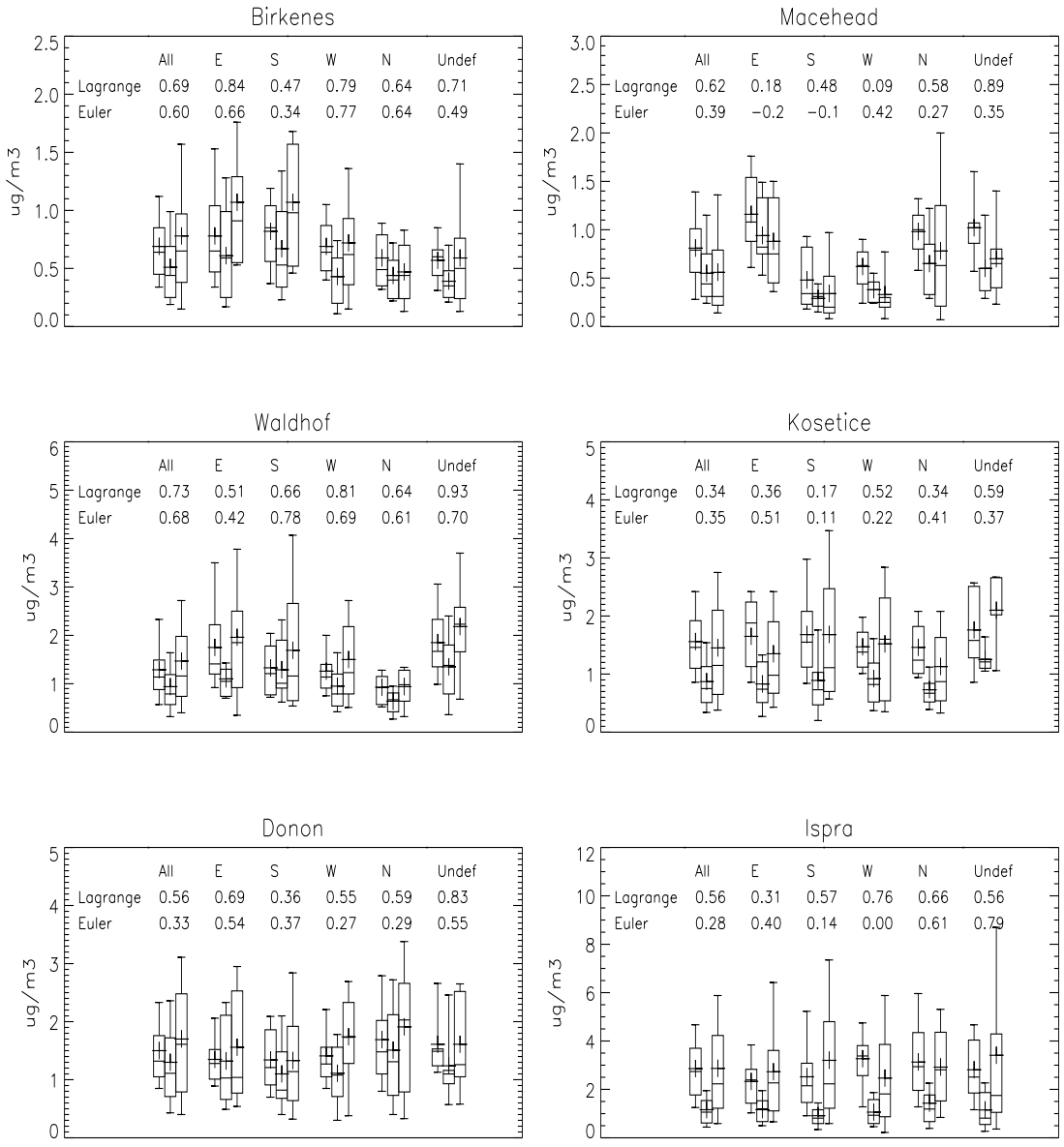

Figure 4.6: Box- and whisker-diagrams showing percentiles and mean formaldehyde concentrations as observed (left bars) and calculated with the Lagrangian model (mid bars) and the Eulerian model (right bars) for 4 transport sectors. The markers indicate the 10- and 90-percentiles (lower and upper bonds), 25- and 75-percentiles (lower and upper boundaries of boxes), medians (horizontal line inside boxes) and means (crosses). The numbers on top give the weighted linear correlation coefficients between modelled and measured concentrations within each sector for the two models, respectively.

4.6 Summary.

Measurements of VOC and CO have been used to evaluate the performance of the EMEP Eulerian and Lagrangian oxidant models. Compared to the last years preliminary comparison the present results with a further developed Eulerian oxidant model is much more encouraging for the Eulerian model and clearly shows that the model development has been successful. Still, the correlation with the measured values are in general somewhat better for the Lagrangian model. The average measured concentration level is, however, closer to the Eulerian model although the Eulerian model in elevated episodes give too high concentration spikes. As aldehydes are directly linked to the atmospheric chemical condition (OH-concentration, dissociation etc.) the somewhat better performance for the Lagrangian model (with regard to correlation with alde-

hyde measurements) suggests that the chemical reaction mechanism applied in the Lagrangian model may give a better description than the one in the Eulerian model. Part of this could be due to isoprene which have been shown to be crucial for the formaldehyde levels calculated in summer. The generally higher concentration levels calculated be the Eulerian model than the Lagrangian model reflects the finer vertical resolution in the 3d-model than the one-layered Lagrangian model. Furthermore, the results for separate transport sectors indicate that the Eulerian model performs better than the Lagrangian model in meteorological situations characterised by anticyclonal flow or wind shear situations; however, the amount of data used in the study is too small to draw firm conclusions.

The two models performed very similar when compared with measurements of CO both with regard to the general concentration level as well as the correlation with the observations. This is a further indication that the differences seen in the modelled aldehydes are caused by differences in the chemical reaction schemes.

As a test of the modelling of anthropogenic VOC emissions, the models were compared with measurements of the sum of light hydrocarbons. This revealed fairly large differences between the two models. The Eulerian model tend to give higher calculated sum of emitted hydrocarbons than the Lagrangian model, particularly in the period August-October. The reason for this discrepancy is not clear. Differences in vertical exchange and mixing are possible reasons for the discrepancies.

4.7 References

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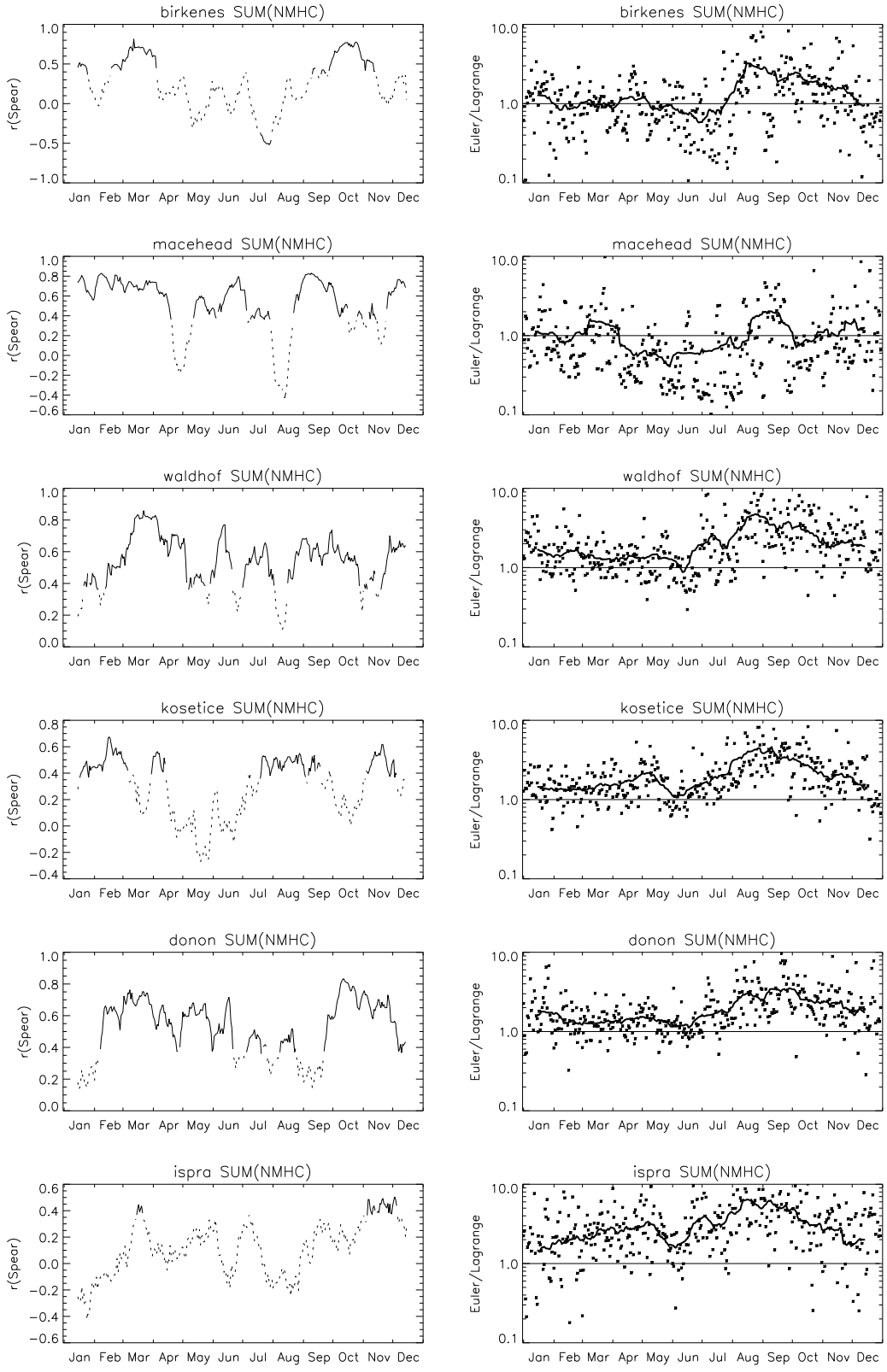

Figure 4.7: Linear correlation coefficients and ratios of the average concentrations between the Eulerian and Lagrangian model for the sum of hydrocarbons based on 30 days running data for six monitoring sites in 1996. Full and dashed lines mark correlations significantly and not significantly different from zero (95% level), respectively.

Chapter 5

Long-term variations in ozone peak values 1989-1998

Anne-Gunn Hjellbrekke and Sverre Solberg

5.1 Introduction

The meteorological variability from one year to another is known to have a major effect on the severity and extent of elevated ozone episodes in Europe. Simpson et al. (1997) found that large "trends" in modelled ozone (AOT40 or mean of daily max) in the period 1985-1995 at Arkona were almost entirely driven by meteorological changes. Furthermore, trend estimates in measured ozone is complicated by occasional shifts in technical equipment, monitoring procedures and changes in the surrounding environment, such as road traffic, growing trees etc. On the other hand the trend in anthropogenic emissions varies substantially between the European countries. Whereas Germany has experienced a 40% reduction in NOx emissions during a 10-years period according to official data, other countries have had constant emissions. Based on this, a preliminary inspection of the long-term variations in ozone peak values has been done and is presented below. Trends studies and compliance monitoring will probably be even more important in the years to come. The aim of this presentation is to give an overview and some suggestions for further research.

5.2 Ozone peak values 1989-1998

Based on the available EMEP ozone monitoring data for the period 1989-1998 the 99-percentiles of hourly data for the summer half year were calculated for each station. The results were used to produce annual kriged maps which are shown in Figures 5.1-5.2. In addition to the variation in the 99-percentiles these maps also indicate the development in the EMEP ozone monitoring network during this 10-years period. Germany, UK and Scandinavia have been covered during the whole period, whereas the network has gradually expanded eastwards and southwards. By the end of this period, in 1998, there are still large gaps in the network, most noticeably in France and southeast Europe. Note also that the number of sites are very few in some areas giving uncertain interpolated fields.

The annual 99-percentile maps all show a maximum area in central Europe with varying levels in UK and Scandinavia from year to year. The level and extent of the maximum area vary considerably from one year to another. The 99-percentiles in southern UK is particularly variable with the highest values in 1989, 1990 and 1995, presumably reflecting the variations in meteorological situations. The values at the Iberian peninsula also varies substantially; however, as noted below, some of the Spanish sites are located at locally influenced suburban sites, and not well representative for a larger region. Based on these maps only, it is not possible to identify any trends in the 99-percentiles of ozone during this period.

According to the officially reported data there has been a 17% reduction in anthropogenic emissions of NOx and VOC in Europe as a whole during the period 1987-1997 (Mylona, 1999).

There are, however, large differences between the countries. Countries presumably most important for ozone episodes in North Europe, such as Poland, Germany and UK, have experienced emission reductions of 30-40% whereas in other countries as e.g. France and Italy the NOx emissions have been almost constant during 1987-1997. Thus, the largest trends in surface ozone should be expected in the North. On the other hand, in Scandinavia the ozone episodes are even more dependent on the occurrence of certain meteorological situations which varies considerably from one year to another. The best area for identifying effects of reductions in European emissions should then be found as a compromise between the inter-annual meteorologically induced variations and the emission induced variations, and that could e.g. be in northern parts of Germany/Poland. Ideally, models should be used to identify regions with the strongest "ozone signal" from the anthropogenic emissions compared to the meteorology. Trend analyses of measurements should then be applied at monitoring sites in these regions.

Another highly important factor in trend studies is the quality of the monitoring data. A study of long-term ozone data at a few Nordic sites (Roemer et al., 1998) indicate that changes in technical procedures and equipment may represent a large problem for trend assessments. Thus, a knowledge of the QA/QC and the history of this is required. In 2000 EMEP/CCC distributed a questionnaire regarding the ozone monitoring to all laboratories, and the results are presented in detail by Aas et al. (2000). The main results are summarised in Table 5.1. These results show that whereas the technical procedures (at present) seem to be good at the sites which have replied to the questionnaire, the level of local emissions vary considerably. Note also that the tabulated results are the laboratories' own views. Thus, the opinion of what is defined as "local", "negligible", "little" etc. will be subjective. Furthermore, whereas some laboratories included very detailed reports on the local conditions, the documentation from other laboratories was very sparse. A fairly detailed documentation of local conditions was included for the Spanish sites, showing that several of these sites are exposed to local traffic emissions. A number of the sites will, however, be moved very soon to more rural areas in Spain.

Table 5.1: Overview of surroundings, inlet heights and calibration/maintenance routines for EMEP's ozone stations

Station	Veg.[1]	local[2] sources	Inlet ht.(m)	L[3]	Tmain[4]	Tcal[4]	T-TSF[4]	NIST[4]
AT02 Illmitz	g; wy; w	l	3.2	0.5-0.8	2w	d	4m	EMPA, y
AT04 St. Koloman	me; f	l	3.2	0.5-0.8	2w	d	4m	EMPA, y
AT05 Vorhegg	me: f	l	3.2	0.5-0.8	2w	d	4m	EMPA, y
CH02 Payerne	g; ar	l	4	6	2w/y	d	3m	EMPA, 4m
CH03 Tänikon	ar	l tr	4	6	2w/3m	d	3m	EMPA, 4m
CH04 Chaumont	g	n	4	6	2w/y	d	3m	EMPA, 4m
CH05 Rigi	g	n	4	6	2w/3m	d	3m	EMPA, 4m
CZ01 Svratouch	g	n	3.5	1.7	2w	6m	y	KLI Libus, y
CZ03 Kosetice	g	n	3.5	1.7	2w	6m	y	KLI Libus, y
DE all stations							6m	EMPA, y
DK05 Keldsnor	t	n	3.6	3	w	d		ITM, y
DK31 Ulborg								
DK32 Fredriksborg								
EE09 Lahemaa	g	n	4	5	m/y	m	†	†
EE11 Vilsandy	g	n	4	5	m/y	m	†	†
ES01 San Pablo	m; f	l	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES03 Roquetas	f; ba	tr	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES04 Logrono	fa; f	m tr	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES05 Noia	bu; f	tr	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES07 Viznar	f; fa	l tr	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES08 Niembro	w; fa	l tr	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES09 Campisábalos	cf	n	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES10 Cabo de Creus	w; f	tr	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES11 Barcarrota	f; w	n	3.5	3.7	15d	15d	6m	CarlosIII, 6m
ES12 Zarra	f; u.s	n	3.5	3.7	15d	15d	6m	CarlosIII, 6m
FI09 Utö	treeless	sbtr	5	4	3m	3m	3m	FMI, y
FI17 Virolahti	f; g		5	4	3m	3m	3m	FMI, y
FI22 Oulanka	f; bh		5	4	3m	3m	3m	FMI, y
FI37 Ähtäri	cf; bh; w		5	4	3m	3m	3m	FMI, y
FR08 Donon	f	n	7,16,30,44	60	15d/m	15d	3m	LNE, 3m
FR09 Revin	f	n	2.5	3	w/15d	w	2m	LNE, 3m
FR10 Morvan	f	n	3	5	m	w	6m	LNE, 3m
FR13_Peyrusse Vieille	g	n	4	6	15d/6m	15d	6m	LNE, 3m
FR14 Montandon	f; gr	n	2.5	5	15d/m	3d	3m	LNE, 3m
GB02 Eskdalemuir	g		3	4	3m/6m	3m	3m	NPL UK, 3
GB06 Lough Navar	f		2,5	4	3m/6m	3m	3m	NPL UK, 3
GB13 Yarner Wood	h		5	4	3m6m	3m	3m	NPL UK, 3
GB14 High Muffles	f		3	4	3m/6m	3m	3m	NPL UK, 3
GB15 Strath Vaich	m		3	4	3m/6m	3m	3m	NPL UK, 3
GB31 Aston Hill	fi		3	4	3m/6m	3m	3m	NPL UK, 3
GB32 Bottesford	fa		5	4	3m/6m	3m	3m	NPL UK, 3

continued on next page

Station	Veg.[1]	local[2] sources	Inlet ht.(m)	L[3]	Tmain[4]	Tcal[4]	T-TSF[4]	NIST[4]
GB33 Bush	g; t		8	4	3m/6m	3m	3m	NPL UK, 3
GB34 Glazerbury	g		3	4	3m/6m	3m	3m	NPL UK, 3
GB35 Great Dun Fell	fa; t		2	40	3m/6m	3m	3m	NPL UK, 3
GB36 Harwell	fi		3	4	3m/6m	3m	3m	NPL UK, 3
GB38_Lullington Heath	h		3	4	3m/6m	3m	3m	NPL UK, 3
GB39 Sibton	fi;f		3	4	3m/6m	3m	3m	NPL UK, 3
GB43 Narberth	fi	s	3	4	3m/6m	3m	3m	NPL UK, 3
GB44 Somerton	fi		3	4	3m/6m	3m	3m	NPL UK, 3
GB45 Wicken Fen	fi; t		2,5	4	3m/6m	3m	3m	NPL UK, 3
GR01 Aliartos								
HU02 K-puszta	f	n	10	12	w	6m	6m	CHMI, y
IE031 Mace Head								
IT01 Montelibretti	g	s tr	2	1.5	15d[5]	15d	3m	IAP, 3m
IT04 Ispra								
LT15 Preila								
LV10 Rucava	fa; t	n	3	8	y/4m	d	no	ITM, y
NL09_Kollumerwaard								
NL10 Vreedepeel								
NO01 Birkenes	f, w	n	2	3	3m	w	y	ITM, y
NO15 Tustervatn	f; w	n	2	3	3m	w	y	ITM, y
NO39 Kaarvatn	f; gr	n	2	3	3m	w	y	ITM, y
NO41 Osen	f; w	n	2	3	3m	w	y	ITM, y
NO42 Zeppelinfjellet	h; w	n	2	3	3m	w	y	ITM, y
NO43 Prestebakke	f	n	2	3	3m	w	y	ITM, y
NO45 Jeløya	f, g	sbtr	2	3	3m	w	y	ITM, y
NO48 Voss	f	n	2	3	3m	w	y	ITM, y
NO52 Sandve	f; fa	n	2	3	3m	w	y	ITM, y
NO55 Karasjok	f; h	n	2	3	3m	w	y	ITM, y
NO56 Hurdal	f	tr	2	3	3m	w	y	ITM, y
PL02 Jarczew							3-4m	NBS, y
PL03 Sniezka							3-4m	NBS, y
PL04 Leba							6m	NBS, y
PL05 Diabla Gora	me; f	l	4.1	5	3m	w[6]	3m	CHMI, y
PT04 Monte Velho								
RU01 Janiskoski								
RU13 Pinega								
RU16 Shepeljovo								
SE02 Rörvik	g	n	5	6	4m	4m	4m	ITM, y
SE11 Vavihill	g	n	5	7	4m	4m	4m	ITM, y
SE12 Aspvreten	f	n	5	6	4m	4m	4m	ITM, y
SE13 Esrange	h; t	n	4	5	4m	4m	4m	ITM, y
SE32 Norra Kvill	g; f	n	5	7	4m	4m	4m	ITM, y
SE35 Vindeln	f	n	3	4	4m	4m	4m	ITM, y
SI08 Iskrba	c.f; g	l tr	5.5	5	m/6m	d	6m	CHMI y
SI31 Zavodnje	f,g p	s	2.5	1.5	6m/y	d	y	CHMI y
SI32 Krvavec	g; c.f	s, pp	10	8	4m	d	4m	CHMI y
SI33 Kovk	f; g; p	s, pp	2.5	1.5	m/y	d	y	CHMI y
SK02 Chopok	g	n	3.5-4	2-2.5	m/y	d	6m	CHMI y
SK04 Stara Lesna	c.f; g	l tr	3.5-4	2-2.5	m/y	d	6m	CHMI y
SK06 Starina	f	n	3.5-4	2-2.5	m/y	d	6m	CHMI y

Notes:
1) ar: arable; g: grass; ba: built up area; bh: bog and heather; bu: bush; cf: coniferous forest; fa: farmland; f: forest; fi: field; gr: graze; h: hillside; m: moor; me: meadow; p: pasture; t: some trees; w: water; wy: wine yard; us: unproductive soil;
2) Local sources of NOx - n: negligible; l: little; s: some; m: much; tr: traffic; sbtr: some boat traffic; pp: power plant
3) L = Length of sample line (m).
4) Tmain = Maintenance interval, Tcal = Calibration interval, T-TSF = Transfer standard interval NIST = NIST location and frequency; d: daily. w: weekly; m; monthly; y: yearly
5)Some maintenance not performed on routine basis
6) span checks not performed
†: No transfer standard, help from FMI yearly

5.3 Summary and recommendations

As indicator for episodes of surface ozone, kriged maps of 99-percentiles based on hourly ozone monitoring values have been prepared for each year in the period 1989-1998. The maps show large inter-annual variations, and it is not possibly to identify any clear trends based on these maps alone. The presented kriged maps clearly shows the importance of increasing the number of ozone monitoring sites, both to get a better regional cover and also to reduce the uncertainty in the interpolated fields. A summary of the results of a questionnaire regarding the ozone