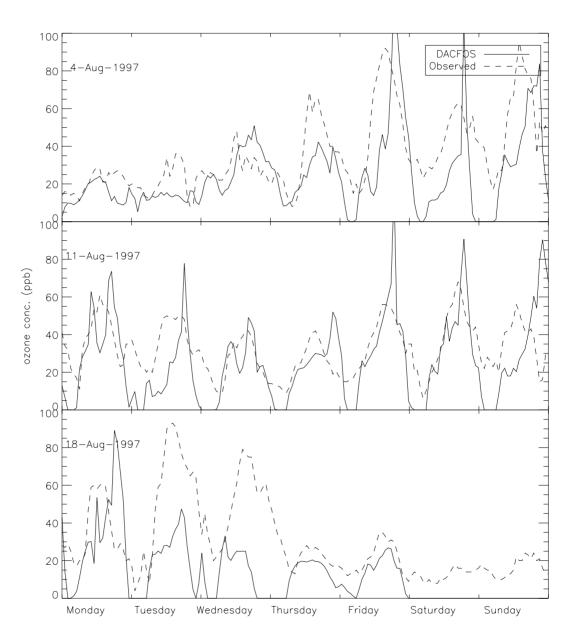
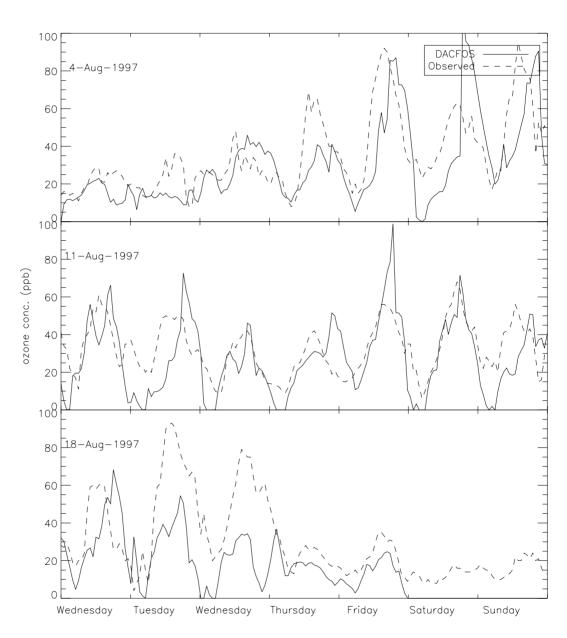


Figure 9: Results from calculations along the trajectory are shown with a lower limit of the hmixa at 200 m. *hmixa* is the mixing height, *ha* is the actual height of the trajectory and *ta* is the temperature at the actual height of the trajectory (not in scale). Every step is 15 min. The trajectories shown in these cases are the lowest possible and are following the surface close in these examples.



Harwell ozone

Figure 10: Results without a lower limit of the mixing height at Harwell in August 1997.



Harwell ozone

Figure 11: Results with a limit at 200 m of the mixing height at Harwell in August 1997.

5.4 LAND-SEA MASK

In the operational DACFOS ver 2.0 a very rough land-sea mask is used, see fig.12. As a surface characterized with a wrong land-sea mask can have big influence on the accuracy

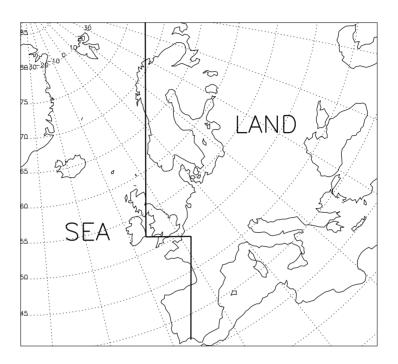


Figure 12: Land-sea mask in the operational version.

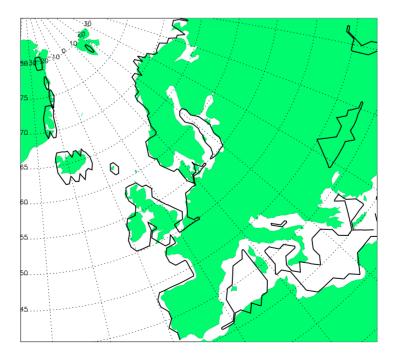


Figure 13: Land-sea mask from Henderson-Sellers (1985) [Wilson, M.F.(1985)].

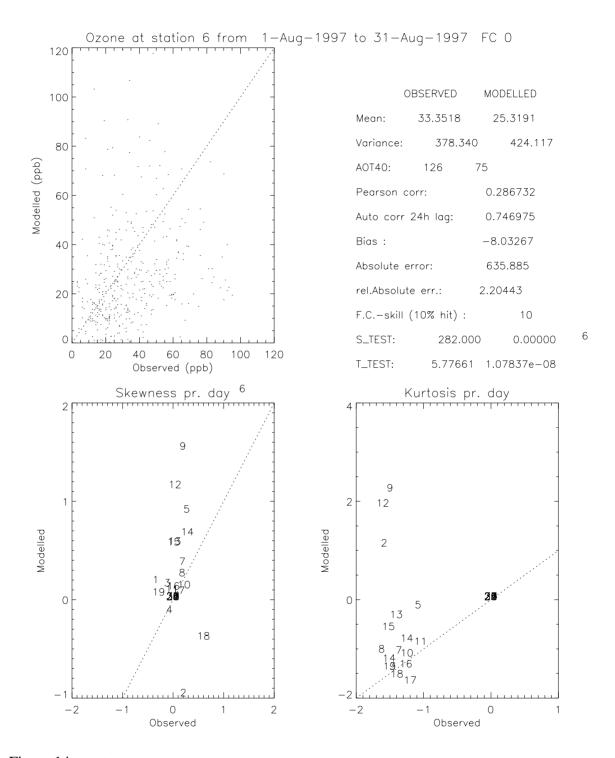


Figure 14: Verification results from calculations of ozone at Harwell in August 4-22 1997 are shown. This case is with the operational DACFOS ver 2.0 land-sea mask. In the skewness and kurtosis scatter plots day numbers are used as marks.

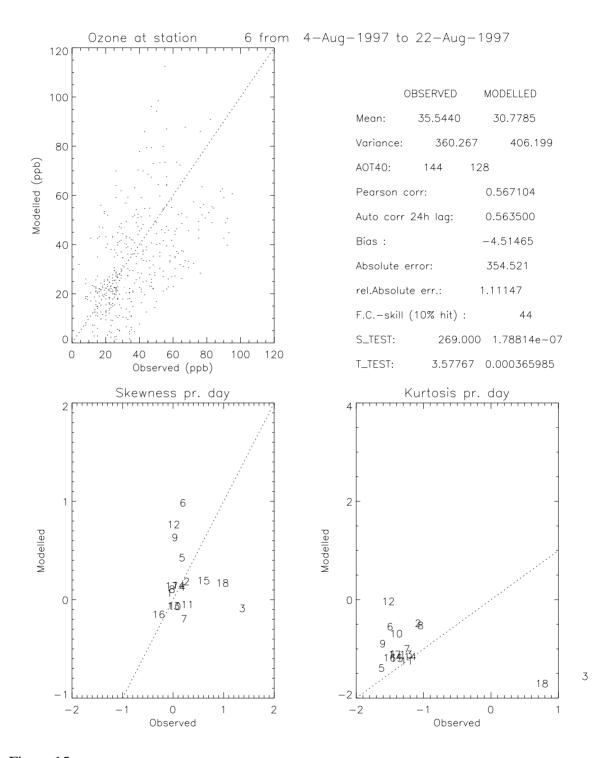


Figure 15: Verification results from calculations of ozone at Harwell in August 4-22 1997 are shown. This case is with a refined land-sea mask [Wilson, M.F.(1985)]. In the skewness and kurtosis scatter plots day numbers are used as marks.

of the calculated concentrations and deposition rates, a test has been made with the Wilson and Henderson-Sellers (WH-S) 1 degree land-sea mask [Wilson, M.F.(1985)].

Verification results for Harwell in August 4-22 1997 are shown in fig. 14 (rough operational land-sea mask) and fig. 15 (the WH-S mask). Clear improvements are seen for all the statistical parameters, e.g. has the FC-skill (Eq. 7) improved from 10 % to 44 % and the bias is significantly reduced. Implementing a new land-sea mask has a special impact on the British stations as they will be influenced by the large changes of sea areas with the lower deposition rate.

This change has not yet been implemented in the operationally DACFOS but only tested and is recommended to be implemented in the next operational version.

6 METHOD OF VERIFICATION

The purpose of the verification is to check the two datasets (modelled and observed) for the equalities in general and to check the ability of forecasting the daily maxima.

Consequently, this verification is divided into two parts, one concerning the problem of detecting systematic differences and the other concerning DACFOS' forecast skill. The verification is based on DACFOS forecasts without KALMAN filtering.

6.1 STANDARD STATISTICS

Simple statistical parameters are used on both model results and observations, e.g.

$$Mean = \overline{x} = \frac{1}{N} \sum_{j=0}^{N-1} x_j \tag{1}$$

$$Variance = \frac{1}{N-1} \sum_{j=0}^{N-1} (x_j - \overline{x})^2$$
(2)

Also some linear correlation coefficients, Pearson's r and autocorrelation $P_x(L)$ calculated by using model results and observations for r and only observations for P:

Pearsons correlation =
$$r = \frac{\sum_{i} (x_i - \overline{x})(y_i - \overline{y})}{\sqrt{\sum_{i} (x_i - \overline{x})^2} \sqrt{\sum_{i} (y_i - \overline{y})^2}}$$
 (3)

$$autocorrelation = P_x(L) = \frac{\sum_{k=0}^{N-L-1} (x_k - \overline{x})(x_{k+L} - \overline{x})}{\sum_{k=0}^{N-1} (x_k - \overline{x})^2} \quad with \ lag \ L$$
(4)

Other statistics for comparing two datasets are bias and Root Mean Square Error (RMSE):

$$bias = \frac{1}{N} \sum_{j=0}^{N-1} (x_j - y_j)$$
(5)

$$RMSE = \sqrt{\frac{1}{N} \sum_{j=0}^{N-1} (x_j - y_j)^2}$$
(6)

The most simple way to calculate a forecast skill parameter is by counting how often the model results hits the observations in an area of an uncertainty interval r (here ± 10 ppb)

$$FC = 100 \cdot \frac{1}{N} \sum (if y \in x \pm r \, then \, 1 \, else \, 0) \tag{7}$$

The skewness characterizes the degree of asymmetry of a distribution around its mean. In this context is it used relative to the skewness of the observations.

$$rel.skewness = \frac{skewness(model)}{skewness(observations)} = \frac{\frac{1}{N_m} \sum_{j=1}^{N_m} \left[\frac{x_{m_j} - \overline{x_m}}{\sigma}\right]^3}{\frac{1}{N_o} \sum_{j=1}^{N_o} \left[\frac{x_{o_j} - \overline{x_o}}{\sigma}\right]^3}$$
(8)

The kurtosis measures the relative peakedness or flatness of a distribution to the normal distribution, in this case is it also used relative to the observations.

$$rel.kurtosis = \frac{kurtosis(model)}{kurtosis(observations)} = \frac{\left\{\frac{1}{N_m}\sum_{j=1}^{N_m} \left[\frac{x_{m_j} - \overline{x_m}}{\sigma}\right]^4\right\} - 3}{\left\{\frac{1}{N_o}\sum_{j=1}^{N_o} \left[\frac{x_{o_j} - \overline{x_o}}{\sigma}\right]^4\right\} - 3}$$
(9)

The S-test function tests the hypothesis that two sample populations X and Y have the same mean of distribution against the hypothesis that they differ at the 0.05 significance level. This type of test is often referred to as the "Sign Test.".

The T-test function computes the Student's T-statistic and the probability that two sample populations X and Y have significantly different means. This type of test is often referred to as the t-means test.

$$T - test = \frac{\overline{x} - \overline{y}}{\sqrt{\frac{\sum_{j=0}^{N-1} (x_i - \overline{x})^2 + \sum_{j=0}^{M-1} (y_i - \overline{y})^2}{(N+M-2)}} \left(\frac{1}{N} + \frac{1}{M}\right)}$$
(10)

 χ^2 is used in comparison of two datasets, model and observation.

$$\chi^2 = \sum_i \frac{(Model_i - Obs_i)^2}{Obs_i} \tag{11}$$

6.2 EEA STATISTICS

The EEA (European Environmental Agency) has recommended use of 6 statistical parameters for verification of ozone forecasting [Van Aalst, R.M.(1998)], [Kiilsholm, S.(1999)]. Three parameters are concerning the skill of forecasting using a contingency table, where N is the total number of data points, f = total number of forecast exceedances, m = to-tal number of observed exceedances, a = number of correctly forecast exceedances. The parameters concerning the skill of forecasting exceedances of a threshold are

• the fraction of correct forecast smog events [Doswell C.A.III.(1990)] (Probability Of Detection, POD)

$$SP = \frac{a}{m} \cdot 100 \tag{12}$$

• the fraction of realized forecast smog events [Doswell C.A.III.(1990)] (Frequency Of Hits, FOH)

$$SR = \frac{a}{f} \cdot 100 \tag{13}$$

• a combination of *SP* and the 'probability of a null event' PON [Doswell C.A.III.(1990)] in the 'Success Index'

$$SI = \left(\frac{a}{m} + \frac{N+a-m-f}{N-m} - 1\right) \cdot 100 \tag{14}$$

The next 3 statistical skill parameters are standard statistics, either using persistency as reference model, as in S, or using an uncertainty range, as in H

• the fractional bias, FB, between averaged values of predictions and measurements

$$FB = 2\frac{\left(\overline{P} - \overline{M}\right)}{\left(\overline{P} + \overline{M}\right)} \tag{15}$$

• the skill score, *S*, where *P_i* and *M_i* are predicted, respectively measured values at day i.

$$S = 100 \cdot \left\{ 1 - \frac{\sum (P_{i+1} - M_{i+1})^2}{\sum (M_i - M_{i+1})^2} \right\}$$
(16)

• the hit score, H, indicates the overlap between the interval $(P \pm r)$ and $(M \pm r)$

$$H = 100 \cdot \frac{1}{N} \sum \frac{2r - \Delta}{2r}, \quad where \ \Delta = \min(|P - M|, 2r)$$
(17)

Both SP, SR and H have a range between 0 - 100%, SI is ranging from -100 to 100, FB ranges between -2 and +2 and S range from $-\infty$ to 100. The perfect model gives for SR, SP, SI, H and S a value of 100%, and for FB a value of 0. FB and SI have been rescaled in the figures to fit the 0-100% scale, which leads to a perfect model score at 50% for FB and at 100% for SI. In this work H is evaluated with a range $r = 10 \ \mu g/m^3$. S is also rescaled so that S equal 0 indicates that the forecast model is worse than the persistency model. A score S = 100 indicates the perfect model.

If warning of the public, affected to smog events, is the main purpose, the number of unexpected smog events (100-*SP*) should be low, at the other hand could a high rate of false-alarms reduces the confidence in the forecasts. In cases, when the forecast are used for smog prevention (e.g. traffic regulations), the false-alarm rate (100 -*SR*) should be minimal. The 'Success Index', *SI*, weights equally the correct forecasting of smog and non-smog events in one index.

7 VERIFICATION RESULTS

This verification has been divided into two parts; verification based on standard statistics and the verification method proposed by EEA.

Because of problems retrieving observations from the forecast stations around in Europe, only 9 of the 34 stations have been verified, these are Jægersborg, Lille Valby, Keldsnor, Harwell, Ladybower, Vavihill, Rorvik and Norra Kvill. Observations from the Swedish stations are only from day-hours which must be taken into account when analyzing the results. The period verified are, for all 9 stations, June 1997 to December 1998. Observations and forecasts from Jægersborg are used from April 1996.

7.1 STANDARD STATISTICS VERIFICATION 1996-98

In the verification using standard statistics all calculations are based on hourly values, which can cause some discrepancy in the results for the different stations because night observations are missing for the Swedish stations. The use of hourly values in the verification, in comparison to daily maximum values, makes it difficult to forecast as nighttime values often are hard to predict.

7.1.1 SEASONAL VERIFICATION

The distribution of hourly observations of ozone concentrations and hourly modelled ozone concentrations are shown for stations in Denmark, England and Germany. The seasons are January, February and March (JFM), April, May and June (AMJ), July, August and September (JAS) and October, November and December (OND).

A general trend in the seasonal ozone distributions for the modelled ozone in JFM and OND, see fig. 16, 17, 18 and 19, are a too high representation of ozone events around 15 ppb and too few events higher than 25 ppb, whereas the seasons AMJ and JAS shows a much better distribution. A displacement of around 10 ppb to the lower ozone concentrations is seen for the modelled distribution of ozone in AMJ compared to the observed distribution, except for Strath Vaich which has problems with too low modelled ozone concentrations in all seasons.

The peak around 1 ppb in the observed ozone values for Jægersborg (fig. 16) and Lille Valby (fig. 17) can be an indication of a zero-point problem in the measurements.

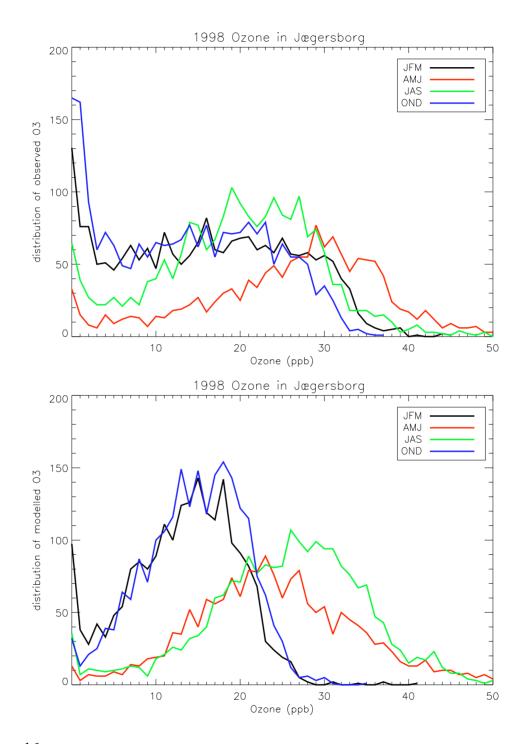


Figure 16: Ozone distribution of observed (upper) and modelled (lower) concentrations for Jægersborg 1998.

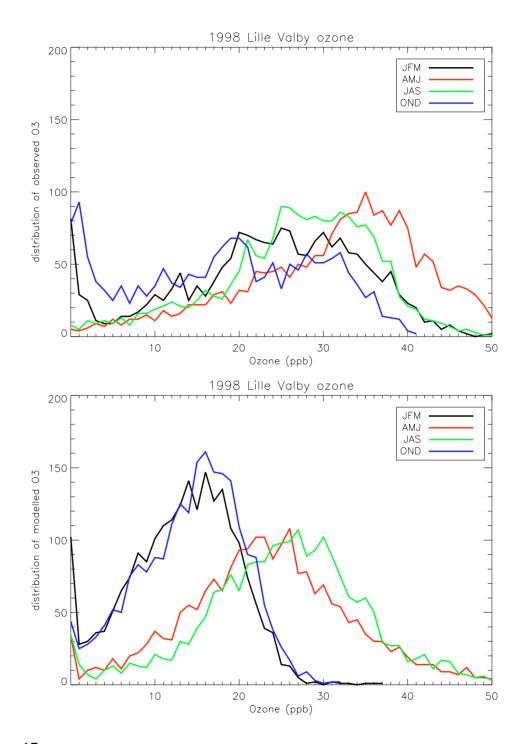


Figure 17: Ozone distribution of observed (upper) and modelled (lower) concentrations for Lille Valby 1998.

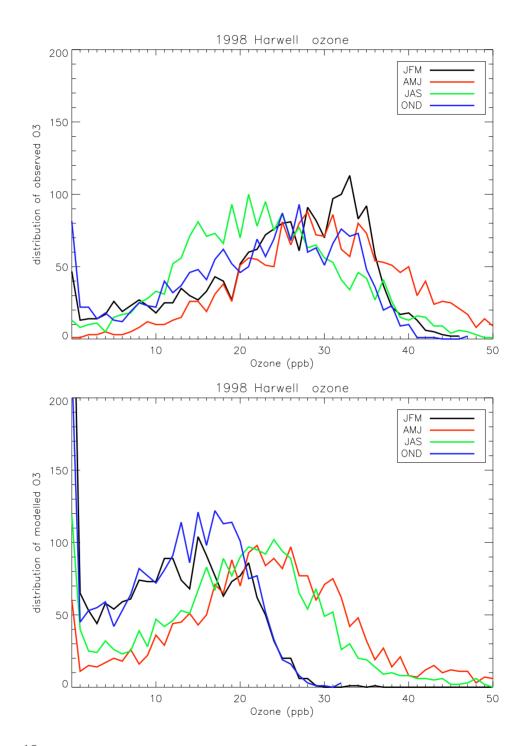


Figure 18: Ozone distribution of observed (upper) and modelled (lower) concentrations for Harwell 1998.

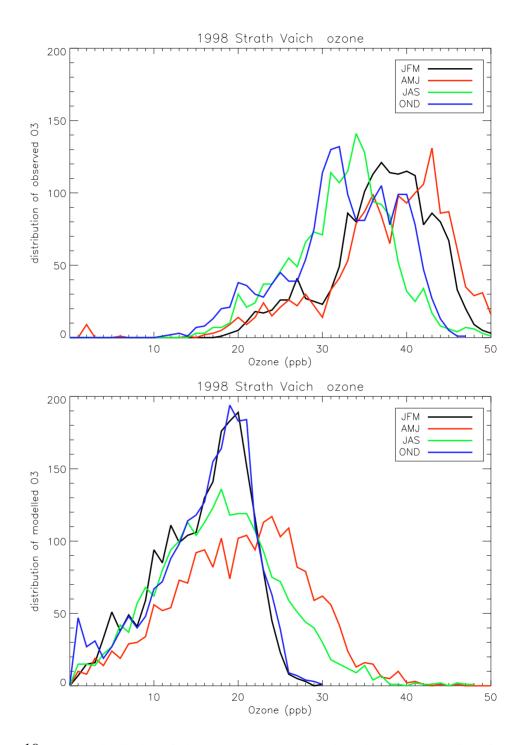


Figure 19: Ozone distribution of observed (upper) and modelled (lower) concentrations for Strach Vaich1998.

7.1.2 MONTHLY VERIFICATION

The bias, MSE and FC-skill for the 9 stations for the period June 1996 to December 1998, fig. 20, 21 and 22, shows a weak seasonal variation and the development in time seems to be quite similar for the stations from the same country.

Jægersborg most often has the lowest MSE and highest FC-skill, above 70 %, in the winters except for 1998. The performance for Lille Valby is very similar to Jægersborg, whereas Keldsnor differs especially in the autumn and winter months by having a less good performance.

For the British stations the best performance is from June to September, with August 1997 as an exception. Having the best performance in the summer is an advantage, as this is the most important period for ozone forecasting because it is the period where high ozone concentrations most often occur. The British stations, gives in general the worst performance compared to the Swedish and Danish stations, except for the late summer months when Harwell and Ladybower gives high FC-skills. The high negative bias for the British stations have been sensitivity tested for different weather types, and a test of this showed that the negative bias was closely related to continental airmasses which had travelled over areas with high emissions [Kiilsholm, S.(2000)]. This can partly be explained by incorrect emissions.

In the scatterplots, fig. 23, 24 and 25, it is clear from χ^2 (Eq. 11) that August 1997 was a month with relatively bad ozone forecasts. This could partly be explained by high bias of the temperatures from HIRLAM [Kiilsholm, S.(1999)].

The forecasts for Harwell and Ladybower (light green) do perform well; as variance (fig. 24) and correlation (fig. 25) seems to fit well, but the mean (fig. 23) shows a displacement against too low values. Strath Vaich, which is the most northern station in U.K., has rather poorly forecasts which can be seen in the mean, bias and correlation parameters. The forecasts for the Swedish stations have relatively higher variance and lower correlation which partly can be caused by the use of exclusively day-time observations. The forecasts for the Danish stations are in general performing well, except for the variance of Keldsnor predictions (dark purple) in the early summer of 1998. The low mean monthly variances in the summer 1998 depict the cold summer with low ozone values very well predicted by DACFOS.

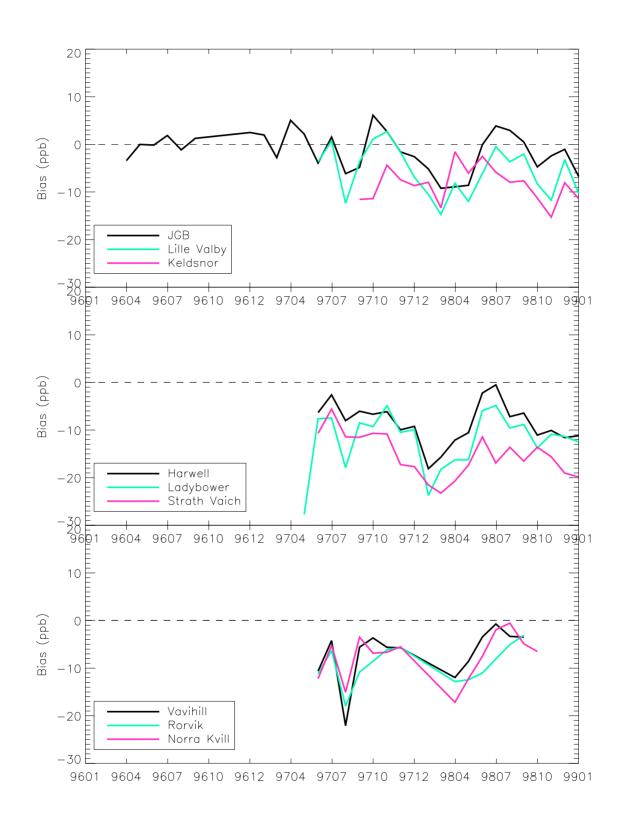


Figure 20: Mean monthly bias for 9 stations in the period Jan. 1996 to Dec. 1998. The statistics are based on hourly values.

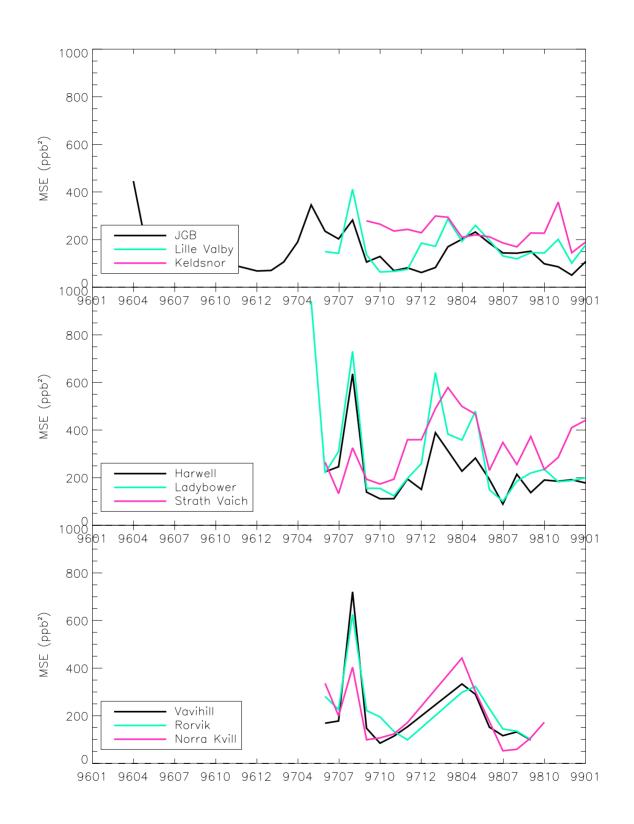


Figure 21: Mean monthly MSE for 9 stations in the period Jan. 1996 to Dec. 1998. The statistics are based on hourly values.

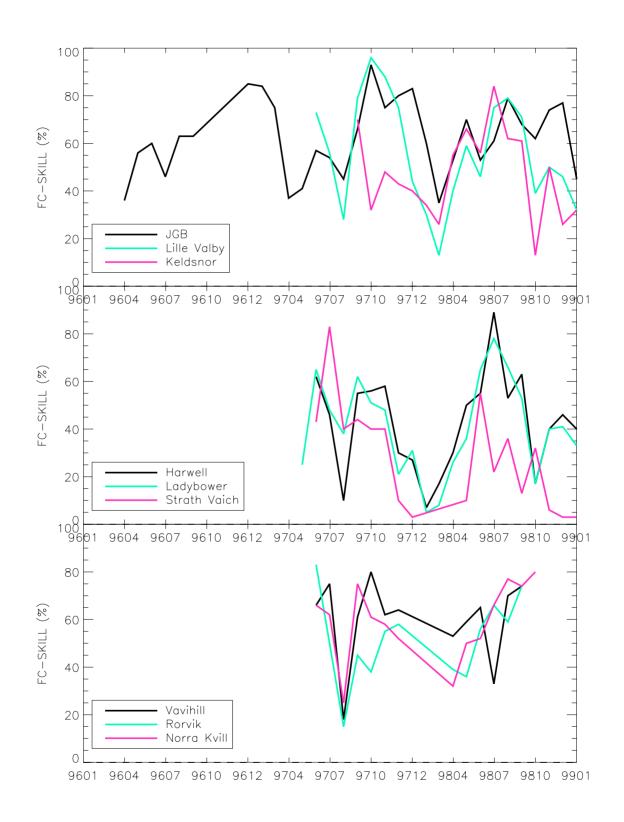


Figure 22: Mean monthly FC-Skill for 9 stations in the period Jan. 1996 to Dec. 1998. The statistics are based on hourly values.

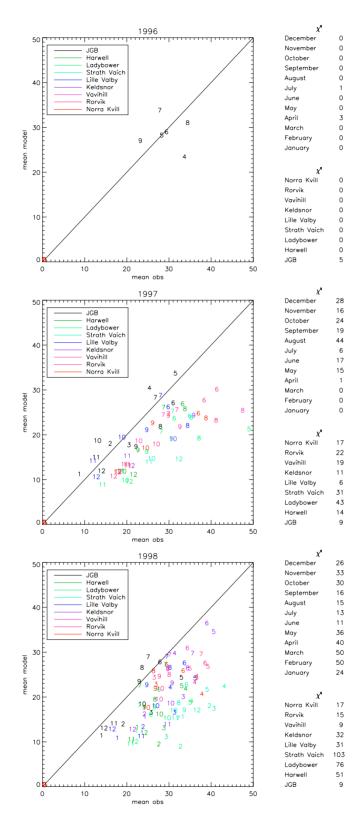


Figure 23: Scatterplot of mean monthly ozone values for 1996-98. The numbers correspond to the month number and the colors correspond to the station (see legend). The χ^2 number is a sum of χ^2 for all stations in one month or for all months at one station.

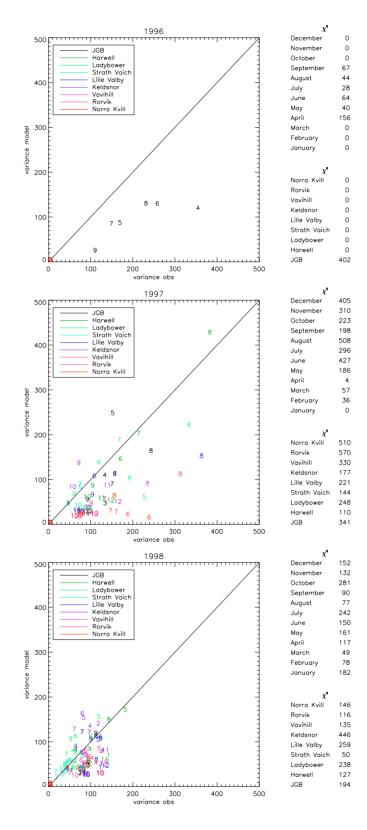


Figure 24: Scatterplot of mean monthly variance on ozone values for 1996-98. The numbers correspond to the month number and the colors correspond to the station (see legend). The χ^2 number is a sum of χ^2 for all stations in one month or for all months at one station.

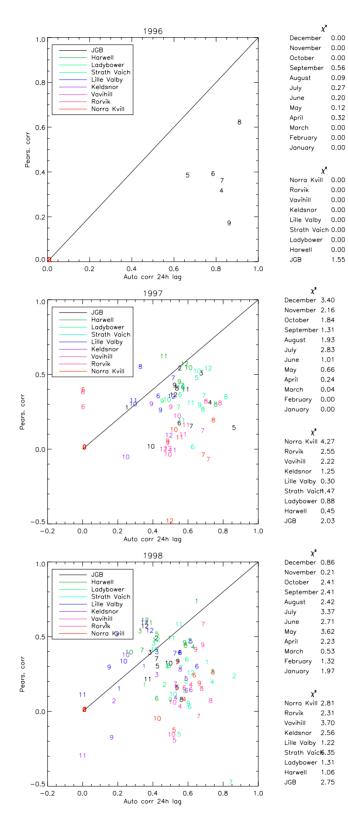


Figure 25: Scatterplot of mean monthly correlation on ozone values for 1996-98. The x-axis show the autocorrelation on observations (corresponding to correlation of persistency) and the y-axis show the correlation between model results and observations. The numbers correspond to the month number and the colors correspond to the station (see legend). The χ^2 number is a sum of χ^2 for all stations in one month or for all months at one station.

7.1.3 DIURNAL VERIFICATION

The skewness is positive for Jægersborg and Lille Valby in autumn and winter and negative in spring and summer. A positive rel. skewness indicates that the predicted peaks of ozone during the day were shifted to a later hour than observed and negative values that the peaking were earlier, by which it can be concluded that diurnal peaking are happening too late in autumn and winter and too early in spring and summer for the Danish stations, whereas the British stations most often reach there ozone maximum too early, except for Harwell in autumn, see fig. 26.

The kurtosis shows no clear trend for the period shown. Positive kurtosis are an indication of a leptokurtic (a more concave and narrow peak than the normal distribution) and negative when the peak are platykurtic (broader and convex). A small tendency to relatively narrow peaks for the British stations and relatively broader peaks for the Danish stations can be claimed.

The T-test and S-test, fig. 27 and 28, are testing if the mean of the observed and modelled ozone concentrations are significant different. When the values are lower than 0.05 the difference is "very significant". Only for a very few cases shows Jægersborg, Lille Valby and Keldsnor significant different means, whereas the northern British stations, especially Strath Vaich are significant different most of the time.

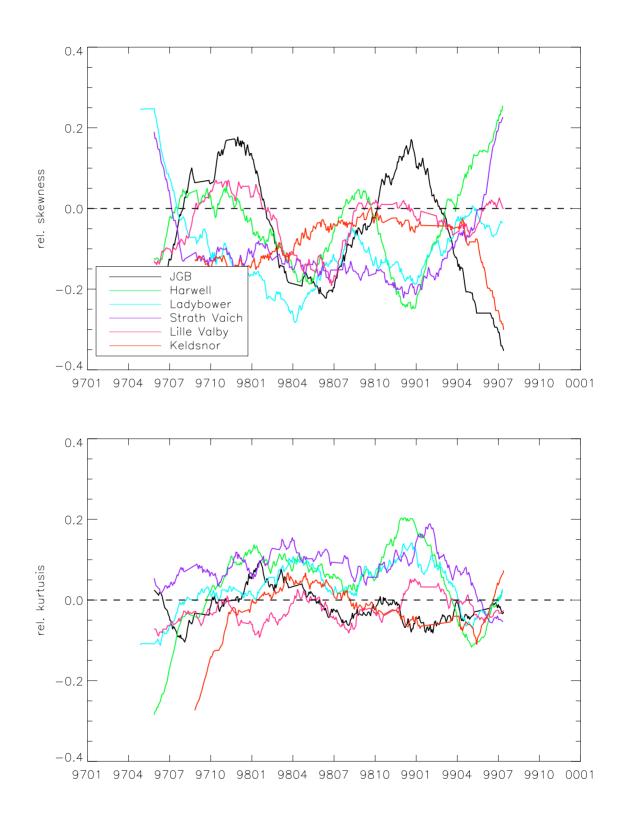


Figure 26: Daily 31 days smoothed rel. skewness, kurtosis for 6 stations in the period Jan. 1996 to July 1999. The statistics are based on hourly values calculated for every day.

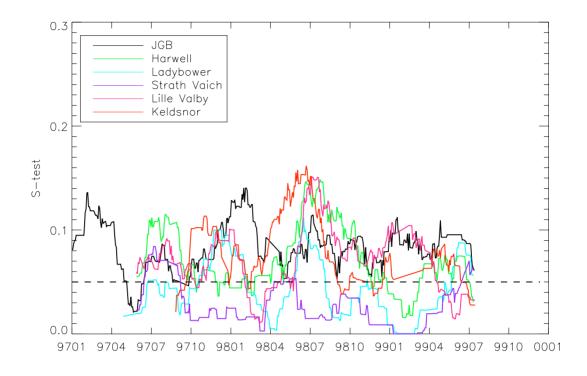


Figure 27: 31 days smoothed S test for 6 stations in the period Jan. 1997 to July 1999. The statistics are based on hourly values calculated per. day.

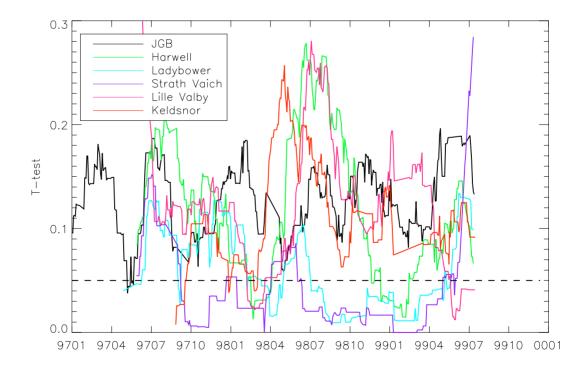


Figure 28: 31 days smoothed T test for 6 stations in the period Jan. 1997 to July 1999. The statistics are based on hourly values calculated per. day.

7.2 EEA VERIFICATION 1995-98

This type of verification differs from the standard verification by using the daily observed/modelled maximum ozone value, which makes it a verification of the ability to forecast ozone peaks.

Comparisons of the forecast skills between years can be complicated by the fact that the ruling weather types differs from year to year with big influence on the concentration of ozone. The geographical differences between stations have a big impact on the forecast skill.

The six calculated statistical parameters are presented in a rose (fig. 29), with the persistency skill value and the forecast skill value overlaying each other, the lowest value are on top. A small black piece of cake with a broad grey rim is a representation of an ideal DACFOS forecast compared to the persistency. In case of equal values for the forecast skill and the persistency skill, the piece of cake has a single color.

In this verification the EEA verification method has been used in three different ways; a long period of four years at one station in order to compare the forecast skill from year to year, a shorter period of two years where the forecast skill at different geographical areas (England compared to Denmark) are verified and periods of a year where the forecast skill of the different information intervals are verified for several stations.

The 'event threshold' level suggested by EEA are at 180 $\mu g/m^3$, and only days with ozone concentrations above 120 $\mu g/m^3$ are taken into account. In Denmark, exceedances of the 180 $\mu g/m^3$ threshold occur very seldom. As an example, during the period 1 April - 30 July 1996 only one exceedance of the population information threshold value was observed in Denmark, [Sluyter, R.(1996)] and in the summer season 1995, five exceedances were detected [De Leeuw F.A.A.M.(1995)]. With these low numbers of exceedances, even for long periods, the statistics gives no meaningful results for the suggested 'event

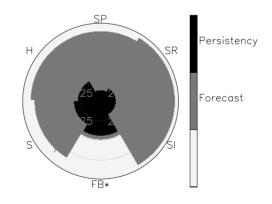


Figure 29: An example of an ideal verification presented in a rose as used for the EEA verification.

threshold'. These conditions are the reason for choosing a lower 'event threshold' in this verification. In the DMI public information system 'high' concentrations are defined to be between 120-180 $\mu g/m^3$. For the verification of DACFOS 120 $\mu g/m^3$ is used as the 'event threshold', which also is close to the 130 $\mu g/m^3$ level used in US as a level for 'limited health Notice' threshold [Maine Department of Environmental Protection.(1998)]. The verification is only based on days with higher concentration than 60 $\mu g/m^3$.

7.2.1 JÆGERSBORG 1995-1998

A verification for the summer seasons 1995 - 98, in Jægersborg (Denmark), fig. 30, shows varying results from year to year partly caused by the change of the weather conditions and partly caused by the few numbers of events in some years.

In general, the number of forecasted exceedances are lower than the number of observed exceedances, giving a SP (Eq.12) lower than SR (Eq.13) for the forecast skills. As seen on fig. 30, the observed exceedances occur around twice as often as the forecasted exceedances. The best forecasted year was 1995 with 'skill scores', S (Eq.16), higher than persistency. High statistical scores for the forecast were also seen in 1998, but persistency had a better 'hit and skill score', H (Eq.17) and S. It must be mentioned here that the 1998 verification only were based on June and July, with a few exceedances. Persistency are doing well in 1996 for 'hit and skill score', H and S, caused by exceedances on consecutive days.

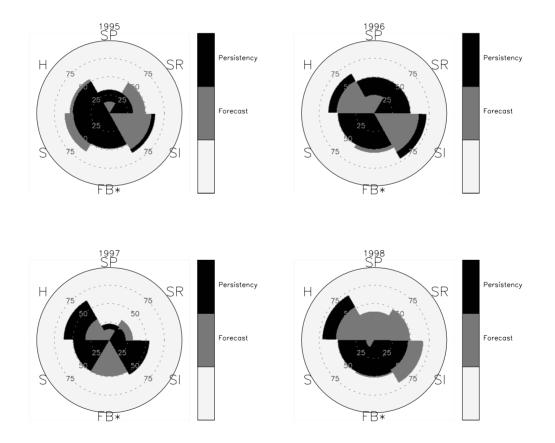


Figure 30: Statistical parameters for Jægersborg (JGB) calculated for the years 1995-1998, based on DACFOS 24-29 hour forecasts. Model and persistency values are shown in the same piece, where the smallest are overlaying the biggest. FB* differs from the other parameters by having an ideal value at 50%.

7.2.2 ENGLISH STATIONS VERSUS DANISH STATIONS

The stations Harwell and Ladybower, in England, and Lille Valby and Jægersborg, in Denmark, are chosen for a verification in the period June, July, August, September 1997 and June, July 1998.

In fig. 32, DACFOS forecast skills are shown for the stations in England and Denmark. Model results are calculated for the three intervals: analysis (-6 to -1 hour), 24h-29h forecast and 43h-48h forecast. With four forecasts per day intervals of 6 hourly values are used to get values 24 hours a day.

DACFOS gives a better forecast performance for England than for Denmark in 1997-1998, especially showed by the skill parameters SR, 'Frequency of hits', and SP, 'Probability of detection'. The change of skill with the forecast length are similar for forecasts for England and Denmark. SP, SI, H and FB has a slightly decreasing skill with forecast length. SR are different from the other parameters by showing an increasing skill, which probably are due to fewer forecasted exceedances. The most drastic changes are detected on S, 'Skill score', in forecasts for England, where it reduces from around 35% to 5 %. Persistency gives higher SP and SR values for forecasts for England than for Denmark (50% versus 25%), which probably can be explained by different ruling weather patterns. Single standing, sporadic exceedances of the 'event threshold' occur more often over Denmark, than over England in 1997-98.



Figure 31: Map of the location of the 4 stations used in the verification.

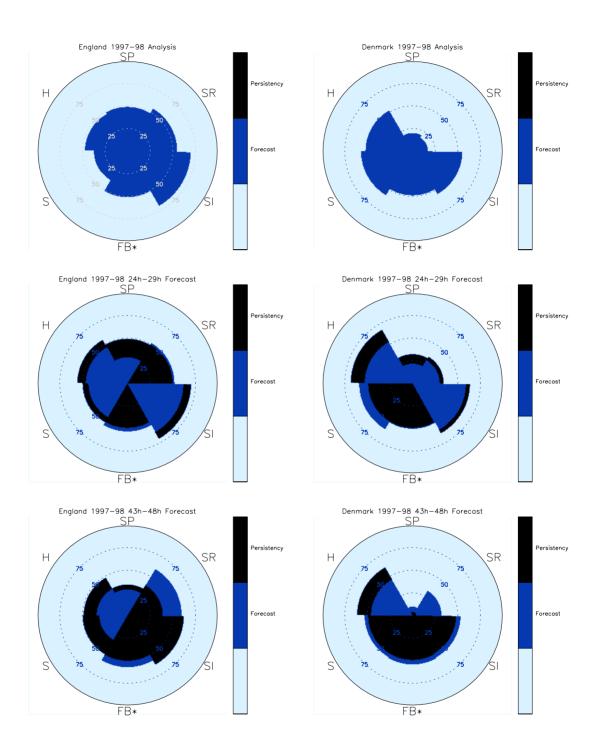


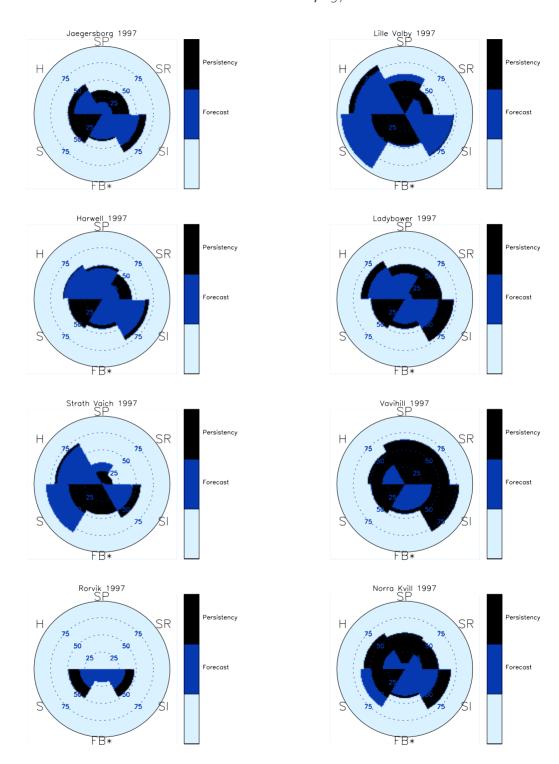
Figure 32: Six statistical parameters for two areas, England and Denmark, shown for three model forecast intervals; analysis, 24h-29h and 43h-48h forecast. Model and persistency values are shown in the same piece, showing the smallest values overlaying the biggest. In case of equal values, only one color is shown. FB* differs from the other parameters by having an ideal value at 50%.

7.2.3 OZONE INTERVALS

In this verification the threshold used for the verification has been changed to intervals used in the DMI public information system. These 3 intervals are [0-60], [60-120], [120-above 120] $\mu g/m^3$ ozone, described by low, mean, and high (see section 4). A verification on these intervals can give some information of the reliability of the forcasts in the different intervals.

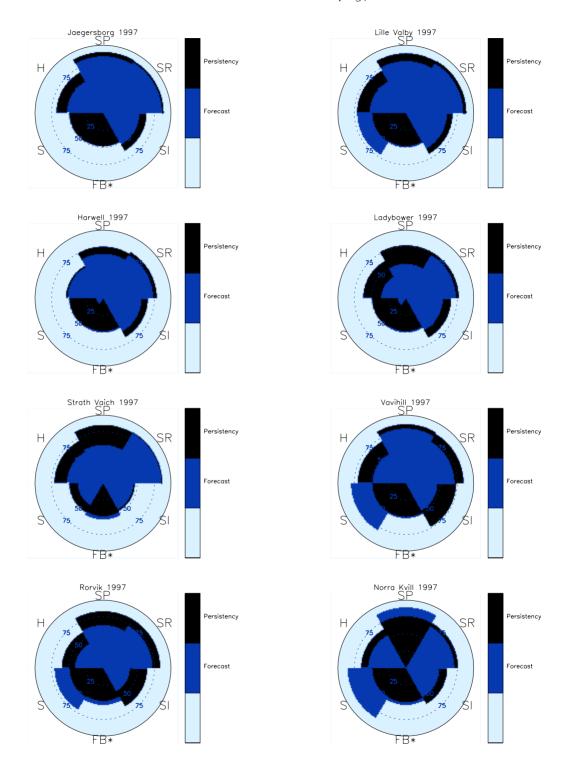
- 0-60 $\mu g/m^3$; The probability of detection 'SP' is generally higher than the frequency of hits 'SR' around values at 50% for the Danish and British stations.
- 60-120 $\mu g/m^3$; In this interval the verification results for 1997 and 1998 do not differ substantially, except for the skill score index 'S'. The high number of events in this interval gives good results around 80% for SP and SR, success index 'SI' around 60%. In June-July 1998, SR was slightly higher than SR for the persistency for 4 stations, Lille Valby, Keldsnor, Harwell and Ladybower, though Ladybower and Strath Vaich seems to have the worst performance when all 6 parameters are weighted equally.
- 120-∞ µg/m³; The probability of detection 'SP' is generally lower than the frequency of hits 'SR', both varies a lot. Hit 'H' and Skill 'S' score shows much better results for 1998 than for 1997 at all stations, except Strath Vaich which had a very high negative bias in 1998. This big difference in scores between 1997 and 1998 is probably due to the bad performance of forecasts in August 1997.

In general the skill score 'S', which shows the skill of forecast compared to the persistency, are high for the Swedish stations, Lille Valby and Keldsnor. Harwell is the only station for which DACFOS hit score 'H' is near equal to persistency hit score. The forecasts are in general too high, which can be seen on the fractional bias 'FB' in fig. 33, 36 being above 50 % at all stations. For medium and high intervals the bias are below 50 %.



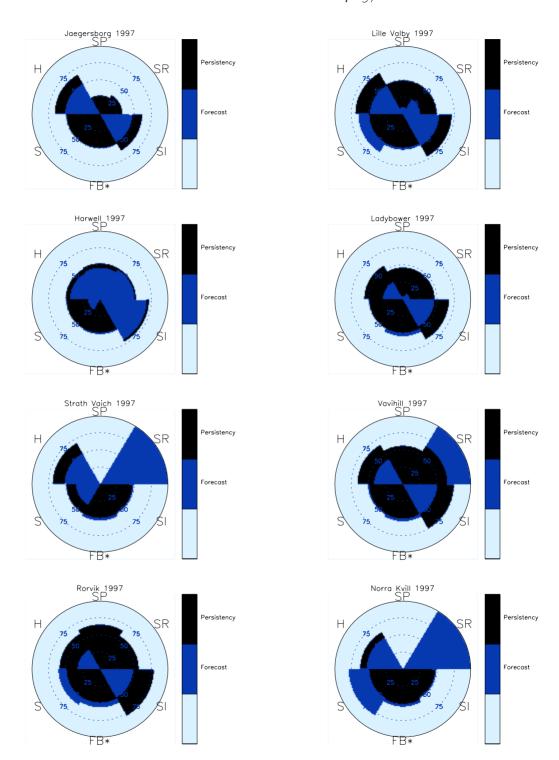
Ozone conc. 0-60 μ g/m³

Figure 33: 8 stations; Jægersborg, Lille Valby, Harwell, Ladybower, Strath Vaich, Vavihill, Rorvik and Norra Kvill verified for the ozone interval 0-60 $\mu g/m^3$ in the period June-Sept. 1997.



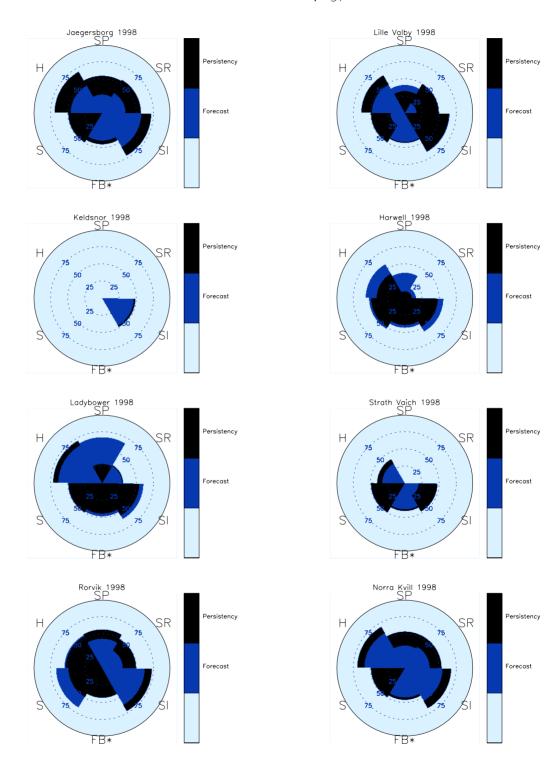
Ozone conc. 60–120 μ g/m³

Figure 34: 8 stations; Jægersborg, Lille Valby, Harwell, Ladybower, Strath Vaich, Vavihill, Rorvik and Norra Kvill verified for the ozone interval 60-120 $\mu g/m^3$ in the period June-Sept.1997.



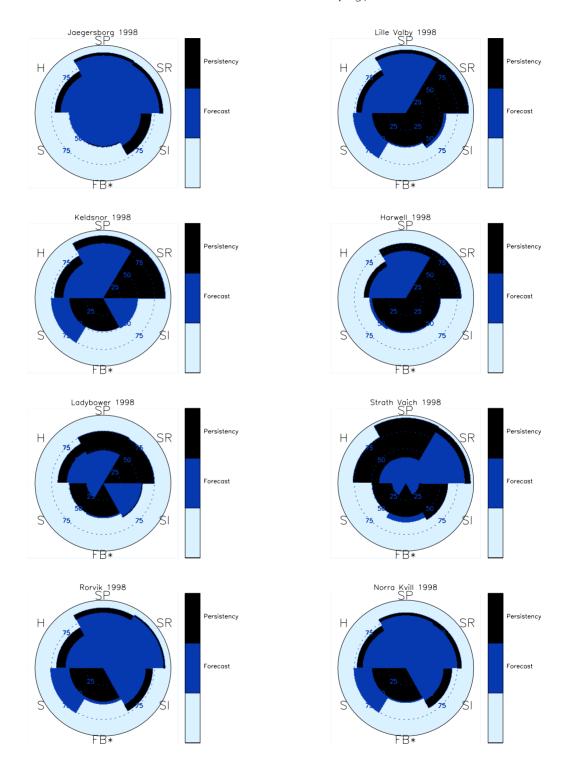
Ozone conc. over 120 μ g/m³

Figure 35: 8 stations; Jægersborg, Lille Valby, Harwell, Ladybower, Strath Vaich, Vavihill, Rorvik and Norra Kvill verified for the ozone interval above $120 \ \mu g/m^3$ in the period June-Sept. 1997.



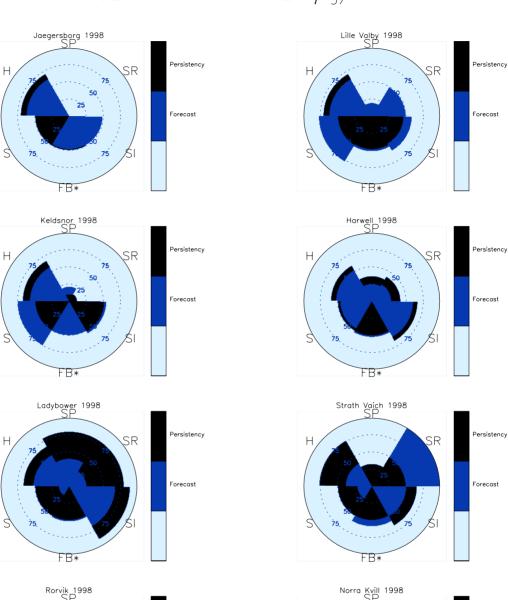
Ozone conc. 0-60 μ g/m³

Figure 36: 8 stations; Jægersborg, Lille Valby, Keldsnor, Harwell, Ladybower, Strath Vaich, Rorvik and Norra Kvill verified for the ozone interval 0-60 $\mu g/m^3$ in the period June-July 1998.



Ozone conc. 60–120 μ g/m³

Figure 37: 8 stations; Jægersborg, Lille Valby, Keldsnor, Harwell, Ladybower, Strath Vaich, Rorvik and Norra Kvill verified for the ozone interval 60-120 $\mu g/m^3$ in the period June-July 1998.



Ozone conc. over 120 μ g/m³

Figure 38: 8 stations; Jægersborg, Lille Valby, Keldsnor, Harwell, Ladybower, Strath Vaich, Rorvik and Norra Kvill verified for the ozone interval above 120 $\mu g/m^3$ in the period June-July 1998.

Persistency

Forecast

SR

FB*

Persistency

Forecast

SR

FB*

In the tabels 4 and 5 below, results from the previous shown EEA forecast skills are summed up in two categories; where the DACFOS model performs better than or are equal to persistency. The statistical parameters are equally weighted.

The British stations show an improved relative forecast skill in 1998 compared to 1997 for the lowest ozone interval, whereas the Swedish and Danish stations showed improved relative forecast skill in 1998 for the two highest ozone intervals (see table 4 and 5). The Swedish stations are in general having a better relative forecast skill for the ozone levels above 60 $\mu g/m^3$.

Ozone ($\mu g/m^3$)	0-60				60-120			
Year	1997		1998		1997		1998	
Model best /equal	model	equal	model	equal	model	equal	model	equal
Jaegersborg	0	1	0	0	0	3	0	4
Lille Valby	2	2	1	2	1	1	1	4
Keldsnor	-	-	-	-	-	-	1	3
Harwell	0	3	4	2	0	3	0	4
Ladybower	0	0	2	3	0	2	0	3
Strath Vaich	2	2	0	3	0	3	1	0
Vavihill	1	0	-	-	1	2	-	-
Rorvik	0	4	1	2	1	0	1	3
Norra Kvill	1	1	0	2	2	2	1	3

Ozone ($\mu g/m^3$)	120-∞						
Year	19	97	1998				
Model best /equal	model	equal	model	equal			
Jaegersborg	0	1	0	3			
Lille Valby	1	1	4	2			
Keldsnor	-	-	3	3			
Harwell	0	3	0	3			
Ladybower	0	0	0	1			
Strath Vaich	1	3	2	0			
Vavihill	1	2	-	-			
Rorvik	2	0	1	4			
Norra Kvill	2	3	1	4			

Table 4: As tabel 5.

Table 5: Number of statistical parameters where the DACFOS model performs better than or are equal to persistency out of 6 possible. The definition of equal performance are when there are less than 10 % difference. For FB an equal performance is defined by +5 < (|model - 50| - |persistency - 50|) < -5.

8 **DISCUSSION**

The source of errors in the ozone results can be numerous; inaccurate emissions, errors in the HIRLAM weather prediction [Kiilsholm, S.(1999)] or bad treatment of the physics and chemistry in DACFOS.

8.1 STANDARD VERIFICATION

A too high representation of ozone events around 15 ppb and too few events higher than 25 ppb is a general trend in the seasonal ozone distributions for the modelled ozone in JFM and OND, whereas the seasons AMJ and JAS shows a much better distribution.

The forecasts for the Danish stations are in general performing well, except for the variance of forecasts for Keldsnor in the early summer of 1998. The forecasts for the British stations gives in general the worst bias, MSE and FC-skill, except for the summer months where forecasts for Harwell and Ladybower gives high FC-skills, whereas forecasts for Jægersborg most often has the lowest MSE around 60 ppb and highest FC-skill above 70 % from October to January, whereas the spring and summer skills varies a lot from year to year

The stations in the same region have nearly the same forecast errors for the whole period, which shows a big regional influence on the ozone forecasts.

8.2 EEA VERIFICATION

The EEA proposed method for verification of the skill performance of ozone forecast models gives an impression of the complexity of verification, as it is clearly seen that the different parameters differs a lot and varies independently of each other. The method can be used when the chosen 'event threshold' is low enough to give some events both for measurements and for forecasts, otherwise undefined values for SP and SR will occur.

Due to very few exceedances of the EEA proposed ozone exceedance threshold in Denmark, an alteration of the exceedance threshold from $180 \ \mu g/m^3$ to $120 \ \mu g/m^3$ has been made. Unfortunately, this modification makes it impossible to compare with other model verifications, based on the EEA proposed verification principles, but comparisons in time and place are made possible for the DACFOS forecast in Denmark and England.

The comparison of the DACFOS forecast skill in England and Denmark, section 7.2.2, shows a better peak forecast for England than for Denmark in 1997-1998. The DACFOS peak forecast for Denmark in 1997, section 7.2.1, had relatively low forecast skill compared to the other years verified, so the fact that the relative best performance of forecasts are seen in England can not be generalized to all years.

In section 7.2.3 it was shown that the skill score 'S' in general are high, around 70%, for the performance in intervals for the Swedish stations, Lille Valby and Keldsnor, whereas

forecast for Harwell is close to the persistency for the hit score 'H'.

Big differences in the statistical parameters between the years 1995-1998 show that factors like weather and number of exceedances have a noticeable effect on the statistics. In cases when the statistical parameters should be used for a comparison between different forecast models, long periods of verification must be used in order to avoid influences from irrelevant factors.

9 CONCLUSION

Some modifications concerning the handling of the trajectories, emissions and mixing height has been implemented in DACFOS and tests on a new land-sea mask showed that further development on DACFOS are needed.

Verification of ozone forecasts are rather difficult as the parameters influencing on the predicted ozone concentrations are numerous, which gives verification results that are changing a lot in time and space. As a result of this a very long period of forecasts and observations are needed to get a reliable basis for any generel conclusions of DACFOS' forecast skill.

The verification concerning systematic differences showed a bad distribution of the modelled ozone in the months from October to March for all stations. The monthly mean forecasts are in generel to low and the mean square error 'MSE' and the forecast skill 'FC' (Eq. 7) varies a lot whitout any clear monthly trend.

The verification concerning DACFOS' peak forecast skill, the EEA verification, showed a somewhat better result for the peak forecasts for the southern English stations. This can partly be explained by the fact that the EEA verification is sensitive to the number of events, and the relatively high number of events at the southern English stations gives better statistics.

10 ACKNOWLEDGMENT

This work was supported by the Danish Ministry of Traffic under the project Surveillance of the Contribution from Traffic to Local Air Pollution Levels in Denmark. The EMEP model was provided from the Norwegian Meteorological Institute (DNMI). Real time ozone data were provided on the internet by NERI (the National Environmental Research Institute), AEA-Technology and IVL.

11 REFERENCES

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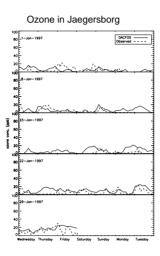
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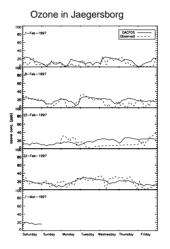
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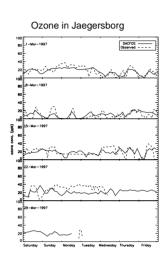
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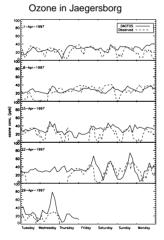
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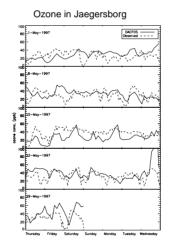
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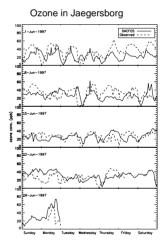


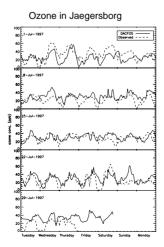


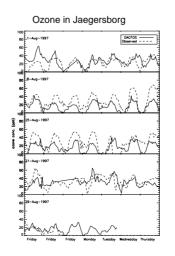


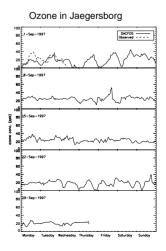


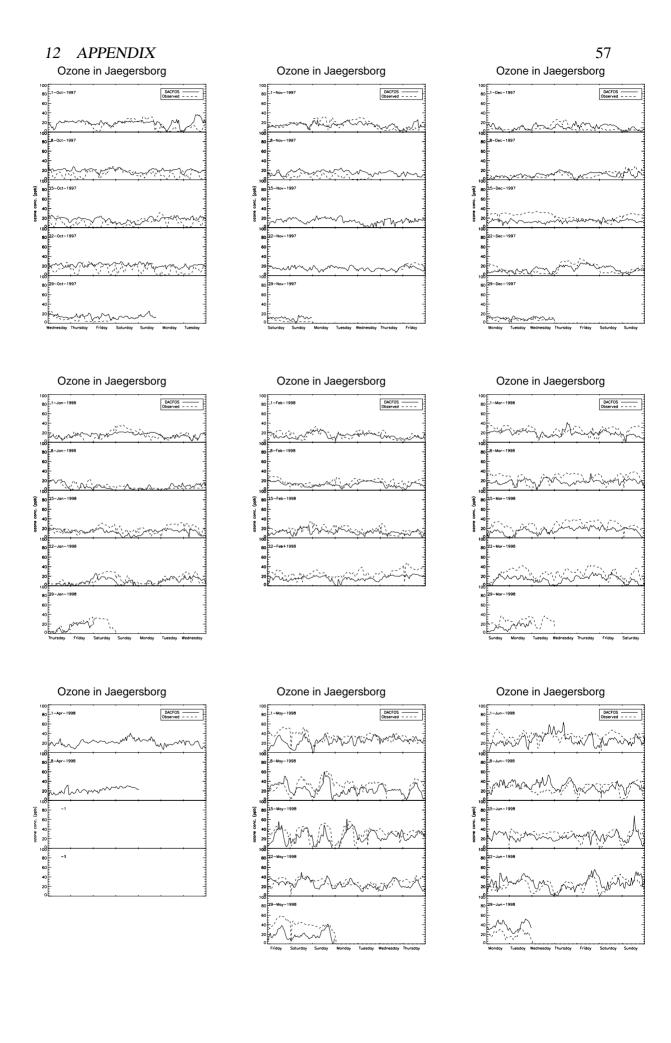


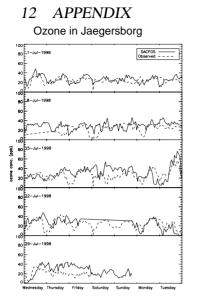


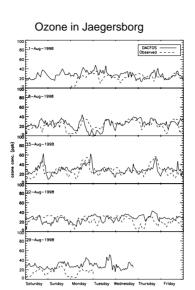


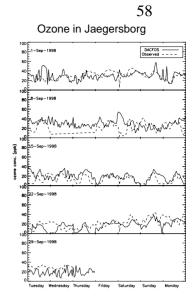




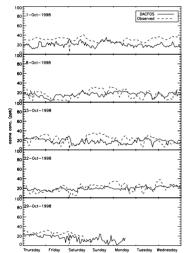


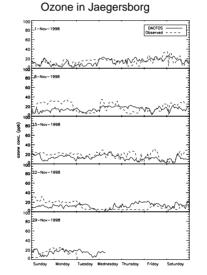




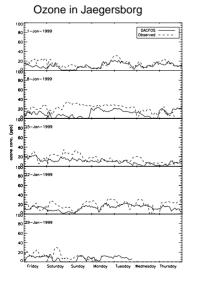


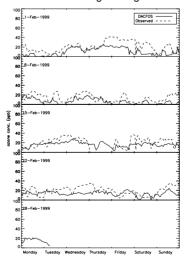
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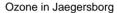


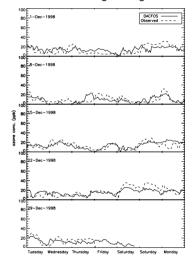




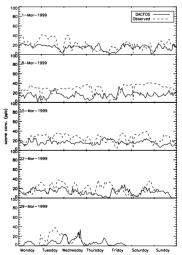


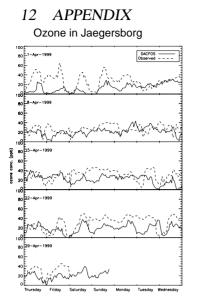


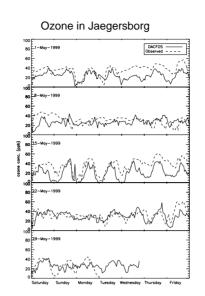


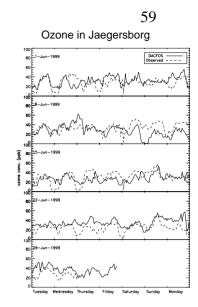




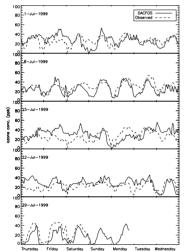




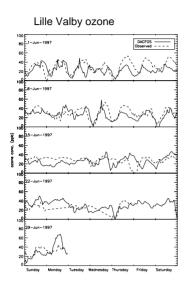


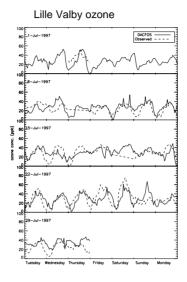


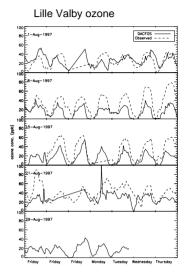
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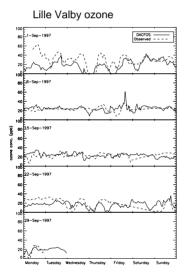


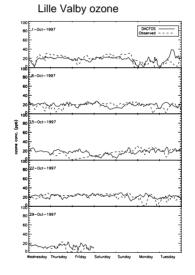
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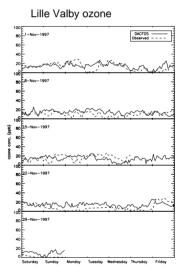




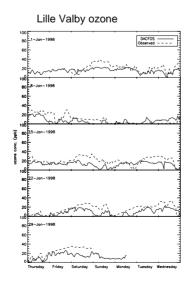




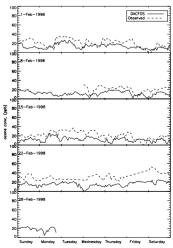




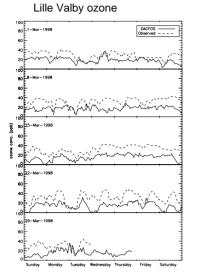
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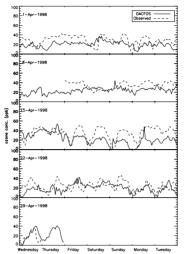
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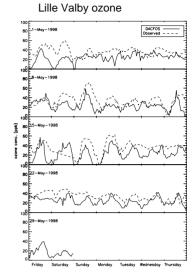


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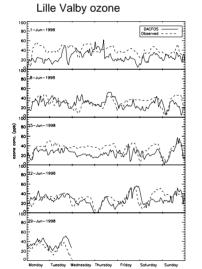


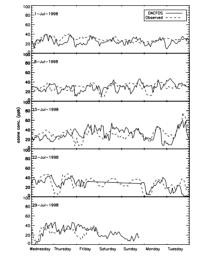
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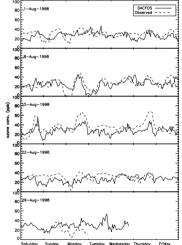


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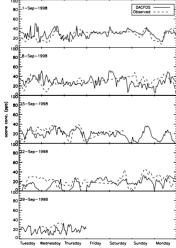


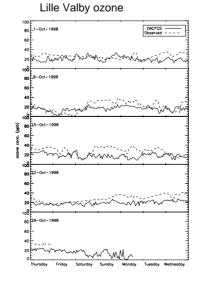




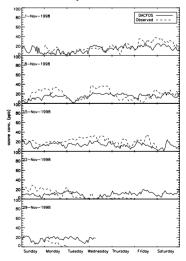






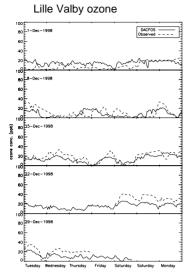


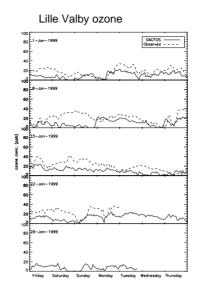
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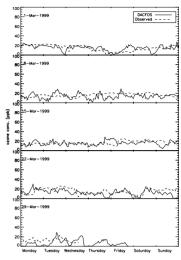


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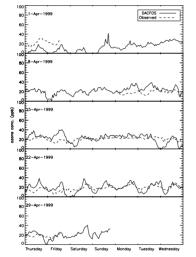
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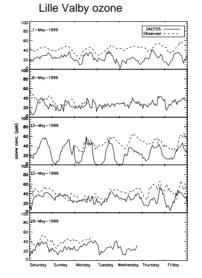




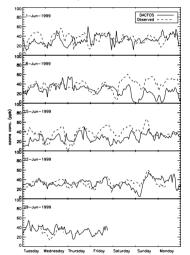


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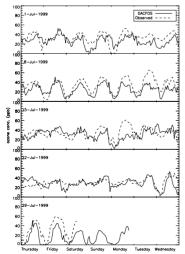








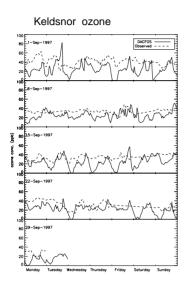


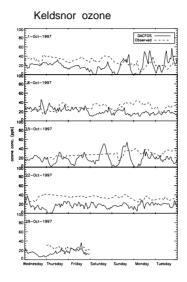


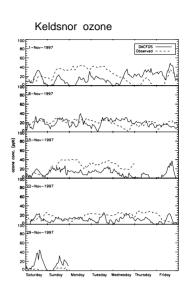
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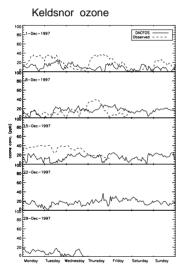
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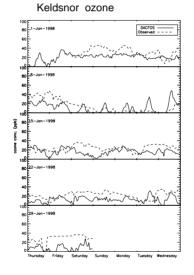
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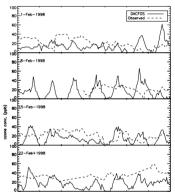




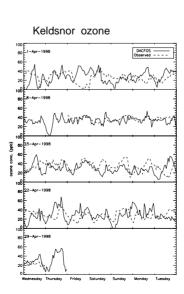


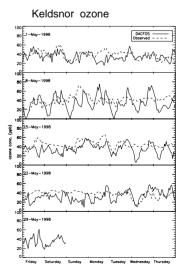


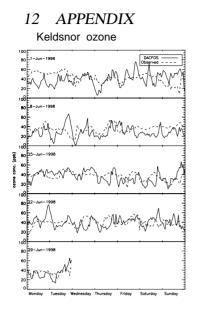
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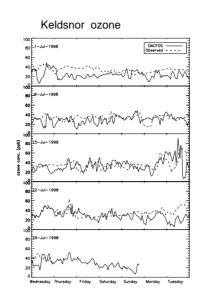


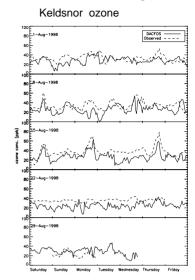
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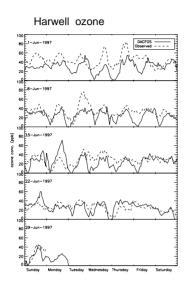




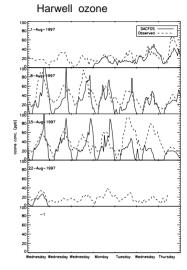


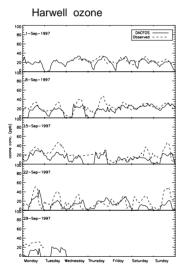


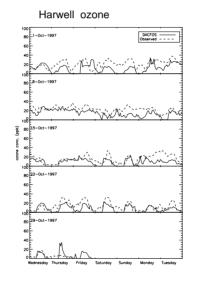
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Harwell ozone DACFOS Observed (qdd ozone

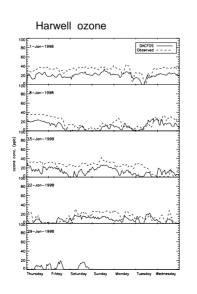






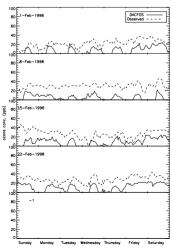
Harwell ozone DACFOS -Observed n.A. N/ What' (ddd) Conc. au 20 10 20 0

Harwell ozone 10 DACFOS ---80 Dec - 1993 20 108 n' M -M NXWA in m 1201 -Dec-1993 (qdd) 500. ozone 20 108 wi iA ~ ...8

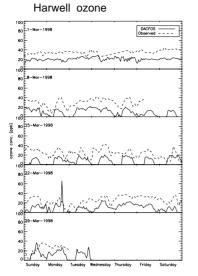


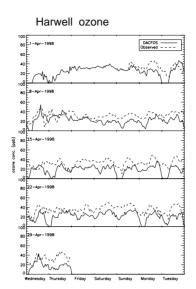
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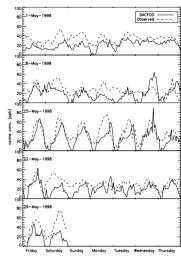
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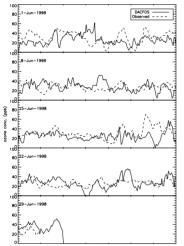
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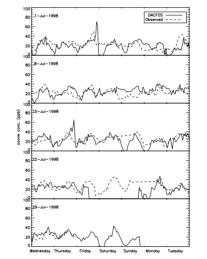




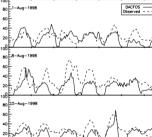


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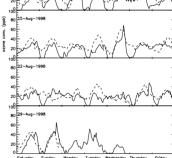


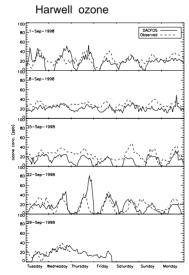


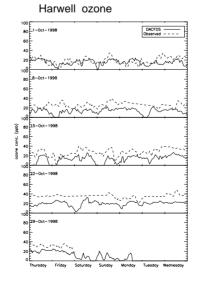
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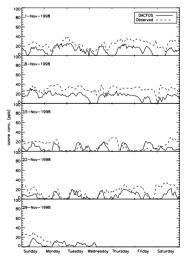
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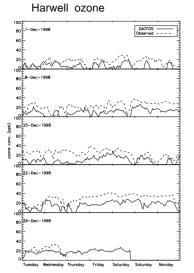
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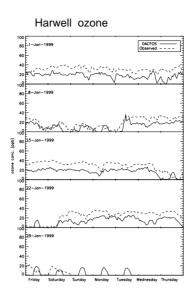


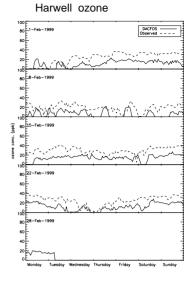
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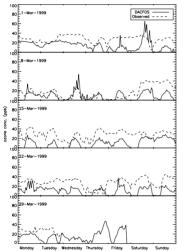
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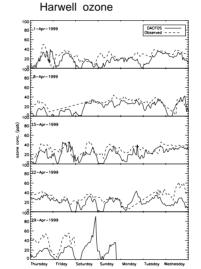


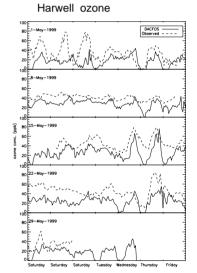




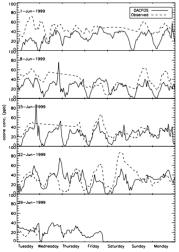


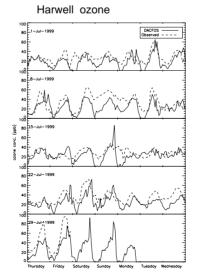






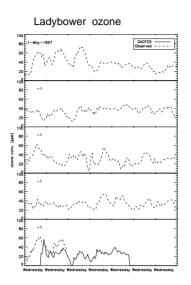


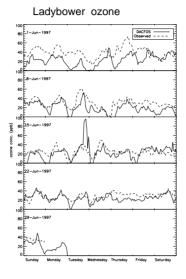


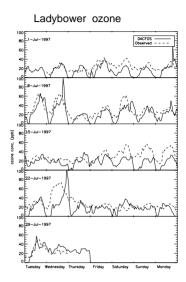


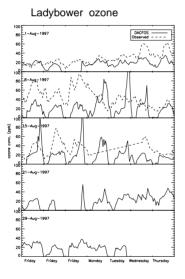
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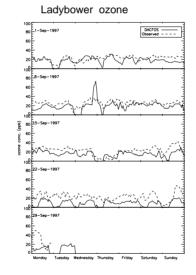
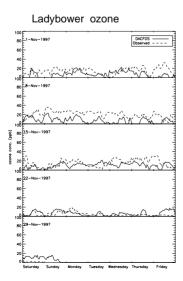
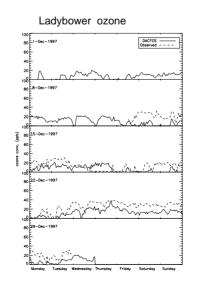


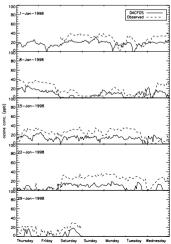
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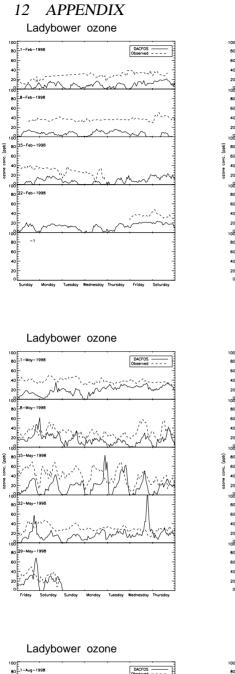
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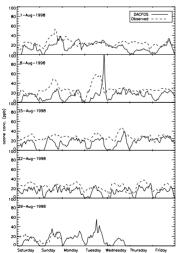


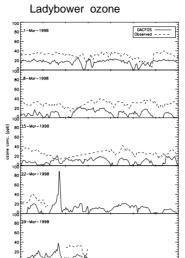


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Ladybower ozone

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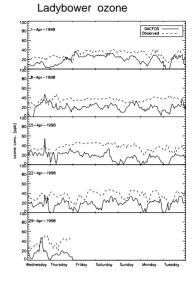
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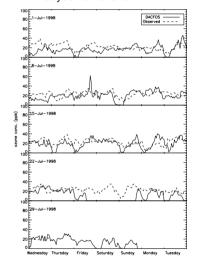
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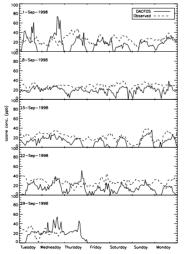


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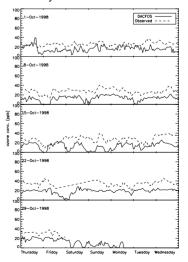




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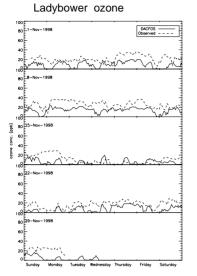


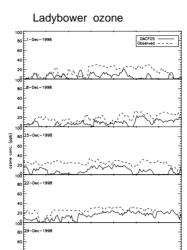
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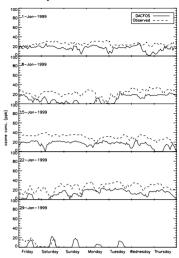
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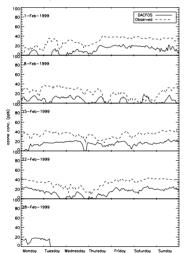


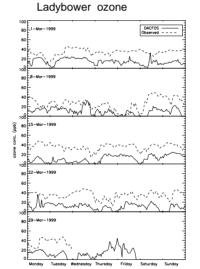


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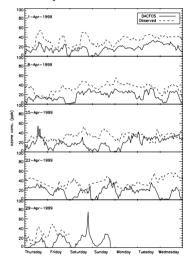


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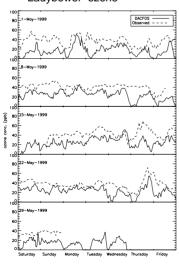


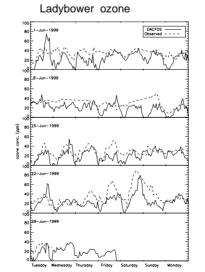








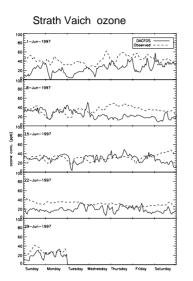


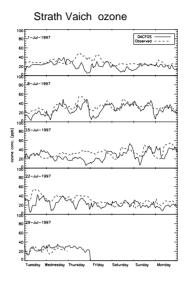


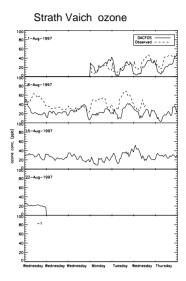
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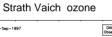
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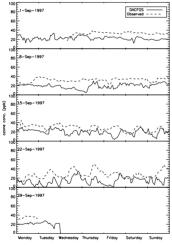
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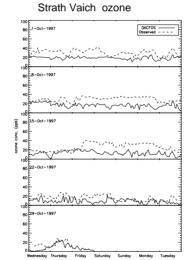




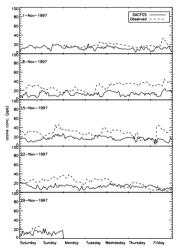


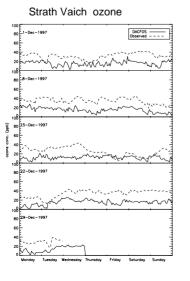


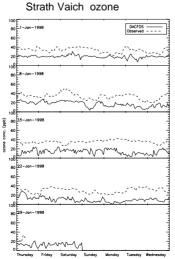




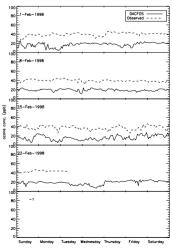
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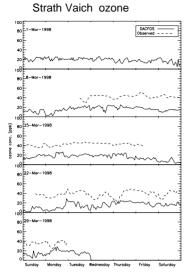




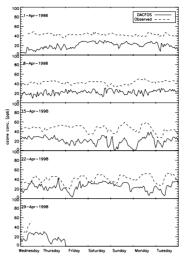
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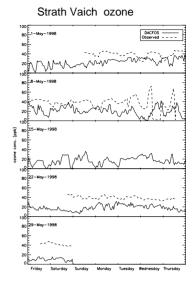




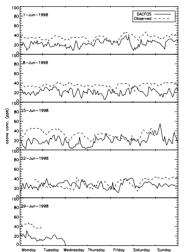


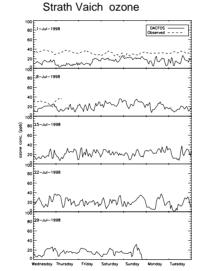
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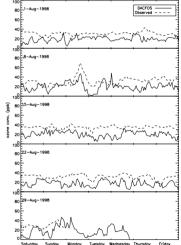
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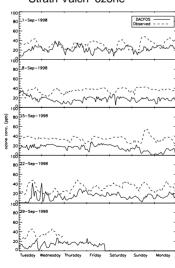


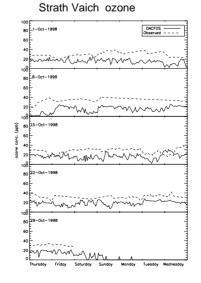


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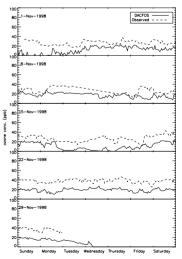




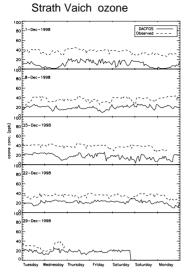


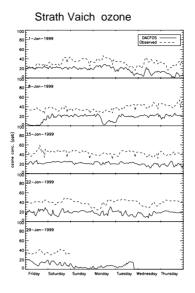


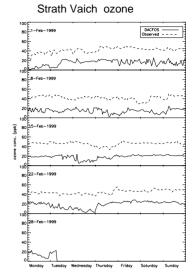
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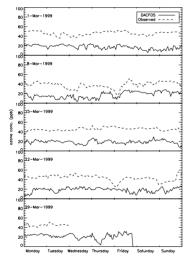
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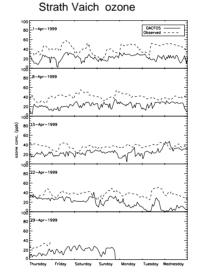


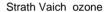


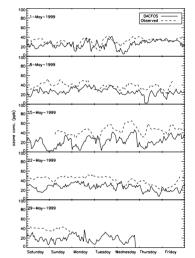


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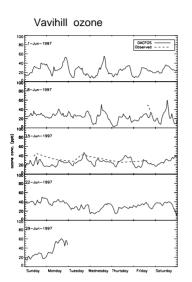


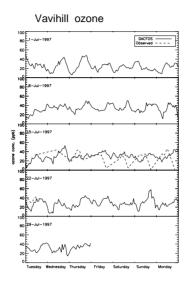


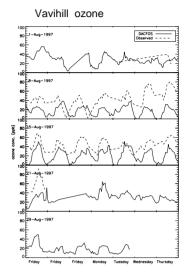


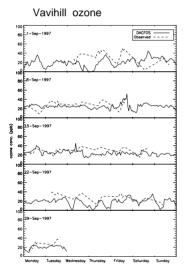


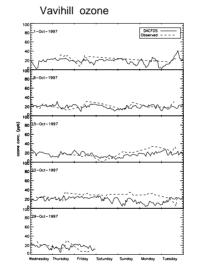
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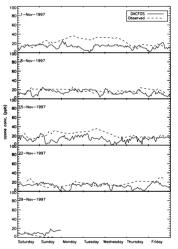


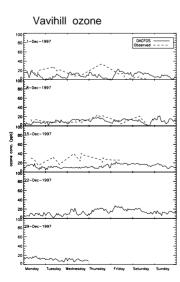


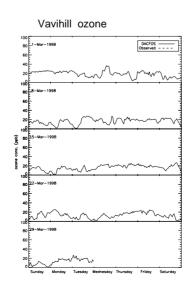




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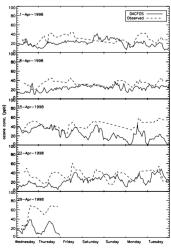


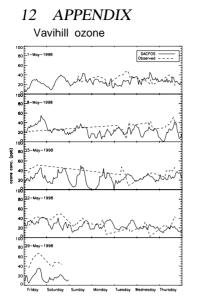


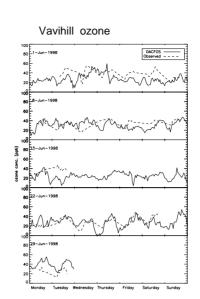


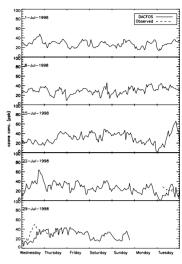
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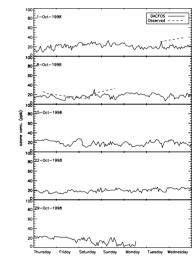


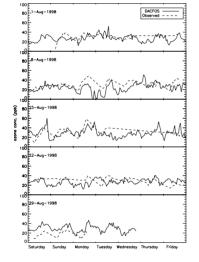




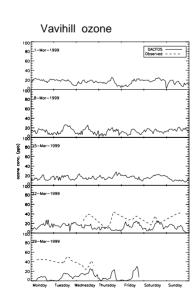


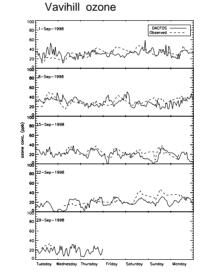
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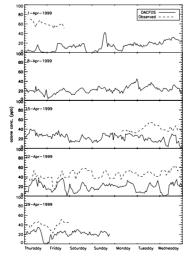


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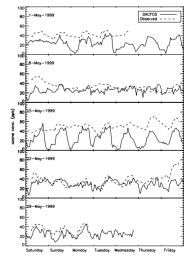




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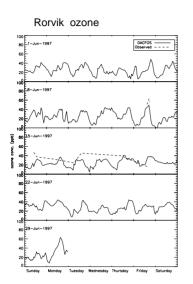
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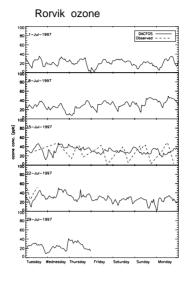


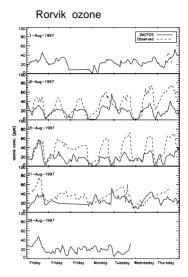
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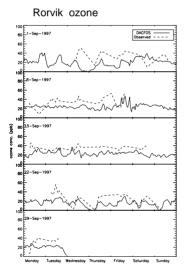
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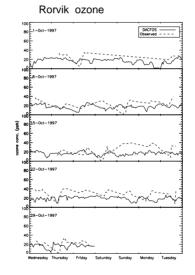
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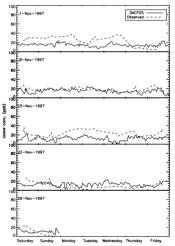


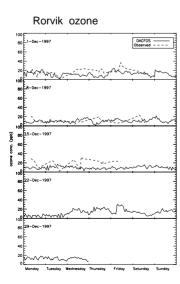


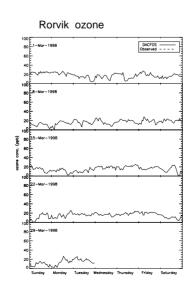




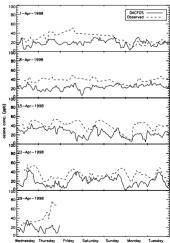
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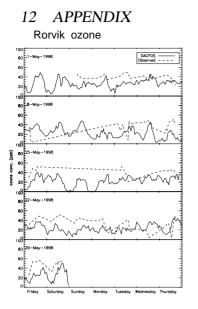


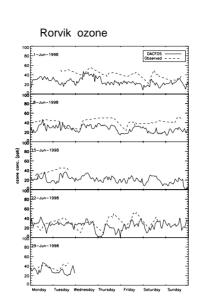


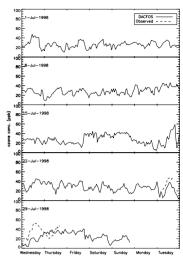


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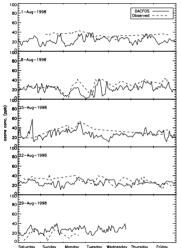


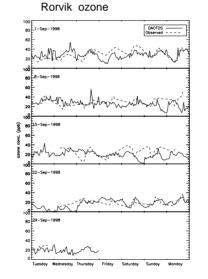


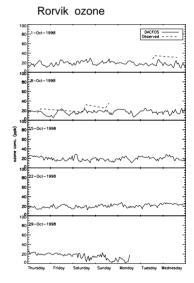




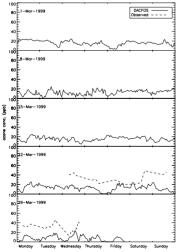
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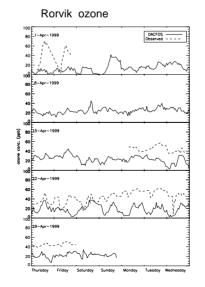




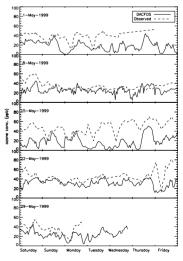






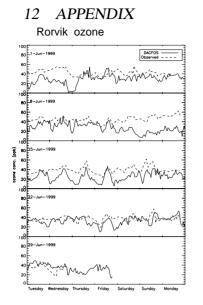


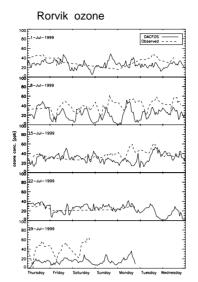




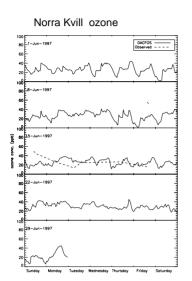
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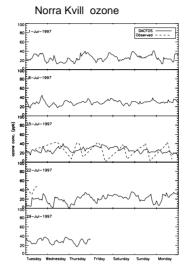
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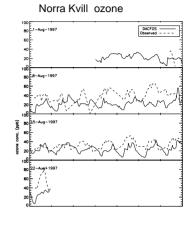


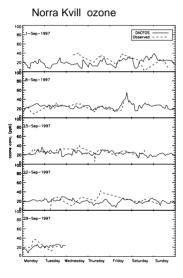


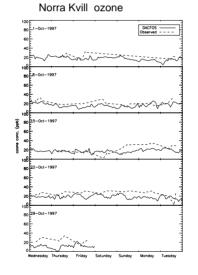
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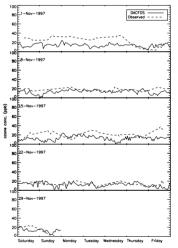




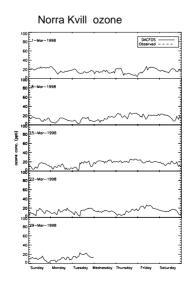




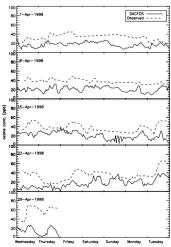
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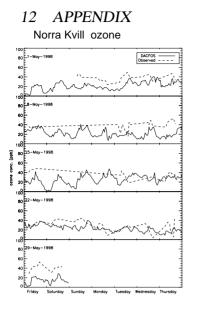


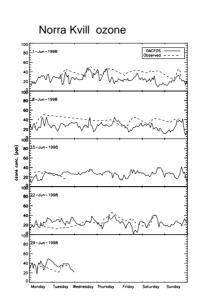
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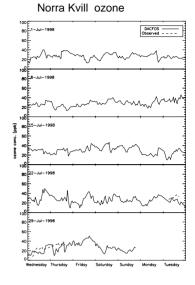


Norra Kvill ozone

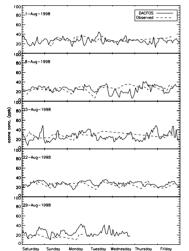


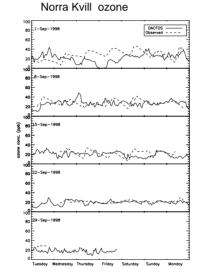


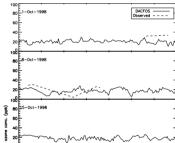




Norra Kvill ozone







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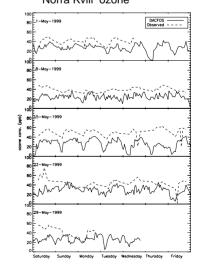
Oct-1998

80 29-Oct-1998

Norra Kvill ozone

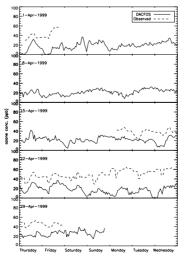


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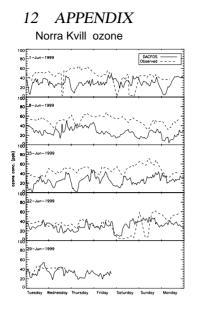


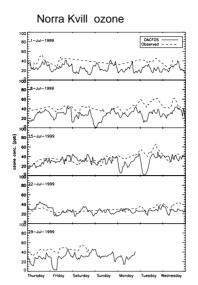
Norra Kvill ozone -Mar - 1999 DACFOS ----60 108 80 8-Mar-1999 60 20 Mymm MA MAR A 108 Mor - 1999 8 ZONE 80 - 1999 40 20 108 -Mar - 1999 80 ٣ m

Norra Kvill ozone

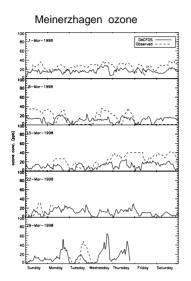


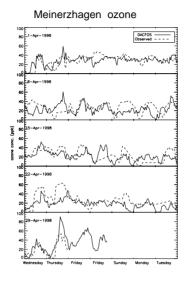
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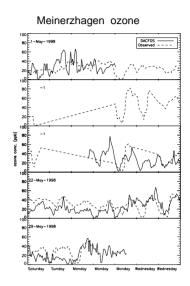


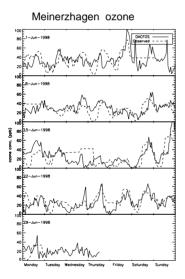


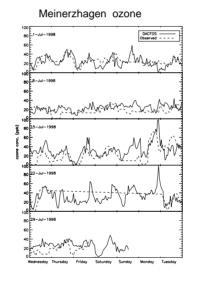
12.1.10 Meinerzhagen NR5

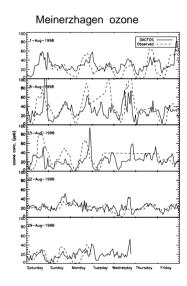


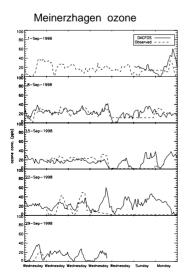


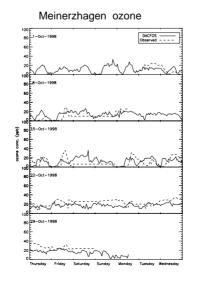




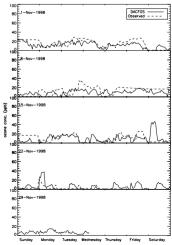








Meinerzhagen ozone



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