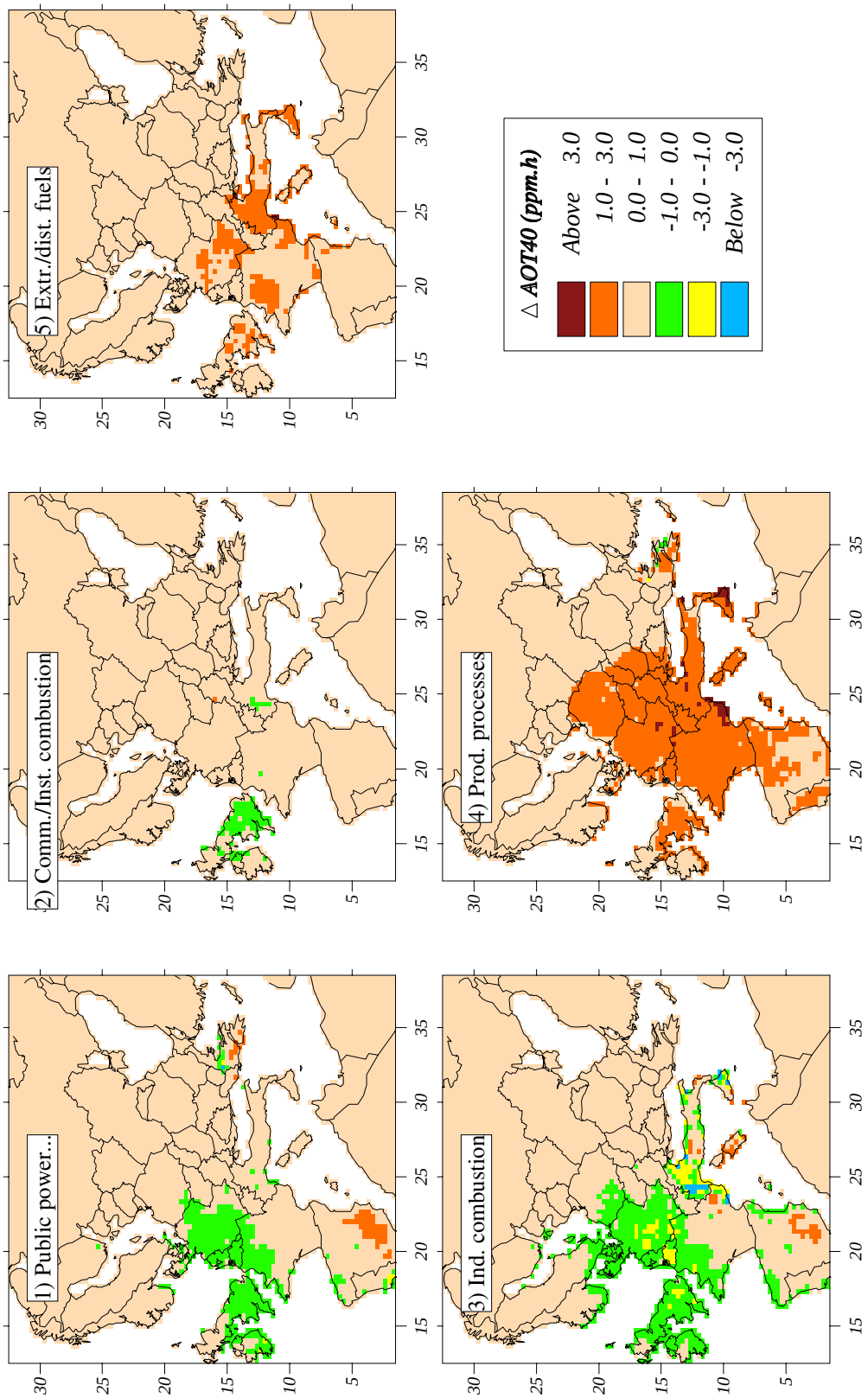

Figure 4.1: Effect of reducing emissions from SNAP sectors (1)-(5) on $AOT40_f$. Calculations with Eulerian model. Base emissions from 2010 (INFOS trend), meteorology of April-Sept. 1996. (Note that negative $AOT40_f$ means that AOT40 has increased when emissions from sector removed.)

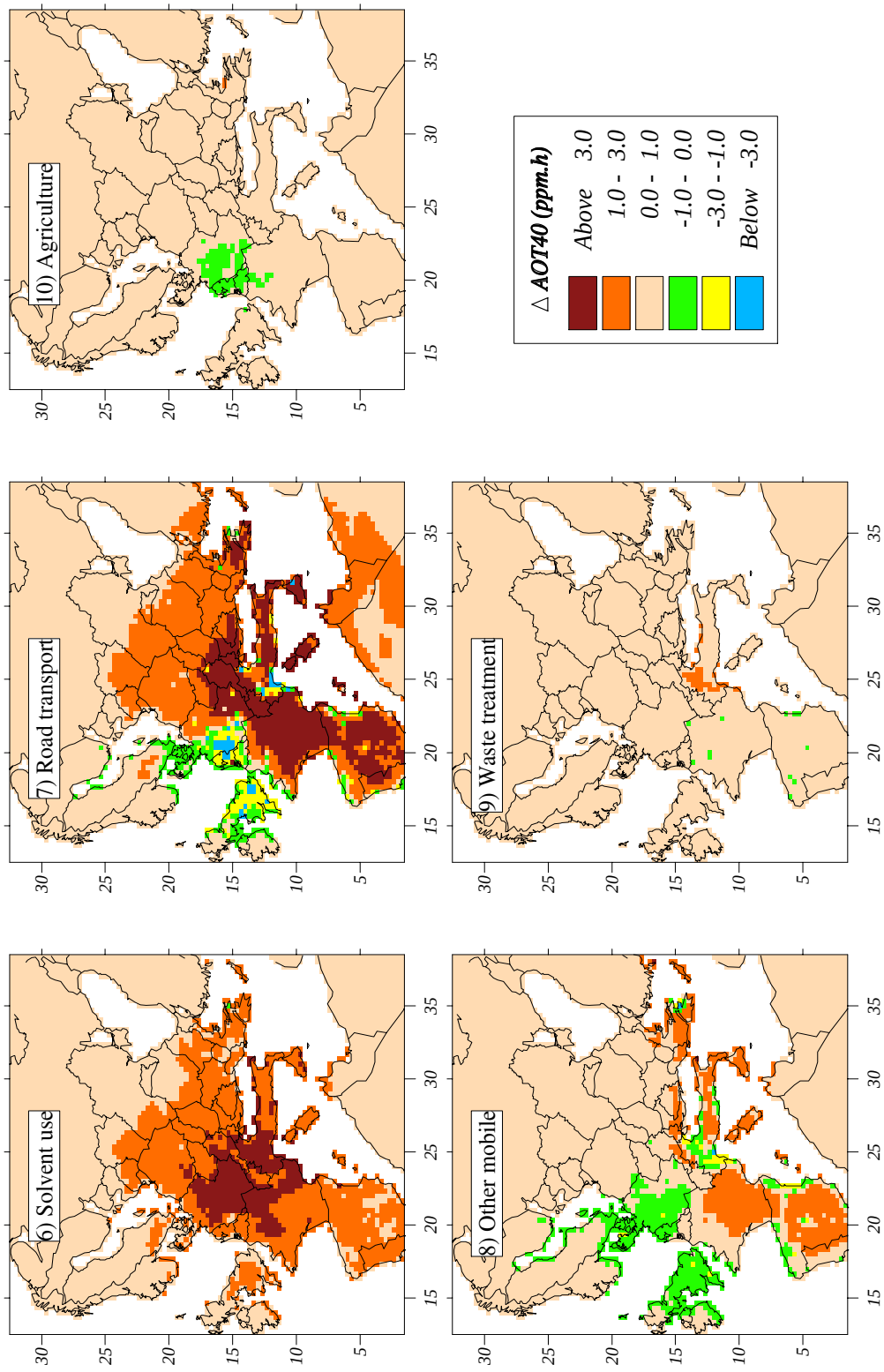

Figure 4.2: Effect of reducing emissions from SNAP sectors (6)-(10) on $AOT40_f$. Calculations with Eulerian model. Base emissions from 2010 (INFOS trend), meteorology of April-Sept. 1996. (Note that negative $AOT40_f$ means that AOT40 has increased when emissions from sector removed.)

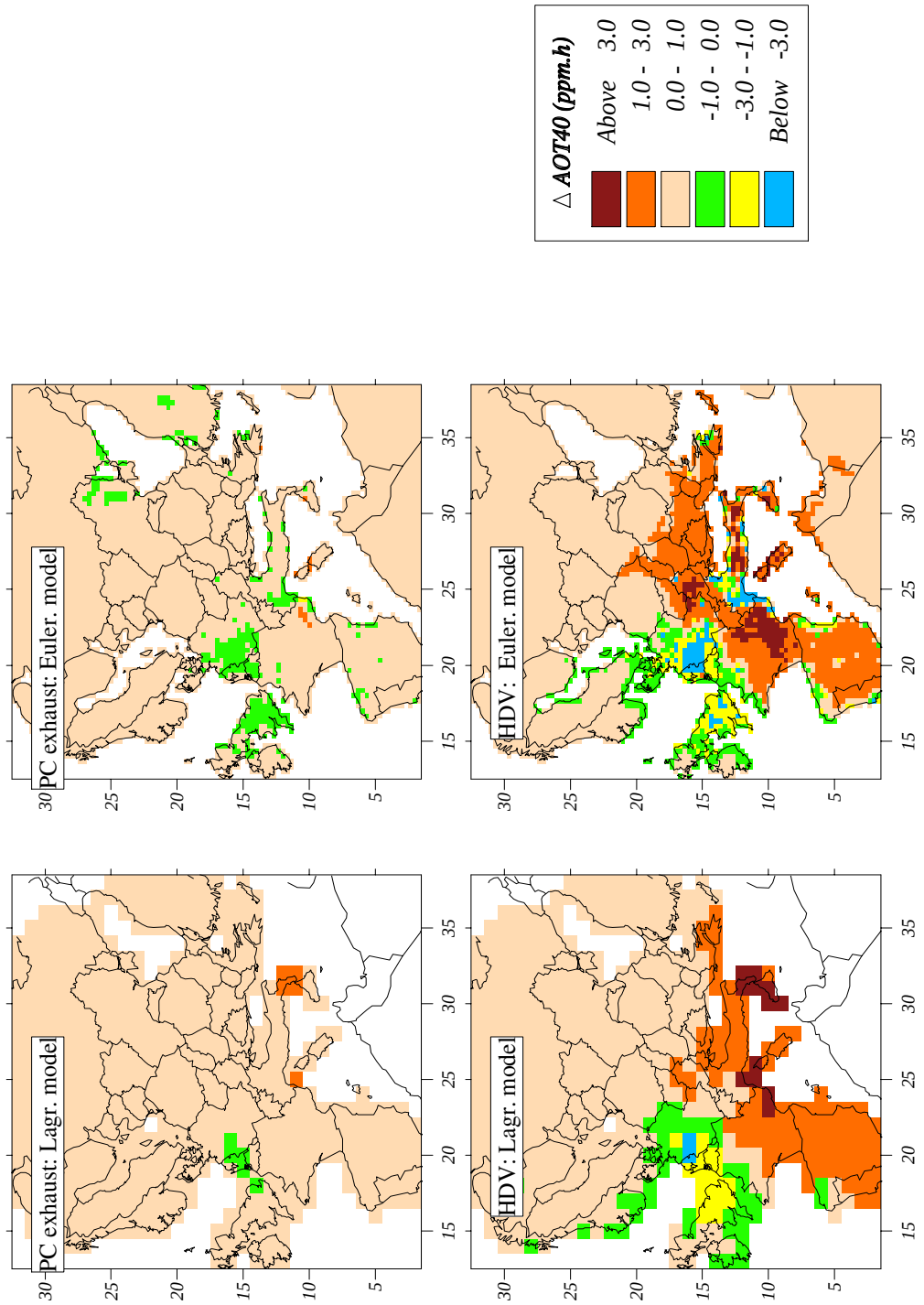

Figure 4.3: Comparison of Lagrangian and Eulerian model predictions for the effect of reducing emissions from SNAP 7.1: passenger car exhaust (top) and SNAP 7.3: heavy-duty vehicles (bottom). Plots show the reduction in $AOT40_f$, as for Figs 4.1,4.2

Chapter 5

Source-receptor calculations

D. Simpson and J.E. Jonson

5.1 Introduction

Source-receptor (S-R) relationships give the change in ozone in each receptor country (or grid square) resulting from a change in emissions of either NO_x or VOC from each emitter country. They are generated for each country by reducing either NO_x or NMVOC emissions (anthropogenic) by a given percentage from that country, re-running the oxidant model, and comparing the resulting fields of ozone or AOTs with the base-case fields.

The first full set of such S-R matrices was presented in Simpson et al. (1997), where calculations were performed with the Lagrangian oxidant model for a 5 year period: 1989, 90, 92, 93, 94. A number of matrices with different assumptions (including 40% reductions) were calculated in order to supply results needed for the IIASA RAINS-ozone model (Schöpp et al., 1999). Extensive discussions of the behaviour of source-receptor matrices for ozone, and of the extent of linearity of these, have been presented elsewhere (Simpson, 1991, Simpson and Malik, 1996, Simpson et al., 1997).

In this chapter we present the results of two new studies. Section 5.2 presents an updated set of S-R matrices, based upon new emission estimates, and now using meteorology from the 5-year period 1992-1996. Section 5.3 presents a first comparison of the results of the Lagrangian and Eulerian models for a number of countries.

5.2 Updated calculations with Lagrangian model

The basic methodology used to calculate S-R matrices is similar to that of Simpson et al. (1997), although here we have only performed calculations for 40% emission reductions. The base-emission case is for the year 2010, using the EMEP emissions estimates as given in chapter 2. The meteorology for the 5-year calculations is now from 1992, 93, 94, 95 and 96. The main advantage of updating these S-R relationships is probably that the estimates of the starting situation, the year 2010 emissions, have been improved, especially since these estimates have formed the basis of negotiations for the 2nd NO_x Protocol and the EU acidification and ozone strategy work (Amann et al., 1999).

The results of the updated S-R calculations are given in Tables 5.1-5.6 for 6-monthly mean of daily max. ozone, $AOT40_c$, and AOT60. For the country codes used in these tables, see chapter 1, section 1.4. These tables, also that for $AOT40_f$ (not shown) and indeed the country-to-grid results from which they are constructed will be made available as ascii files on the EMEP web-site, http://www.emep.int.

The matrices will not discussed further here as the results are very similar to those previously reported in (Simpson et al., 1997).

Table 5.1: Source-receptor relationships for 6-monthly mean of daily max. ozone - NOx

(Emission base-year 2010, meteorology: from 5 years - 1992-1996)

emitters

Country	AL	AT	BY	BE	BA	BG	HR	CZ	DK	EE	FI	FR	DE	GR	HU	IS	IE	IT	LV
Albania	105	6	4	0	62	88	49	5	0	0	0	16	-6	162	36	0.01	0	231	0
Austria	0	22	4	-15	5	1	27	-26	-4	0	1	84	-27	2	43	0.16	0	-23	1
Belarus	0	0	157	-3	1	2	2	-8	1	19	33	0	-16	1	7	0.26	0	3	39
Belgium	0	3	1	-320	0	0	1	-9	-12	0	0	165	-389	0	3	1.13	3	6	1
Bosnia and Herzegovina	8	19	4	-5	97	25	112	5	0	0	0	19	-9	20	97	0.07	0	226	0
Bulgaria	7	2	13	-3	7	118	9	-4	0	1	1	3	-13	55	33	0.07	0	45	1
Croatia	3	25	5	-7	61	13	122	-5	-1	0	0	34	-15	10	110	0.08	0	254	0
Czech Republic	0	36	11	-32	3	0	16	-207	-6	0	2	51	-137	1	38	0.34	0	12	3
Denmark	0	-1	12	-22	0	0	0	-14	-167	3	7	3	-70	0	1	2.75	2	0	8
Estonia	0	0	33	-5	0	1	0	-4	2	51	95	0	-9	0	1	0.30	0	0	54
Finland	0	0	8	-1	0	0	0	-1	1	13	181	0	-4	0	0	0.63	0	0	10
France	0	1	0	-44	0	0	1	-7	-5	0	0	527	-58	0	1	0.58	4	53	0
Germany	0	11	5	-75	0	0	3	-39	-18	0	1	113	-309	0	7	0.88	1	-14	2
Greece	31	2	6	0	10	149	10	1	0	0	1	13	-5	333	17	0.02	0	179	1
Hungary	1	20	15	-10	17	11	49	-34	-2	0	1	23	-45	4	150	0.15	0	84	1
Iceland	0	0	0	-12	0	0	0	0	-2	0	0	21	-30	0	0	4.31	19	0	0
Ireland	2	6	1	-2	22	5	39	-3	-1	0	0	165	-11	13	26	0.07	0	60	0
Italy	0	0	57	-4	0	1	1	-4	-4	40	63	0	-10	0	3	0.41	0	2	73
Latvia	0	0	69	-5	0	1	1	-7	-7	18	35	1	-15	0	5	0.44	0	1	49
Lithuania	0	3	1	-121	0	0	1	-15	-17	0	0	391	-217	0	3	0.90	3	10	1
Luxembourg	0	2	2	-219	0	0	0	-14	-13	0	1	53	-491	0	2	1.58	3	-3	1
Netherlands	0	0	3	-3	0	0	0	-1	-7	2	22	0	-12	0	0	2.28	2	0	4
Norway	0	5	45	-18	2	2	5	-47	-9	3	8	11	-70	1	20	0.67	0	7	10
Poland	0	0	0	0	0	0	0	0	0	0	0	27	0	0	0	0.13	3	4	0
Portugal	0	2	53	-2	2	20	5	-5	0	3	7	2	-12	6	25	0.13	0	20	6
Republic of Moldova	2	5	23	-4	7	42	15	-9	-1	1	3	4	-19	10	65	0.09	0	40	2
Romania	0	0	13	0	0	1	0	0	0	6	23	0	0	0	0	0.05	0	0	6
Russian Federation	0	18	26	-15	8	5	25	-62	-4	1	2	19	-67	2	92	0.22	0	30	3
Slovakia	0	20	4	-9	21	5	78	-15	-3	0	0	51	-13	4	81	0.10	0	96	0
Slovenia	0	0	0	0	0	0	0	-2	0	0	0	92	-2	0	0	0.13	3	21	0
Spain	0	0	8	-5	0	0	0	-3	-12	6	49	0	-13	0	0	1.22	1	0	10
Sweden	0	2	0	-8	1	1	4	-3	-3	0	0	270	-29	1	4	0.13	0	-117	0
Switzerland	44	4	5	-2	24	125	19	0	0	0	0	7	-10	123	41	0.09	0	125	0
The FYR Macedonia	2	0	7	0	0	24	0	0	0	0	1	2	-1	61	2	0.	0	26	1
Turkey	0	1	66	-2	1	14	3	-6	0	5	11	1	-12	4	13	0.12	0	8	10
Ukraine	0	0	0	-34	0	0	0	-3	-3	0	0	38	-65	0	0	4.19	23	1	0
United Kingdom	17	10	5	-4	49	59	49	-2	0	0	0	8	-15	36	91	0.07	0	127	0
Yugoslavia	1	2	17	-9	3	8	5	-6	-2	4	19	46	-25	11	9	0.39	1	17	6
Europe*																			

receivers

Country	LT	LU	NL	NO	PL	PT	MD	RO	RU	SK	SI	ES	SE	CH	MK	TR	UA	GB	YU
Albania	1	0	-1	0	13	1	3	72	14	9	1	17	0	0	36	30	52	-1	114
Austria	3	0	-24	2	-15	0	0	15	7	12	7	12	0	3	0	0	19	-26	9
Belarus	61	0	-10	9	54	0	3	21	191	3	0	1	23	0	0	4	178	-10	3
Belgium	2	0	-415	15	-1	0	0	3	3	1	0	30	0	6	0	0	6	-424	1
Bosnia and Herzegovina	1	0	-6	1	13	1	1	68	9	20	6	15	0	0	6	6	30	-10	117
Bulgaria	3	0	-4	0	11	0	15	234	70	9	0	4	1	0	6	42	253	-8	49
Croatia	2	0	-9	1	0	1	0	53	8	20	10	24	0	0	2	3	27	-15	68
Czech Republic	7	0	-56	5	-60	0	0	12	15	19	5	8	1	1	0	0	33	-66	6
Denmark	12	0	-55	66	35	0	0	1	25	0	0	3	-1	0	0	0	5	-106	0
Estonia	33	0	-9	25	24	0	0	4	142	0	0	0	74	0	0	1	34	-18	0
Finland	8	0	-3	38	7	0	0	1	86	0	0	0	76	0	0	0	9	-5	0
France	0	0	-71	4	-6	3	0	0	0	0	0	133	-1	9	0	0	1	-120	0
Germany	4	0	-157	14	10	0	0	3	10	3	1	15	0	-1	0	0	14	-157	1
Greece	1	0	-1	0	9	0	6	85	51	5	0	12	0	0	11	221	165	-1	37
Hungary	4	0	-19	2	-10	0	1	87	18	30	5	10	0	2	1	2	81	-22	40
Iceland	0	0	-26	10	1	0	0	0	2	0	0	3	0	0	0	0	2	-196	0
Ireland	0	0	-3	0	-1	3	0	12	2	6	4	60	0	-6	1	4	8	-6	18
Italy	58	0	-11	20	41	0	1	7	142	1	0	0	60	0	0	1	52	-20	1
Latvia	81	0	-17	19	73	0	1	9	106	2	0	1	47	0	0	1	65	-28	2
Lithuania	2	-10	-160	8	-4	1	0	1	1	2	0	42	-2	12	0	0	3	-184	1
Luxembourg	2	0	-642	26	4	0	0	2	6	1	0	18	2	0	0	0	11	-475	0
Netherlands	5	0	-9	112	5	0	0	0	19	0	0	0	42	0	0	0	2	-26	0
Norway	28	0	-40	13	-15	0	1	16	49	9	0	4	11	0	0	1	83	-53	4
Poland	0	0	-1	0	0	159	0	0	0	0	0	345	0	0	0	0	0	0	0
Portugal	13	0	-5	2	38	0	36	165	107	8	0	1	4	0	0	22	427	-7	8
Republic of Moldova	6	0	-7	1	15	0	15	262	53	21	1	4	1	0	2	15	238	-11	33
Romania	5	0	0	3	3	0	0	3	236	0	0	0	6	0	0	13	50	0	0
Russian Federation	9	0	-27	3	-76	0	1	43	24	21	3	7	1	2	0	1	105	-33	16
Slovakia	3	0	-13	1	-22	1	0	32	7	15	5	24	0	0	0	2	22	-17	27
Slovenia	0	0	-3	0	-2	99	0	0	0	0	0	734	0	1	0	0	0	-4	0
Spain	12	0	-12	78	10	0	0	1	36	0	0	0	108	0	0	0	5	-23	0
Sweden	0	0	-15	1	-1	0	0	1	0	2	2	30	-1	-74	0	0	2	-19	2
Switzerland	1	0	-2	0	14	0	4	117	19	12	1	10	0	0	32	22	71	-5	104
The FYR Macedonia	2	0	0	0	5	0	5	29	148	0	0	2	1	0	1	430	214	0	2
Turkey	18	0	-4	3	28	0	11	60	249	5	0	1	6	0	0	30	436	-6	5
Ukraine	0	0	-72	25	-1	0	0	1	3	0	0	6	1	0	0	0	4	-1019	0
United Kingdom	1	0	-6	0	12	0	3	141	13	19	2	8	0	0	10	8	55	-9	119
Yugoslavia	7	0	-18	11	5	7	2	22	135	2	0	53	11	0	0	35	80	-44	6
Europe*																			

Units: ppt per 40% emission reduction; Average over all land-areas of Europe

Table 5.2: Source-receptor relationships for 6-monthly mean of daily max. ozone - VOC

(Emission base-year 2010, meteorology: from 5 years - 1992-1996)

emitters

Country	AL	AT	BY	BE	BA	BG	HR	CZ	DK	EE	FI	FR	DE	GR	HU	IS	IE	IT	LV
Albania	58	10	4	4	17	20	21	15	1	0	0	21	38	52	12	0.	0	290	1
Austria	0	60	5	21	2	1	11	49	6	0	1	112	172	1	14	0.	1	306	4
Belarus	0	6	43	5	0	2	2	13	12	4	5	14	41	1	6	0.	1	17	32
Belgium	0	10	2	177	0	0	1	27	13	0	0	241	370	0	2	0.	14	20	3
Bosnia and Herzegovina	4	23	5	9	25	8	36	28	3	0	0	39	72	7	25	0.	1	327	3
Bulgaria	4	11	10	4	4	62	7	19	2	0	0	15	44	29	21	0.	0	44	4
Croatia	2	34	6	12	14	5	39	39	4	0	0	62	98	4	31	0.	1	411	4
Czech Republic	0	50	10	35	1	1	8	132	12	1	1	101	263	1	16	0.	3	104	7
Denmark	0	8	13	28	0	0	2	26	77	3	5	56	144	0	3	0.	9	14	15
Estonia	0	2	17	7	0	0	0	8	14	20	21	14	35	0	2	0.	2	6	44
Finland	0	0	5	2	0	0	0	2	6	6	16	5	15	0	0	0.	1	1	10
France	0	6	1	50	0	0	1	13	6	0	0	266	153	0	1	0.	6	67	1
Germany	0	26	5	69	0	0	2	57	17	1	1	155	378	0	5	0.	7	73	5
Greece	25	6	7	2	5	29	7	10	1	0	0	14	25	174	10	0.	0	123	2
Hungary	1	39	11	15	6	7	20	60	7	1	1	56	128	2	58	0.	1	161	6
Iceland	0	1	1	16	0	0	0	3	4	0	0	42	50	0	0	0.01	50	3	2
Ireland	1	24	2	7	6	3	20	21	2	0	0	121	58	4	12	0.	1	824	1
Italy	0	4	23	7	0	1	1	11	19	12	13	15	42	0	3	0.	2	10	64
Latvia	0	6	30	8	0	1	2	16	27	7	7	20	58	0	5	0.	3	17	56
Lithuania	0	11	2	118	0	0	1	29	12	0	0	249	368	0	1	0.	7	32	2
Luxembourg	0	9	4	134	0	0	0	32	18	0	1	178	384	0	3	0.	16	20	4
Netherlands	0	1	4	5	0	0	0	4	10	1	3	13	31	0	0	0.	2	2	5
Norway	0	18	19	22	1	2	5	56	23	3	4	49	153	1	14	0.	4	45	17
Poland	0	0	0	3	0	0	0	1	0	0	0	28	10	0	0	0.	2	3	0
Portugal	0	9	20	5	1	13	5	19	6	1	1	18	42	5	15	0.	1	26	9
Republic of Moldova	1	15	12	7	3	25	9	28	4	0	1	25	62	9	32	0.	1	51	5
Romania	0	0	4	0	0	0	0	1	1	1	2	1	4	0	0	0.	0	1	4
Russian Federation	0	36	14	20	3	5	12	81	10	1	2	60	143	1	43	0.	2	103	9
Slovakia	0	54	6	14	5	2	29	43	5	0	1	93	119	2	24	0.	1	521	4
Slovenia	0	0	0	6	0	0	0	3	1	0	0	62	22	0	0	0.	2	12	0
Spain	0	1	6	7	0	0	0	5	18	3	6	14	36	0	1	0.	2	3	11
Sweden	0	17	1	14	0	0	4	16	3	0	0	190	120	0	3	0.	1	320	1
Switzerland	24	10	5	4	9	33	11	15	1	0	0	15	38	53	14	0.	0	109	1
The FYR Macedonia	1	2	6	0	0	7	1	2	1	0	0	4	7	31	2	0.	0	13	2
Turkey	0	6	23	4	1	6	3	14	6	2	2	13	34	3	9	0.	1	19	11
Ukraine	0	2	2	39	0	0	0	9	8	0	1	86	103	0	1	0.07	25	6	3
United Kingdom	8	19	6	7	15	18	20	28	2	0	0	26	67	12	29	0.	0	152	2
Yugoslavia	1	6	7	10	1	3	3	11	5	1	2	39	50	5	5	0.	2	54	6
Europe*																			

receivers

Country	LT	LU	NL	NO	PL	PT	MD	RO	RU	SK	SI	ES	SE	CH	MK	TR	UA	GB	YU
Albania	0	0	4	0	29	0	2	25	14	9	1	4	1	1	9	19	36	13	45
Austria	2	1	19	2	86	0	0	8	8	18	6	5	6	37	0	1	24	52	5
Belarus	13	0	10	4	86	0	3	12	92	7	0	1	16	2	0	5	94	35	3
Belgium	1	4	144	7	46	1	0	4	6	3	0	13	9	10	0	0	10	425	1
Bosnia and Herzegovina	1	0	9	1	61	0	1	24	9	18	3	4	2	5	1	5	32	28	36
Bulgaria	2	0	6	1	45	0	7	73	50	15	0	1	2	1	2	45	106	19	34
Croatia	2	0	12	1	84	0	1	21	10	24	6	7	4	10	0	3	35	37	24
Czech Republic	4	2	40	4	183	0	1	9	17	27	3	5	10	16	0	0	44	101	5
Denmark	6	0	39	17	121	0	0	3	31	5	0	3	39	3	0	0	31	217	1
Estonia	9	0	10	6	46	0	1	3	62	2	0	0	34	1	0	1	30	56	1
Finland	2	0	4	4	14	0	0	1	24	1	0	0	20	0	0	0	9	24	0
France	0	2	45	3	24	3	0	1	2	2	0	59	4	16	0	0	4	172	0
Germany	2	3	76	6	103	0	0	4	13	9	1	8	14	22	0	0	22	210	1
Greece	1	0	3	0	23	0	4	32	42	7	0	2	1	0	3	163	78	10	21
Hungary	3	1	19	2	137	0	2	38	17	47	4	4	6	10	0	2	71	48	26
Iceland	0	0	22	5	13	1	0	0	4	0	0	4	2	0	0	0	4	268	0
Ireland	1	0	7	1	37	1	0	8	4	10	5	14	3	19	0	3	13	24	9
Italy	15	0	12	5	66	0	1	5	71	4	0	1	31	1	0	2	36	64	2
Latvia	25	0	17	7	105	0	1	8	63	7	0	1	32	2	0	3	46	70	2
Lithuania	1	10	79	5	41	1	0	2	4	2	0	16	9	16	0	0	6	217	1
Luxembourg	1	3	188	9	62	1	0	4	12	5	0	8	12	8	0	0	16	485	0
Netherlands	2	0	8	16	22	0	0	0	9	1	0	0	15	0	0	0	5	59	0
Norway	9	0	32	6	268	0	1	12	38	19	1	3	18	5	0	1	65	97	7
Poland	0	0	3	0	1	180	0	0	0	0	0	200	0	0	0	0	0	26	0
Portugal	4	0	7	2	75	0	19	68	75	13	0	1	5	1	0	25	187	24	9
Republic of Moldova	2	0	9	1	77	0	7	94	38	24	1	1	4	3	1	16	113	26	27
Romania	1	0	1	0	8	0	0	2	73	0	0	0	3	0	0	5	27	5	0
Russian Federation	5	1	24	3	211	0	2	25	23	53	3	3	9	9	0	1	79	63	14
Slovakia	2	1	13	1	93	0	1	13	9	22	14	7	6	24	0	2	30	36	11
Slovenia	0	0	6	0	5	53	0	0	0	0	0	263	0	2	0	0	0	33	0
Spain	3	0	10	10	33	0	0	1	18	1	0	0	32	0	0	0	10	58	0
Sweden	0	1	13	1	19	0	0	2	1	3	1	13	3	85	0	0	4	42	1
Switzerland	1	0	5	1	30	0	2	30	16	10	0	2	1	1	10	16	41	13	42
The FYR Macedonia	1	0	1	0	11	0	2	13	66	2	0	0	1	0	0	669	74	4	3
Turkey	5	0	6	2	68	0	7	27	134	9	0	0	6	1	0	22	228	22	5
Ukraine	1	1	48	7	30	0	0	1	5	1	0	4	5	2	0	0	8	636	0
United Kingdom	1	0	9	1	60	0	2	39	13	20	1	2	2	3	4	6	43	25	73
Yugoslavia	2	0	12	2	36	4	1	9	51	5	0	20	6	4	0	43	43	53	4
Europe*																			

Units: ppt per 40% emission reduction; Average over all land-areas of Europe

Table 5.3: Source-receptor relationships for AOT40c (crops) - NOx

(Emission base-year 2010, meteorology: from 5 years - 1992-1996)

emitters

Country	AL	AT	BY	BE	BA	BG	HR	CZ	DK	EE	FI	FR	DE	GR	HU	IS	IE	IT	LV
Albania	245	10	5	0	58	99	45	11	0	0	0	13	14	264	46	0.04	0	270	0
Austria	0	233	6	-6	6	1	49	35	0	1	1	122	195	1	93	0.15	0	208	2
Belarus	0	2	219	-1	0	3	2	5	7	12	12	2	12	0	11	0.06	0	6	31
Belgium	0	11	1	-236	0	0	1	4	0	0	2	463	-253	0	4	0.77	9	14	1
Bosnia and Herzegovina	5	34	5	-1	223	26	185	32	0	0	0	20	40	9	144	0.06	0	248	1
Bulgaria	5	7	14	0	7	449	10	11	1	1	2	4	13	57	52	0.03	0	27	1
Croatia	2	59	7	-1	136	14	297	35	1	0	0	44	63	6	208	0.10	0	406	1
Czech Republic	0	136	16	-19	4	0	25	139	0	1	3	85	115	1	83	0.22	0	49	4
Denmark	0	2	24	-6	0	0	0	9	23	9	14	19	103	0	3	0.75	5	3	13
Estonia	0	0	43	0	0	1	0	3	18	102	42	0	11	0	2	0.10	0	0	77
Finland	0	0	7	0	0	0	0	0	5	17	56	0	4	0	0	0.03	0	0	9
France	0	5	0	-19	0	0	2	3	0	0	0	1231	63	0	3	0.41	6	113	0
Germany	0	47	8	-50	0	0	5	48	2	1	3	205	316	0	18	0.51	3	32	3
Greece	31	3	7	0	9	204	9	6	0	0	1	10	7	569	22	0.02	0	150	1
Hungary	0	84	17	-5	24	12	103	39	1	1	2	42	55	3	499	0.10	0	123	2
Iceland	0	0	1	-10	0	0	0	1	2	0	1	58	0	0	0	0.60	89	1	0
Ireland	4	22	2	0	31	10	59	11	0	0	0	213	27	14	47	0.08	0	808	0
Italy	0	0	67	0	0	2	0	5	11	41	25	1	16	0	4	0.13	0	1	116
Latvia	0	1	84	-2	0	2	1	7	11	15	17	2	24	0	10	0.17	1	4	59
Lithuania	0	13	1	0	0	0	3	-10	0	0	1	945	243	0	3	1.02	7	32	1
Luxembourg	0	11	3	-193	0	0	0	18	-2	1	2	119	-356	0	7	1.07	9	4	2
Netherlands	0	0	3	0	0	0	0	2	6	2	3	3	15	0	0	0.02	0	0	3
Norway	0	15	60	-10	2	2	6	24	7	4	7	20	64	1	33	0.38	1	16	11
Poland	0	0	0	1	0	0	0	0	0	0	0	37	4	0	0	0.15	3	5	0
Portugal	0	7	49	0	2	37	7	10	3	4	6	7	11	3	39	0.07	0	22	6
Republic of Moldova	1	14	23	0	8	79	20	19	1	1	2	12	19	8	115	0.05	0	46	2
Romania	0	0	11	0	0	1	0	0	0	3	4	0	1	0	0	0.01	0	0	3
Russian Federation	0	84	30	-10	8	6	36	54	2	2	3	40	28	1	289	0.22	0	63	3
Slovakia	0	138	6	-1	38	5	208	22	1	0	1	76	104	4	151	0.17	0	485	1
Slovenia	0	0	0	2	0	0	0	0	0	0	0	129	10	0	0	0.14	2	30	0
Spain	0	0	10	-1	0	0	0	2	12	7	12	2	19	0	0	0.06	0	0	10
Sweden	0	22	0	-3	1	0	5	9	0	0	0	380	117	0	9	0.08	0	131	0
Switzerland	58	8	6	0	22	215	18	10	0	0	0	7	13	187	51	0.04	0	91	0
The FYR Macedonia	2	0	9	0	0	34	0	0	0	0	1	1	1	50	2	0.01	0	17	1
Turkey	0	4	65	0	1	18	4	7	3	5	8	4	10	2	23	0.09	0	12	9
Ukraine	0	1	2	-35	0	0	0	2	4	1	2	88	-28	0	1	0.96	20	2	1
United Kingdom	16	21	6	0	59	70	54	25	0	0	0	10	25	20	149	0.05	0	110	0
Yugoslavia	1	9	18	-5	4	15	8	7	2	3	5	94	22	14	20	0.12	1	50	5
Europe*																			

receivers

Country	LT	LU	NL	NO	PL	PT	MD	RO	RU	SK	SI	ES	SE	CH	MK	TR	UA	GB	YU
Albania	1	0	0	0	22	0	4	92	15	13	2	9	0	0	70	23	56	1	134
Austria	5	1	-14	2	30	0	0	23	7	45	37	10	1	48	0	1	33	-13	12
Belarus	62	0	-2	4	110	0	4	20	125	7	0	0	13	0	0	3	214	1	2
Belgium	3	26	-443	11	20	1	0	2	4	4	0	34	3	18	0	0	9	-397	0
Bosnia and Herzegovina	1	0	-1	1	41	0	1	99	7	36	11	10	0	1	5	5	41	-1	182
Bulgaria	4	0	0	0	28	0	16	367	56	18	1	1	1	0	11	64	226	-1	71
Croatia	3	0	-4	1	51	0	1	83	7	45	28	21	1	5	2	3	43	-3	97
Czech Republic	9	2	-42	4	81	0	1	20	16	81	11	5	2	11	0	0	64	-39	5
Denmark	23	1	-24	32	148	0	0	1	38	2	0	2	21	1	0	0	24	-22	0
Estonia	50	0	0	9	54	0	1	5	75	1	0	0	45	0	0	1	44	6	0
Finland	8	0	0	3	11	0	0	0	23	0	0	0	20	0	0	0	8	2	0
France	0	6	-55	4	5	4	0	1	1	1	0	177	1	41	0	0	1	-60	0
Germany	7	9	-130	9	109	0	0	6	13	14	3	13	4	31	0	0	26	-100	1
Greece	1	0	0	0	15	0	7	103	39	7	0	7	0	0	15	275	140	0	40
Hungary	5	1	-14	2	96	0	2	137	16	135	16	10	2	7	0	2	118	-9	65
Iceland	0	0	-16	6	10	0	0	0	3	0	0	5	1	2	0	0	3	-66	0
Ireland	1	0	0	0	15	2	0	22	2	13	11	51	0	12	2	4	12	1	30
Italy	110	0	-1	8	77	0	1	9	74	2	0	0	41	0	0	1	59	0	1
Latvia	201	0	-6	9	157	0	1	12	84	8	0	0	33	0	0	1	84	-12	1
Lithuania	2	112	-147	6	11	1	0	2	2	3	0	48	2	32	0	0	4	-105	0
Luxembourg	4	2	-644	19	43	1	0	3	7	6	0	22	6	6	0	0	13	-393	0
Netherlands	4	0	0	19	13	0	0	0	5	0	0	0	14	0	0	0	1	-9	0
Norway	32	0	-22	8	483	0	2	22	55	33	1	3	10	2	0	0	148	-16	6
Poland	0	0	1	0	0	503	0	0	0	0	0	573	0	0	0	0	0	5	0
Portugal	11	0	-1	2	75	0	89	303	93	19	1	1	4	1	0	28	528	0	13
Republic of Moldova	5	0	-2	1	62	0	18	593	43	48	3	3	1	1	2	15	249	-2	55
Romania	4	0	0	0	6	0	0	3	199	0	0	0	2	0	0	8	69	0	0
Russian Federation	8	1	-25	3	134	0	2	65	24	244	8	5	3	5	0	1	180	-24	21
Slovakia	5	1	-6	1	27	0	0	50	7	41	97	23	1	15	0	2	38	-3	37
Slovenia	0	0	1	0	0	134	0	0	0	0	0	1398	0	2	0	0	0	4	0
Spain	15	0	-2	8	33	0	0	0	19	0	0	0	51	0	0	0	9	-1	0
Sweden	0	1	-11	0	6	0	0	3	0	4	3	20	0	171	0	0	2	-3	2
Switzerland	1	0	0	0	20	0	5	143	16	15	1	3	0	0	112	16	67	0	146
The FYR Macedonia	2	0	0	0	8	0	5	32	107	0	0	1	1	0	1	1101	211	0	2
Turkey	17	0	0	2	79	0	19	81	249	15	0	1	6	0	0	25	807	0	6
Ukraine	2	0	-64	14	19	0	0	0	5	1	0	8	4	2	0	0	5	-773	0
United Kingdom	2	0	-1	0	40	0	3	188	12	35	3	4	0	1	33	4	64	-1	342
Yugoslavia	8	0	-13	3	37	12	3	35	113	9	1	91	5	5	1	69	113	-26	11
Europe*																			

Units: ppb.h per 40% emission reduction; Average over all land-areas of Europe

Table 5.4: Source-receptor relationships for AOT40c (crops) - VOC

(Emission base-year 2010, meteorology: from 5 years - 1992-1996)

emitters

Country	AL	AT	BY	BE	BA	BG	HR	CZ	DK	EE	FI	FR	DE	GR	HU	IS	IE	IT	LV
Albania	58	7	4	2	13	14	16	12	1	0	0	12	30	41	11	0.	0	229	1
Austria	0	73	5	20	1	1	12	54	6	0	0	81	170	1	16	0.01	1	264	4
Belarus	0	2	25	4	0	1	0	7	7	2	2	8	26	0	2	0.	1	7	16
Belgium	0	16	3	318	0	0	1	45	9	0	0	342	570	0	3	0.04	15	18	4
Bosnia and Herzegovina	1	15	4	6	20	4	29	25	2	0	0	22	66	2	22	0.	0	234	2
Bulgaria	3	6	9	2	2	57	4	13	2	0	0	8	32	27	14	0.	0	25	3
Croatia	0	31	6	10	14	5	44	34	4	0	0	47	101	3	34	0.01	1	361	3
Czech Republic	0	56	13	38	1	1	9	193	11	1	1	76	289	1	19	0.01	2	73	7
Denmark	0	4	13	22	0	0	0	27	98	3	2	36	146	0	1	0.02	7	4	15
Estonia	0	1	7	1	0	0	0	5	9	8	6	2	14	0	1	0.	1	0	21
Finland	0	0	2	1	0	0	0	1	2	2	4	1	6	0	0	0.	0	0	4
France	0	8	0	63	0	0	2	16	4	0	0	355	194	0	2	0.02	6	87	1
Germany	0	30	7	86	0	0	2	71	12	0	0	150	495	0	6	0.02	7	49	5
Greece	25	4	7	1	3	29	5	7	1	0	0	8	17	221	8	0.	0	106	2
Hungary	1	38	11	16	5	5	21	56	7	0	0	43	129	2	69	0.01	1	139	5
Iceland	0	1	1	19	0	0	0	3	2	0	0	62	54	0	0	0.01	37	2	1
Ireland	2	27	3	7	7	4	24	24	2	0	0	116	64	4	16	0.01	1	1036	2
Italy	0	1	11	4	0	1	0	6	15	5	4	5	25	0	1	0.01	1	1	39
Latvia	0	2	16	7	0	0	0	8	20	3	3	11	39	0	3	0.01	2	4	34
Lithuania	0	17	2	187	0	0	2	40	6	0	0	312	544	0	3	0.09	6	35	3
Luxembourg	0	14	5	217	0	0	0	46	17	0	0	209	633	0	4	0.04	16	7	3
Netherlands	0	0	2	2	0	0	0	2	6	0	0	6	19	0	0	0.	0	0	2
Norway	0	14	18	20	0	2	3	57	20	2	2	34	141	1	10	0.01	3	22	12
Poland	0	0	0	3	0	0	0	0	0	0	0	33	10	0	0	0.01	2	4	0
Portugal	0	6	15	4	1	8	3	12	5	0	1	12	29	6	8	0.01	0	22	5
Republic of Moldova	1	9	10	4	2	16	6	20	3	0	0	15	47	7	22	0.	1	40	4
Romania	0	0	2	0	0	0	0	0	0	0	0	0	2	0	0	0.	0	0	2
Russian Federation	0	37	14	22	2	4	11	94	10	1	1	43	140	1	47	0.01	2	75	7
Slovakia	0	63	6	12	5	3	35	40	5	0	0	72	125	2	26	0.01	1	489	4
Slovenia	0	0	0	6	0	0	0	1	0	0	0	63	17	0	0	0.01	1	17	0
Spain	0	0	3	4	0	0	0	3	15	1	1	6	26	0	0	0.	1	0	6
Sweden	0	25	1	13	0	0	5	17	1	0	0	172	137	0	5	0.	1	408	1
Switzerland	15	6	4	2	4	23	5	10	1	0	0	8	24	40	10	0.	0	55	1
The FYR Macedonia	1	0	7	0	0	6	0	1	1	0	0	1	5	31	1	0.	0	9	3
Turkey	0	4	17	3	0	3	1	9	5	1	1	9	23	2	5	0.01	0	14	7
Ukraine	0	2	2	54	0	0	0	11	8	0	0	124	134	0	1	0.03	15	4	2
United Kingdom	4	12	5	5	9	11	13	24	2	0	0	14	55	5	24	0.	0	91	2
Yugoslavia	0	5	5	12	0	2	2	11	4	0	0	41	54	6	4	0.01	1	56	3
Europe*																			

receivers

Country	LT	LU	NL	NO	PL	PT	MD	RO	RU	SK	SI	ES	SE	CH	MK	TR	UA	GB	YU
Albania	0	0	4	0	26	0	2	22	12	8	1	2	0	0	6	17	34	12	34
Austria	2	1	18	1	86	0	0	9	7	22	7	3	2	31	0	2	26	57	6
Belarus	7	0	7	1	48	0	1	6	44	3	0	0	6	0	0	2	49	19	1
Belgium	1	8	203	5	58	0	0	4	6	6	0	10	6	14	0	0	12	507	1
Bosnia and Herzegovina	1	0	8	1	53	0	1	28	7	16	2	2	1	1	0	5	31	23	31
Bulgaria	1	0	4	0	33	0	6	65	34	10	0	0	2	0	2	34	82	13	24
Croatia	1	0	13	1	80	0	1	26	8	25	6	5	2	6	0	3	37	37	23
Czech Republic	4	1	42	3	214	0	1	9	16	36	3	3	5	12	0	1	46	100	5
Denmark	6	0	42	9	129	0	0	1	28	3	0	2	37	2	0	0	17	207	0
Estonia	4	0	5	2	30	0	0	2	23	1	0	0	13	0	0	1	15	31	0
Finland	1	0	2	0	6	0	0	0	8	0	0	0	5	0	0	0	3	9	0
France	0	3	50	2	19	2	0	1	1	2	0	63	2	23	0	0	3	187	0
Germany	2	4	97	4	115	0	0	4	12	12	1	6	6	22	0	0	24	227	1
Greece	1	0	2	0	20	0	4	32	31	6	0	1	1	0	2	161	67	7	17
Hungary	2	0	23	2	139	0	2	35	13	55	4	3	4	5	0	2	71	52	22
Iceland	0	0	26	2	11	0	0	0	4	0	0	2	1	1	0	0	6	251	0
Ireland	1	0	8	1	42	1	0	13	4	14	6	11	1	21	0	3	16	21	13
Italy	9	0	7	2	35	0	1	3	25	2	0	0	15	0	0	1	19	47	1
Latvia	19	0	14	3	65	0	0	5	29	4	0	0	16	0	0	1	24	63	1
Lithuania	1	20	93	3	41	0	0	2	3	4	0	14	3	24	0	0	5	215	0
Luxembourg	1	4	304	6	83	0	0	4	8	8	0	6	8	8	0	0	19	540	0
Netherlands	0	0	4	2	13	0	0	0	3	0	0	0	4	0	0	0	1	25	0
Norway	7	0	32	4	292	0	1	7	31	20	1	1	10	3	0	1	56	82	3
Poland	0	0	3	0	1	247	0	0	0	0	0	221	0	0	0	0	0	26	0
Portugal	2	0	6	1	53	0	15	49	44	8	0	0	3	1	0	20	122	18	5
Republic of Moldova	2	0	7	1	60	0	5	82	24	17	1	1	2	1	0	12	89	20	19
Romania	0	0	0	0	4	0	0	1	48	0	0	0	1	0	0	3	20	2	0
Russian Federation	4	0	31	2	229	0	2	19	19	67	2	2	5	5	0	1	79	74	11
Slovakia	2	0	14	1	91	0	1	16	8	24	19	6	2	21	0	3	35	33	11
Slovenia	0	0	5	0	1	56	0	0	0	0	0	302	0	1	0	0	0	29	0
Spain	2	0	7	1	19	0	0	0	9	0	0	0	15	0	0	0	4	33	0
Sweden	0	1	14	0	19	0	0	3	1	4	2	7	0	131	0	0	5	36	2
Switzerland	0	0	4	0	23	0	2	23	11	7	0	0	1	0	8	7	33	10	28
The FYR Macedonia	1	0	0	0	10	0	2	11	55	1	0	0	1	0	0	790	70	3	2
Turkey	3	0	4	1	50	0	5	18	96	6	0	0	4	1	0	17	185	17	3
Ukraine	0	1	58	5	25	0	0	1	7	1	0	3	4	3	0	0	9	630	0
United Kingdom	1	0	7	0	50	0	2	38	9	18	1	1	1	2	2	3	38	21	65
Yugoslavia	1	0	13	1	32	5	1	8	34	4	0	22	3	4	0	48	34	49	3
Europe*																			

Units: ppb.h per 40% emission reduction;
Average over all land-areas of Europe

Table 5.5: Source-receptor relationships for AOT60 - NOx

(Emission base-year 2010, meteorology: from 5 years - 1992-1996)

emitters

Country	AL	AT	BY	BE	BA	BG	HR	CZ	DK	EE	FI	FR	DE	GR	HU	IS	IE	IT	LV
Albania	7	0	0	0	3	2	3	0	0	0	0	0	1	8	4	0.	0	0	0
Austria	0	39	0	0	1	0	9	18	0	0	0	15	35	0	20	0.	0	52	0
Belarus	0	0	5	0	0	0	0	2	0	0	0	0	0	0	4	0.	0	1	0
Belgium	0	8	1	81	0	0	0	14	0	0	0	284	144	0	3	0.02	0	7	0
Bosnia and Herzegovina	0	4	0	0	7	0	12	6	0	0	0	2	4	0	28	0.	0	25	0
Bulgaria	0	1	0	0	0	38	2	1	0	0	0	0	2	1	8	0.	0	2	0
Croatia	0	14	0	0	10	1	43	11	0	0	0	13	17	0	57	0.	0	108	0
Czech Republic	0	42	2	0	1	0	8	144	0	0	0	26	99	0	32	0.	0	23	0
Denmark	0	2	2	1	0	0	1	17	22	0	0	16	70	0	3	0.	0	4	0
Estonia	0	0	0	0	0	0	0	1	4	2	0	2	8	0	0	0.	0	0	1
Finland	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0.	0	0	0
France	0	2	0	13	0	0	1	4	0	0	0	263	58	0	1	0.	0	34	0
Germany	0	17	1	5	0	0	2	46	0	0	0	93	264	0	8	0.01	0	18	0
Greece	1	0	0	0	0	21	0	0	0	0	0	0	1	55	3	0.	0	15	0
Hungary	0	36	3	0	4	3	28	29	0	0	0	19	31	0	216	0.	0	49	0
Iceland	0	0	0	0	0	0	0	1	0	0	0	18	4	0	0	0.01	6	1	0
Ireland	0	6	0	0	5	1	16	4	0	0	0	25	7	1	16	0.	0	277	0
Italy	0	1	0	0	0	0	0	2	1	0	0	1	7	0	2	0.	0	1	2
Latvia	0	1	1	0	0	0	1	4	1	0	0	2	8	0	5	0.	0	2	1
Lithuania	0	6	1	79	0	0	0	10	0	0	0	363	410	0	1	0.01	0	20	0
Luxembourg	0	7	1	10	0	0	0	23	0	0	0	124	61	0	8	0.05	0	3	0
Netherlands	0	0	0	0	0	0	0	0	1	0	0	2	3	0	0	0.	0	0	0
Norway	0	11	5	0	0	0	2	35	1	0	0	10	40	0	19	0.	0	7	0
Poland	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0.	0	0	0
Portugal	0	2	1	0	0	8	2	4	0	0	0	3	4	0	14	0.	0	5	0
Republic of Moldova	0	3	0	0	0	9	3	4	0	0	0	3	5	0	27	0.	0	7	0
Romania	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	-0.00	0	0	0
Russian Federation	0	37	2	0	1	0	9	53	0	0	0	20	28	0	132	0.	0	22	0
Slovakia	0	24	0	0	4	0	29	11	0	0	0	14	21	1	30	0.	0	183	0
Slovenia	0	0	0	0	0	0	0	0	0	0	0	8	0	0	0	0.	0	1	0
Spain	0	0	0	0	0	0	0	1	4	0	0	1	10	0	0	0.	0	0	0
Sweden	0	3	0	0	0	0	1	0	0	0	0	30	37	0	2	-0.00	0	74	0
Switzerland	0	0	0	0	0	5	0	0	0	0	0	0	0	0	1	0.	0	0	0
The FYR Macedonia	0	0	0	0	0	7	0	0	0	0	0	0	0	13	0	0.	0	4	0
Turkey	0	1	1	0	0	2	1	2	0	0	0	1	2	0	7	0.	0	3	0
Ukraine	0	1	0	-3	0	0	0	4	0	0	0	50	17	0	1	0.01	0	1	0
United Kingdom	0	4	0	0	2	7	8	7	0	0	0	2	6	0	43	0.	0	13	0
Yugoslavia	0	2	0	1	0	1	1	5	0	0	0	22	17	1	6	0.	0	14	0
Europe*																			

receivers

Country	LT	LU	NL	NO	PL	PT	MD	RO	RU	SK	SI	ES	SE	CH	MK	TR	UA	GB	YU
Albania	0	0	0	0	3	0	0	2	0	0	0	0	0	0	0	3	0	0	5
Austria	0	0	-1	0	14	0	0	4	0	13	6	0	0	6	0	0	5	-1	2
Belarus	1	0	0	0	15	0	0	2	4	3	0	0	0	0	0	0	8	0	0
Belgium	0	22	-54	0	12	0	0	4	1	3	0	13	0	18	0	0	5	-45	0
Bosnia and Herzegovina	0	0	0	0	5	0	0	7	0	9	2	0	0	0	0	0	3	0	9
Bulgaria	0	0	0	0	2	0	0	39	1	3	0	0	0	0	0	9	9	0	7
Croatia	0	0	-1	0	14	0	0	17	0	16	7	1	0	3	0	0	7	0	13
Czech Republic	0	1	-1	0	57	0	0	6	1	33	3	0	0	5	0	0	12	-1	3
Denmark	0	0	6	0	35	0	0	1	3	2	0	0	1	2	0	0	5	-12	0
Estonia	1	0	0	0	6	0	0	0	0	0	0	0	3	0	0	0	0	0	0
Finland	0	0	0	0	1	0	0	0	0	0	0	0	1	0	0	0	0	0	0
France	0	6	-5	0	3	0	0	0	0	0	0	13	0	15	0	0	0	-8	0
Germany	0	7	-7	0	30	0	0	2	0	7	1	2	0	18	0	0	6	-8	0
Greece	0	0	0	0	2	0	0	11	4	1	0	0	0	0	0	78	13	0	3
Hungary	0	0	-2	0	46	0	0	44	1	63	7	1	0	4	0	0	21	0	23
Iceland	0	0	0	0	2	0	0	0	0	0	0	0	0	1	0	0	1	7	0
Ireland	0	0	0	0	5	0	0	5	0	5	4	2	0	2	0	0	2	0	5
Italy	4	0	0	0	16	0	0	0	1	1	0	0	1	0	0	0	0	0	0
Latvia	9	0	0	0	39	0	0	1	2	2	0	0	1	0	0	0	1	-11	1
Lithuania	0	87	-15	0	8	0	0	0	0	1	0	5	0	31	0	0	1	0	0
Luxembourg	0	4	-45	0	21	0	0	5	1	7	0	11	0	6	0	0	9	-27	0
Netherlands	0	0	1	0	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Norway	2	0	0	0	145	0	0	7	3	19	1	0	0	2	0	0	18	-1	2
Poland	0	0	0	0	0	92	0	0	0	0	0	42	0	0	0	0	0	0	0
Portugal	0	0	0	0	9	0	6	37	1	7	0	0	0	0	0	7	23	0	3
Republic of Moldova	0	0	0	0	8	0	0	65	0	11	0	0	0	0	0	2	11	0	9
Romania	0	0	0	0	0	0	0	0	6	0	0	0	0	0	0	0	2	0	0
Russian Federation	0	0	-3	0	81	0	0	17	1	99	3	1	0	3	0	0	26	-1	6
Slovakia	0	0	0	0	14	0	0	10	0	11	18	1	0	3	0	1	5	0	4
Slovenia	0	0	0	0	0	10	0	0	0	0	0	75	0	0	0	0	0	0	0
Spain	0	0	0	0	8	0	0	0	0	0	0	0	3	0	0	0	1	-1	0
Sweden	0	0	0	0	0	0	0	0	0	0	0	0	0	30	0	0	0	0	0
Switzerland	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	0	1	0	0
The FYR Macedonia	0	0	0	0	0	0	0	4	7	0	0	0	0	0	0	406	14	0	0
Turkey	0	0	0	0	10	0	1	11	7	4	0	0	0	0	0	4	57	0	1
Ukraine	0	1	-9	0	8	0	0	1	0	1	0	1	0	1	0	0	3	-79	0
United Kingdom	0	0	0	0	10	0	0	20	0	11	1	0	0	0	0	0	4	0	39
Yugoslavia	0	0	-1	0	9	1	0	4	4	3	0	5	0	2	0	23	7	-2	1
Europe*																			

Units: ppb.h per 40% emission reduction; Average over all land-areas of Europe

Table 5.6: Source-receptor relationships for AOT60 - VOC

(Emission base-year 2010, meteorology: from 5 years - 1992-1996)

emitters

Country	AL	AT	BY	BE	BA	BG	HR	CZ	DK	EE	FI	FR	DE	GR	HU	IS	IE	IT	LV
Albania	2	0	0	0	0	0	1	1	0	0	0	1	3	1	1	0.	0	49	0
Austria	0	7	0	3	0	0	2	6	0	0	0	10	22	0	3	0.	0	46	0
Belarus	0	1	0	0	0	0	0	2	0	0	0	0	2	0	1	0.	0	3	0
Belgium	0	10	1	107	0	0	1	23	1	0	0	113	260	0	2	0.	1	14	1
Bosnia and Herzegovina	0	2	0	1	0	0	1	3	0	0	0	3	7	0	3	0.	0	35	0
Bulgaria	0	1	0	0	0	4	0	2	0	0	0	1	7	3	2	0.	0	3	0
Croatia	0	7	1	2	0	0	5	6	0	0	0	16	23	0	7	0.	0	123	0
Czech Republic	0	16	2	4	0	0	3	40	0	0	0	17	47	0	7	0.	0	32	0
Denmark	0	3	1	10	0	0	0	15	6	0	0	20	56	0	1	0.	0	8	0
Estonia	0	0	0	0	0	0	0	2	1	0	0	5	8	0	0	0.	0	1	0
Finland	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0.	0	0	0
France	0	3	0	17	0	0	1	5	0	0	0	63	60	0	1	0.	0	28	0
Germany	0	11	2	22	0	0	1	23	0	0	0	46	126	0	3	0.	0	27	0
Greece	1	0	0	0	0	4	0	1	0	0	0	1	2	59	1	0.	0	22	0
Hungary	0	12	3	3	1	1	5	16	0	0	0	18	37	0	23	0.	0	64	0
Iceland	0	0	0	6	0	0	0	2	0	0	0	16	20	0	0	0.	1	2	0
Ireland	0	7	0	1	1	1	7	6	0	0	0	25	13	1	5	0.	0	320	0
Italy	0	1	0	0	0	0	0	4	0	0	0	3	6	0	0	0.	0	4	0
Latvia	0	2	0	0	0	0	0	4	0	0	0	4	7	0	1	0.	0	4	1
Lithuania	0	8	1	60	0	0	0	12	2	0	0	90	227	0	1	0.	0	25	0
Luxembourg	0	9	2	80	0	0	0	29	1	0	0	92	273	0	3	0.01	1	17	1
Netherlands	0	0	0	1	0	0	0	0	0	0	0	2	4	0	0	0.	0	1	0
Norway	0	9	3	2	0	0	1	21	1	0	0	11	24	0	6	0.	0	14	1
Poland	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0.	0	0	0
Portugal	0	2	0	1	0	1	1	3	0	0	0	5	7	1	2	0.	0	6	0
Republic of Moldova	0	2	0	0	0	1	1	3	0	0	0	4	9	0	4	0.	0	11	0
Romania	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0.	0	0	0
Russian Federation	0	11	2	4	0	0	3	24	0	0	0	16	30	0	17	0.	0	31	0
Slovakia	0	9	0	1	0	0	4	6	0	0	0	16	19	1	4	0.	0	201	0
Slovenia	0	0	0	0	0	0	0	0	0	0	0	4	2	0	0	0.	0	1	0
Spain	0	0	0	1	0	0	0	1	1	0	0	3	10	0	0	0.	0	1	0
Sweden	0	2	0	0	0	0	1	1	0	0	0	10	12	0	1	0.	0	67	0
Switzerland	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0.	0	0	0
The FYR Macedonia	0	0	0	0	0	1	0	0	0	0	0	0	1	12	0	0.	0	3	0
Turkey	0	1	1	0	0	0	0	2	0	0	0	2	4	0	1	0.	0	4	0
Ukraine	0	2	1	23	0	0	0	7	1	0	0	48	66	0	1	0.	0	3	0
United Kingdom	0	2	0	0	0	1	1	5	0	0	0	3	11	0	5	0.	0	14	0
Yugoslavia	0	1	0	3	0	0	0	3	0	0	0	9	14	1	1	0.	0	17	0
Europe*																			

receivers

Country	LT	LU	NL	NO	PL	PT	MD	RO	RU	SK	SI	ES	SE	CH	MK	TR	UA	GB	YU
Albania	0	0	0	0	2	0	0	1	0	0	0	0	0	0	0	3	1	0	1
Austria	0	0	1	0	10	0	0	1	0	4	0	0	0	3	0	0	4	5	1
Belarus	0	0	0	0	6	0	0	0	2	1	0	0	0	0	0	0	3	0	0
Belgium	0	4	56	1	27	0	0	2	2	4	0	3	1	10	0	0	7	69	0
Bosnia and Herzegovina	0	0	0	0	3	0	0	2	0	2	0	0	0	0	0	0	2	0	1
Bulgaria	0	0	0	0	3	0	0	8	2	2	0	0	0	0	0	10	5	0	2
Croatia	0	0	3	0	12	0	0	6	1	6	1	0	0	3	0	0	6	5	3
Czech Republic	0	0	6	0	35	0	0	3	2	12	1	0	0	4	0	0	11	9	2
Denmark	0	0	12	0	25	0	0	1	3	2	0	0	1	3	0	0	5	18	0
Estonia	0	0	0	0	4	0	0	0	0	0	0	0	0	0	0	0	0	1	0
Finland	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
France	0	1	10	0	7	0	0	0	0	1	0	5	0	5	0	0	1	15	0
Germany	0	1	18	0	27	0	0	1	1	6	0	1	0	9	0	0	7	25	0
Greece	0	0	0	0	2	0	0	4	5	0	0	0	0	0	0	65	10	1	2
Hungary	0	0	3	0	31	0	0	12	3	17	1	1	0	5	0	0	17	7	6
Iceland	0	0	7	0	4	0	0	0	0	0	0	0	0	1	0	0	2	38	0
Ireland	0	0	1	0	9	0	0	4	0	4	2	1	0	4	0	0	4	3	3
Italy	0	0	0	0	9	0	0	0	0	1	0	0	0	0	0	0	0	1	0
Latvia	0	0	0	0	18	0	0	0	0	2	0	0	0	0	0	0	1	0	0
Lithuania	0	6	23	0	17	0	0	0	0	2	0	2	1	14	0	0	2	15	0
Luxembourg	0	3	75	0	36	0	0	3	3	6	0	2	0	6	0	0	12	70	0
Netherlands	0	0	1	0	2	0	0	0	0	0	0	0	0	0	0	0	0	3	0
Norway	0	0	2	0	58	0	0	3	3	11	0	0	0	1	0	0	11	4	1
Poland	0	0	0	0	0	56	0	0	0	0	0	31	0	0	0	0	0	2	0
Portugal	0	0	1	0	5	0	0	6	1	2	0	0	0	0	0	13	6	3	1
Republic of Moldova	0	0	1	0	7	0	0	10	1	3	0	0	0	1	0	4	6	1	2
Romania	0	0	0	0	0	0	0	0	3	0	0	0	0	0	0	0	1	0	0
Russian Federation	0	0	4	0	42	0	0	5	3	20	1	1	0	3	0	0	18	9	3
Slovakia	0	0	1	0	13	0	0	4	1	5	2	1	0	5	0	1	6	3	1
Slovenia	0	0	0	0	0	6	0	0	0	0	0	26	0	0	0	0	0	1	0
Spain	0	0	1	0	4	0	0	0	0	0	0	0	0	0	0	0	0	2	0
Sweden	0	0	0	0	1	0	0	0	0	0	0	0	0	6	0	0	1	0	0
Switzerland	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0
The FYR Macedonia	0	0	0	0	1	0	0	2	8	0	0	0	0	0	0	371	10	0	0
Turkey	0	0	0	0	5	0	0	2	5	1	0	0	0	0	0	8	17	1	0
Ukraine	0	1	22	0	12	0	0	1	1	1	0	0	0	2	0	0	5	95	0
United Kingdom	0	0	1	0	8	0	0	5	0	4	0	0	0	0	0	0	4	1	5
Yugoslavia	0	0	2	0	6	0	0	1	2	1	0	2	0	1	0	21	3	5	0
Europe*																			

Units: ppb.h per 40% emission reduction; Average over all land-areas of Europe

5.3 Comparison of Eulerian versus Lagrangian S-R calculations

For 6 countries, both the Eulerian and Lagrangian model have been used to calculate country-to-grid changes in $AOT40_f$ caused by 40% reductions in NO_x and VOC emissions. The 6 countries were chosen to give a mix of high and low emitters, in different parts of Europe. The aim is to obtain a first estimate of the likely effect of using the 3-D modelling system in future S-R studies, and to quantify the uncertainties in such calculations to some extent by running two models of very different physical character. As for the Lagrangian model calculations presented above, the base-emissions used were from the 2010 EMEP inventory described in chapter 2. Meteorology was from April-September 1996.

Figures 5.1-5.3 show the calculated $AOT40_f$ changes obtained from these calculations. Note that the scale used for plotting is identical for all four plots from a particular emitter country, but varies greatly from country to country. These plots share many common features so will be discussed together.

In general, both models show rather similar behaviour when comparing the effects of NO_x control against that of VOC control. Thus, for high NO_x countries such as Germany and the United Kingdom, the negative effects of NO_x control is seen in both models, and in approximately the same areas. The negative effects are generally more spatially limited in the 3-D model though, with the exception of Italy where the 3-D model predicts increased AOT40 values even in southern Italy, whereas the Lagrangian model only shows such effects in northern Italy. However, both models predict similar behaviour for the effect of Italian NO_x emissions on other countries, with reduced ozone over the Balkans, southern France, etc. The biggest discrepancy occurs for Norwegian emissions, where the 3-D model predicts substantially greater effects than the Lagrangian model. This may well be due to the fact that AOT40 values are lowest in this part of Europe, and hence very uncertain and very sensitive to systematic differences.

A general pattern is that the 3-D model predicts stronger changes than the Lagrangian model. Thus, both the negative and positive NO_x effects are stronger in the 3-D model, as are the always-positive VOC effects.

This situation is very consistent with what should be expected from the different resolutions of the two models, as has been partly discussed in chapter 4 and will be discussed further in chapter 8. Indeed, such resolution effects were also apparent in the 4-model study of Hass et al. (1997), but in that study so many other factors (including basic emission input) were different between the models that a good explanation of different control strategy responses was impossible to make. Indeed, even with the two EMEP models currently used, a number of factors other than resolution may influence the results shown, but especially the vertical resolution does seem the most likely reason for the 3-D model's greater sensitivity to emissions.

For a number of reasons (see chapter 8), it is difficult to say which set of calculations is 'better' at this stage, but rather these calculations offer two alternative views. Given the known uncertainties of modelling in general and AOT predictions in particular, the large similarity between the results of the two models is encouraging and gives continues confidence in the usage of source-receptor matrices in emission control analysis.

5.4 References

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Schöpp, W., Amann, M., Cofala, J., Heyes, C., , and Klimont, Z , 1999, Integrated assessment of european air pollution emission control strategies, *Environ. Mod. & Software*, 14, 1–9.

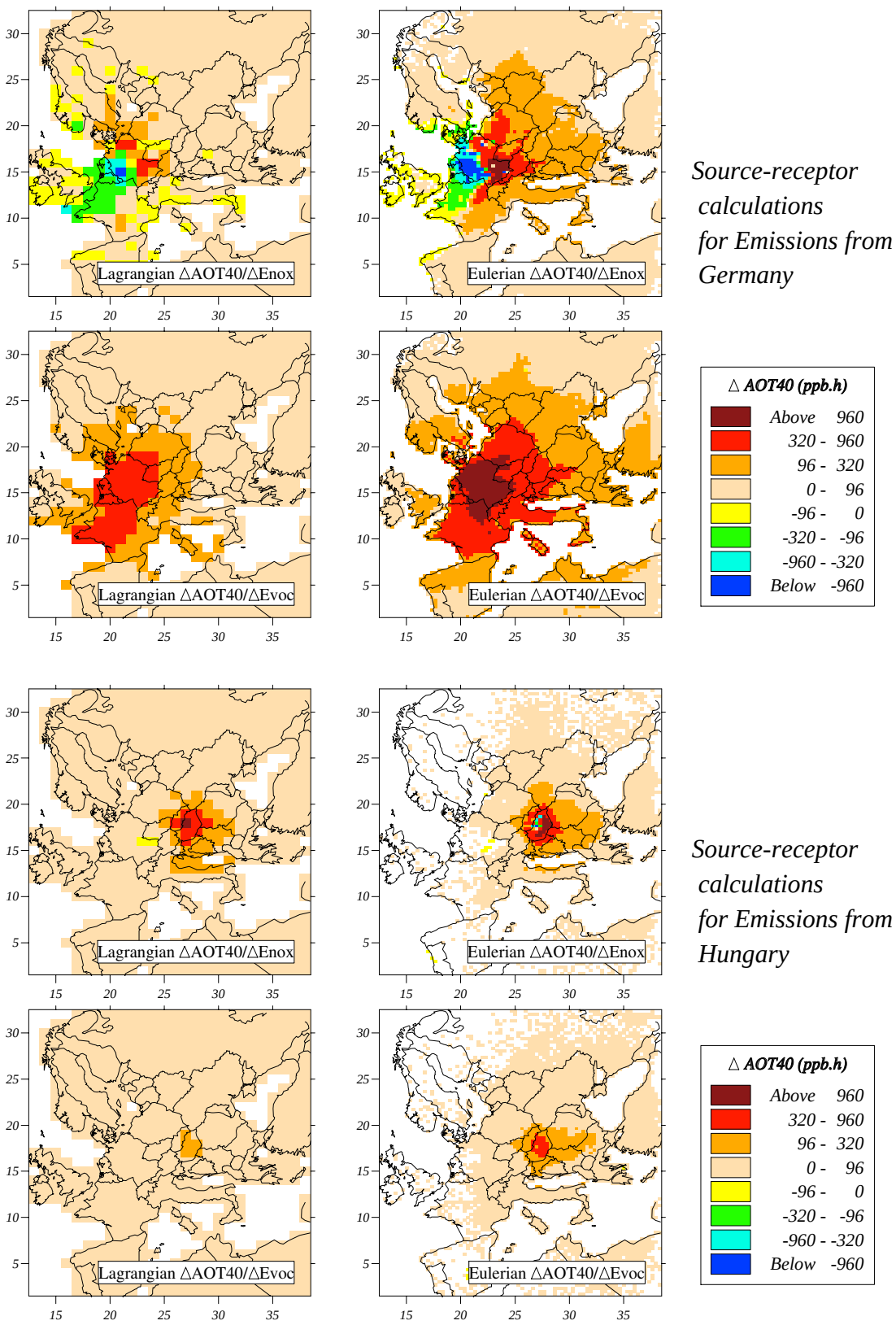
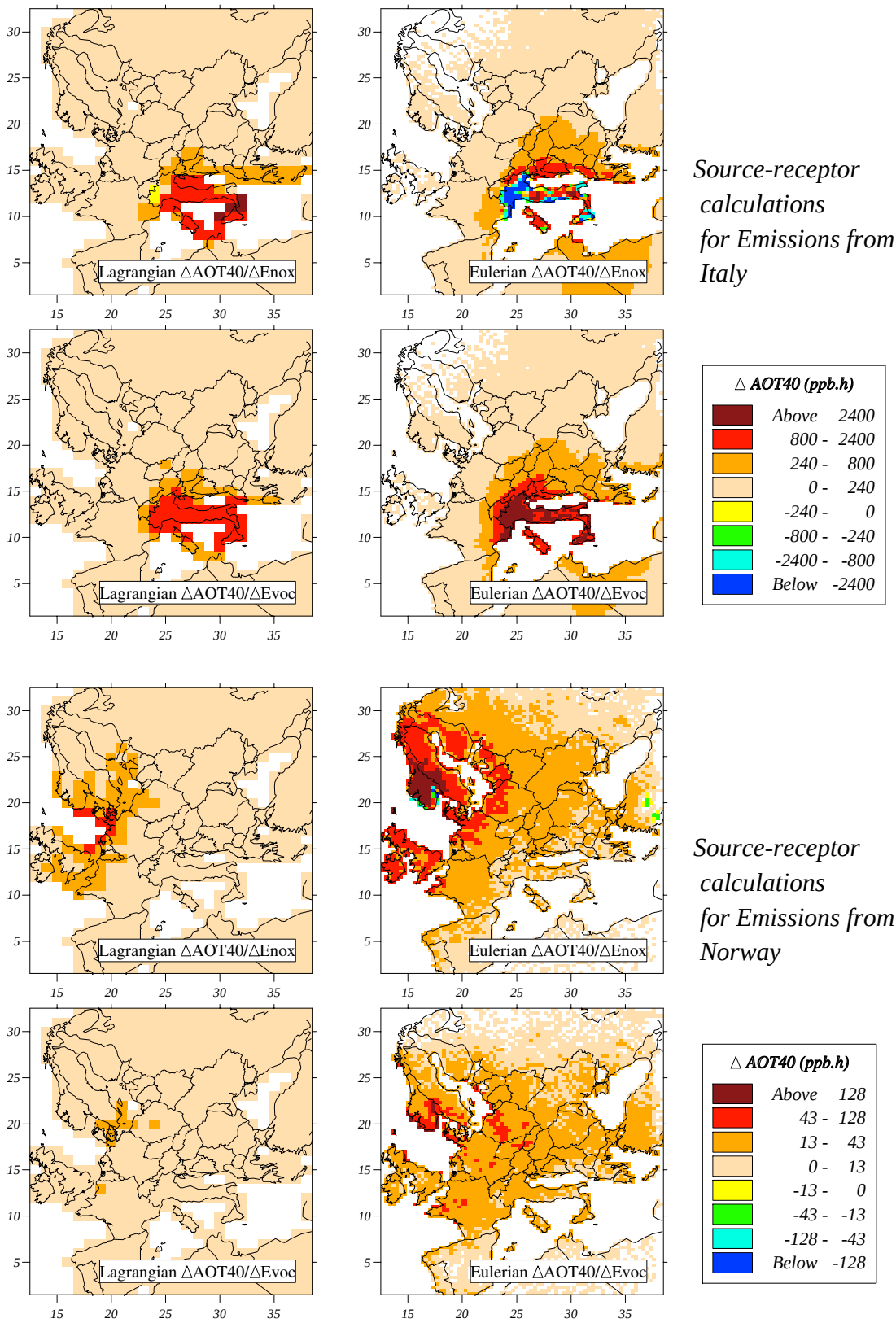


Figure 5.1: Comparison of source-receptor calculations from Eulerian and Lagrangian models. Plots show reductions in $AOT40_f$ levels caused by 40% reductions of NO_x or VOC emissions from the given emitter country. Plots are arranged in groups of 4 per country, here Germany (top) and Hungary (bottom).



Source-receptor calculations for Emissions from Italy

Source-receptor calculations for Emissions from Norway

Figure 5.2: *Cont. from Fig. 5.1*

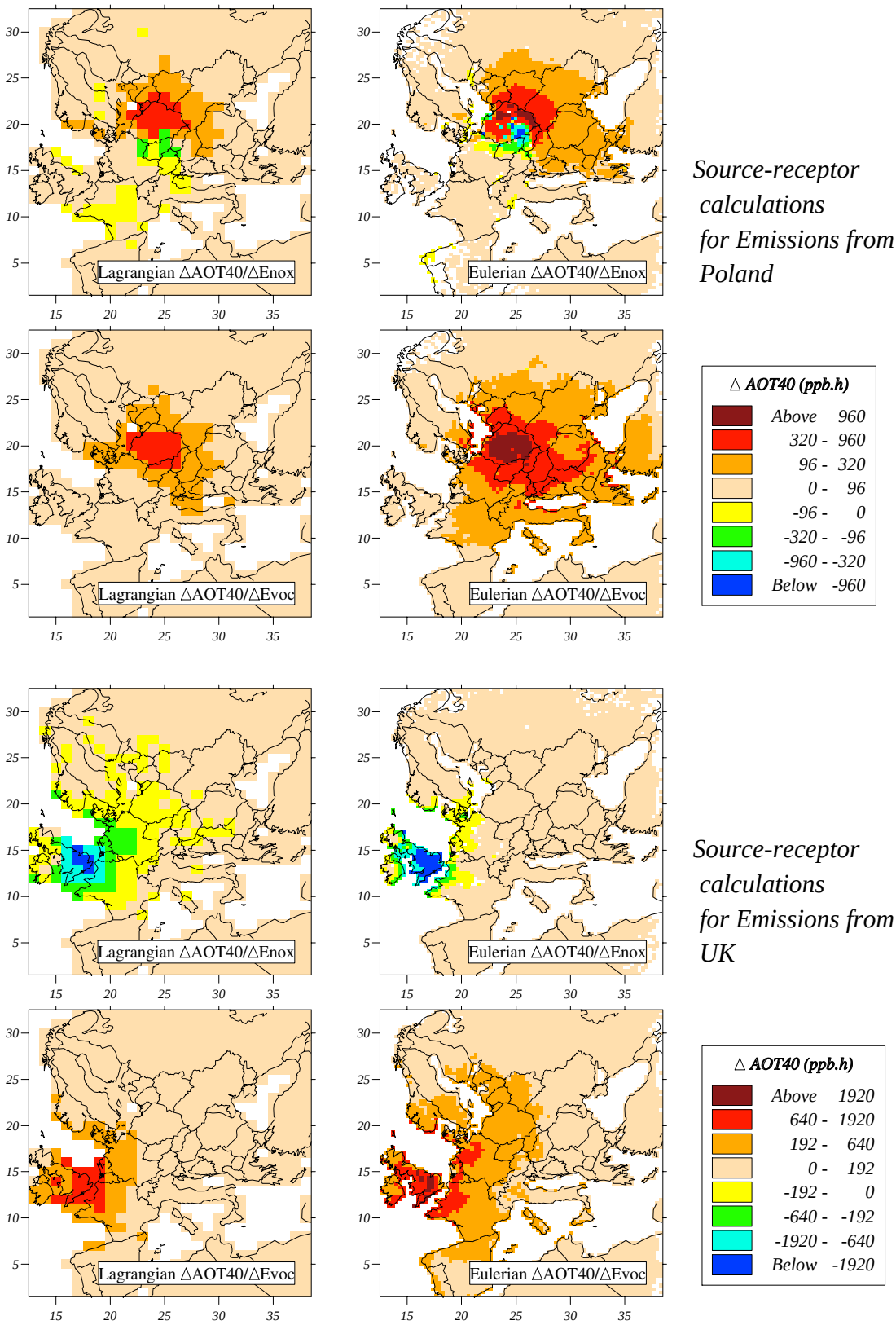


Figure 5.3: *Cont. from Fig. 5.1*

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Chapter 6

Comparison of Eulerian and Lagrangian model calculations for carbonyls: preliminary results

S. Solberg, D. Simpson and J.E. Jonson

6.1 Introduction:

In Simpson and Jonson (1998), results from the EMEP Lagrangian and Eulerian photochemistry models were compared. The comparison was carried out mainly for ozone and ozone related parameters as AOT40 and AOT60. Comparisons was also made for NO_2, total nitrate, SO_2 and sulphate. For ozone the results from the two models were comparable, but with a tendency for the Eulerian model to give more ozone in the Mediterranean (there are virtually no measurements in the area to validate this difference). Compared to measurements of daily maximum ozone there was a tendency that the Eulerian model performed better relative to the measurements than the Lagrangian model. The same was also the case for most other components compared. For NO_2 results from the Lagrangian model were about a factor of two lower than measured, averaged over all sites compared, for the period considered (April - September). For the same period results from the Eulerian model was much closer to the measurements.

Here we report first results of an evaluation of the model performance based on comparison with the EMEP VOC measurements. The results are not very encouraging at this stage, but unfortunately time has not yet allowed a more detailed investigation of the causes of this, and the presented results and conclusions should therefore be regarded as very preliminary. A more thorough follow-up study is foreseen in the coming year.

VOC measurements are particularly useful for oxidant model evaluations for several reasons. Firstly, a large number of VOC are measured, giving a good basis for evaluating the model performance for VOC in general and for the VOC speciation in particular. Secondly, carbonyls are to a large extent formed by the atmospheric oxidation of other VOC, thus directly linked to the atmospheric chemical condition (OH-concentration, dissociation etc), and thereby useful for evaluating the model simulation of atmospheric oxidation processes. Previous studies (Solberg et al., 1995) have shown a very good agreement between some of the modelled and measured VOC (particularly formaldehyde).

6.2 Model comparison

The EMEP Eulerian oxidant model (Jonson et al., 1998, Jonson et al., 1999) and the Lagrangian oxidant model (EMEP, 1998) have been run for the whole year 1996 for comparison with the measurements of VOC; here we present some preliminary results for carbonyls. The carbonyl measurements are 8 hours samples, centered around noon, and are normally sampled twice a week. In the model comparison daily average concentrations were prepared from both the Eulerian and the Lagrangian models. Previous comparisons between the Lagrangian model and the VOC measurements (Solberg et al., 1995) have shown that the agreement was improved when

using the average of the calculated values at 6, 12 and 18 compared to using either the value at 12 only or the average of the whole day. However, to make the results the most comparable for the two models, daily averages were used in the present study.

As both the Eulerian and the Lagrangian models use lumped chemical reaction schemes for the organic chemistry, the model components are not directly comparable to the measured components. However, previous comparisons between the Lagrangian model and VOC measurements (Solberg et al., 1995) have shown a good agreement between modelled and measured formaldehyde and acetaldehyde. This probably reflects that these aldehydes are not particularly reaction specific, i.e. they originate from a large number of reactions, and in addition that they are in the bottom of the reaction chain, implying that a significant part of the initial pool of emitted VOC eventually goes through formaldehyde and acetaldehyde before being decomposed further to CO and CO2.

Figure 6.1 and Figure 6.2 show the measured formaldehyde and acetaldehyde, respectively, compared with the Lagrangian model (left column) and the Eulerian model (right column). Table 6.1 and Table 6.2 give the linear correlation coefficients and the average modelled concentration relative to the average measured concentration.

Table 6.1: Linear correlation coefficients between modelled and measured concentrations and the ratio between average modelled and average measured concentrations of formaldehyde for the Lagrangian and the Eulerian models, respectively. Only days with measurements were used.

Station	Linear corr. coeff.		[C]mod/[C]obs	
	Lagrange	Euler	Lagrange	Euler
Birkenes	0.68	0.27	0.69	2.71
Mace Head	0.67	0.32	0.66	1.72
Waldhof	0.68	0.41	0.62	2.16
Kosetice	0.37	0.12	0.49	1.68
Donon	0.41	0.06	0.67	1.99
Ispra	0.45	0.08	0.35	1.34

Table 6.2: Linear correlation coefficients between modelled and measured concentrations and the ratio between average modelled and average measured concentrations of acetaldehyde for the Lagrangian and the Eulerian models, respectively. Only days with measurements were used.

Station	Linear corr. coeff.		[C]mod/[C]obs	
	Lagrange	Euler	Lagrange	Euler
Birkenes	0.55	0.34	0.57	2.24
Mace Head	0.36	0.17	0.29	0.77
Waldhof	0.51	0.43	0.65	2.10
Kosetice	0.24	0.11	0.40	1.22
Donon	0.23	0.19	0.61	1.65
Ispra	0.28	0.09	0.36	1.22

The results show relatively large differences between the two models. The Lagrangian model gives higher linear correlation coefficients than the Eulerian model for both formaldehyde and acetaldehyde and for all the sites shown. Furthermore, whereas the Lagrangian model gives systematically lower concentrations than the measurements, the Eulerian model concentration are on average mostly higher than the measurements. These differences are present through the whole year, but the differences in averge concentrations between the two models are most pronounced in the winter season. The calculations for Mace Head show that in January-March the formaldehyde concentrations calculated by the Eulerian model are about a factor 3 higher compared to the Lagrangian model (and also significantly higher than the measurements). However, from April to the end of the year the Eulerian model gives formaldehyde concentration very close to the observations (the number of samples from Mace Head was few, though).

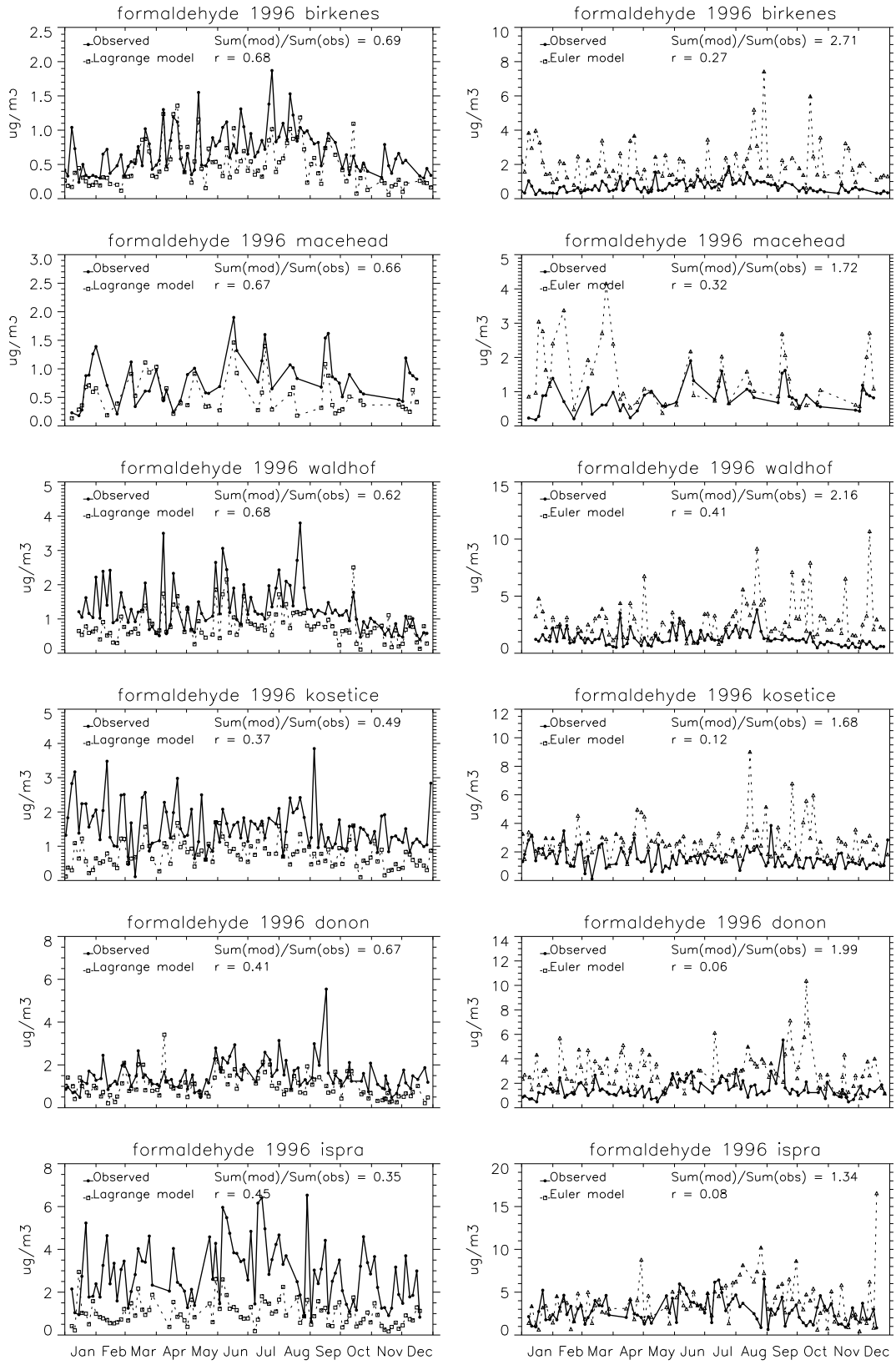

Figure 6.1: Measurements of formaldehyde in 1996 on six EMEP VOC stations (full line) together with daily avereges of concentrations calculated by the Lagrangian model (left side) and the Eulerian model (right side). All values are in ug/m3. The linear correlation coefficients (r) and the ratio of the average modelled concentration relative to the average measured concentration is also given.

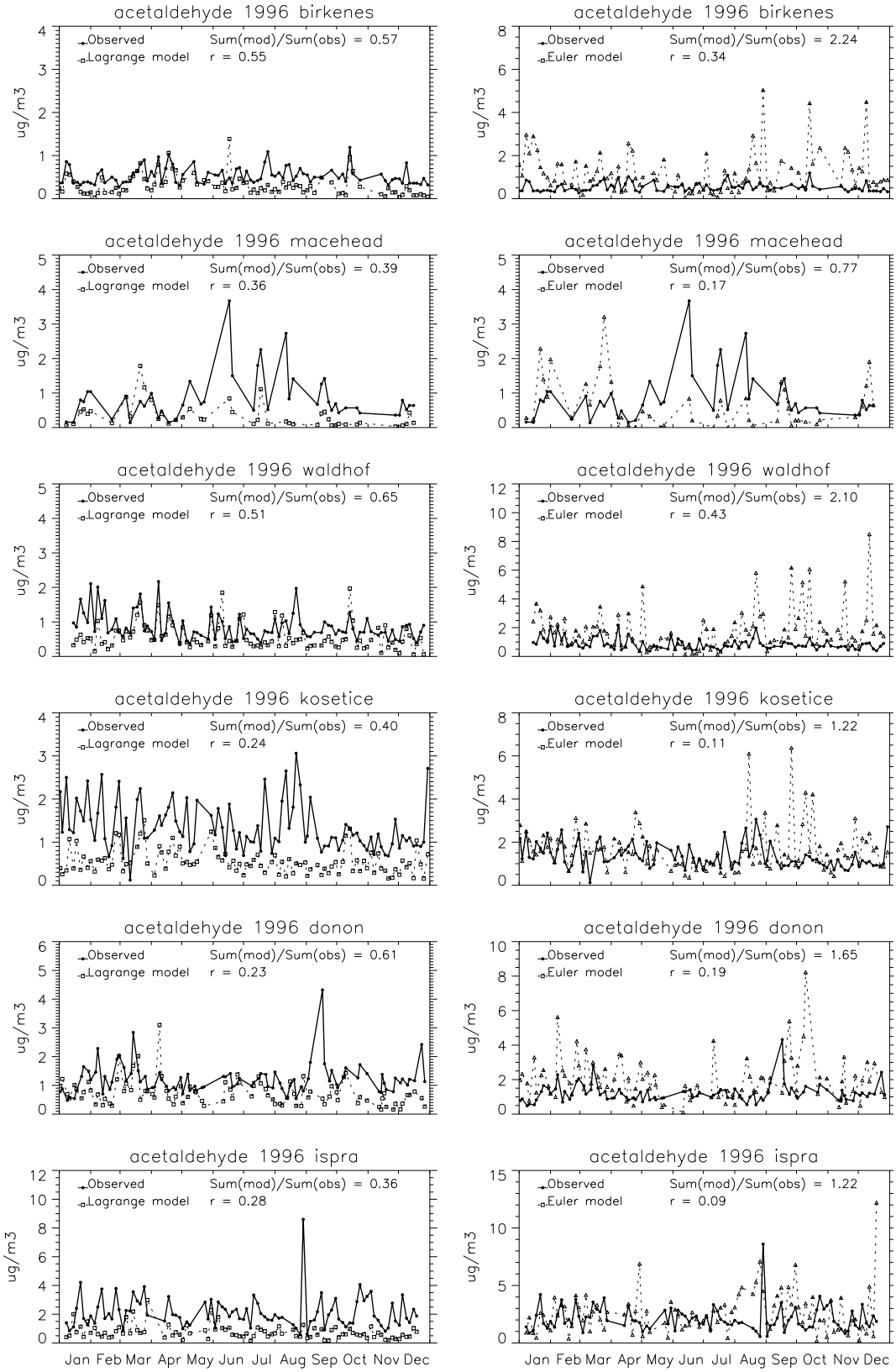

Figure 6.2: Same as Figure 6.1 for acetaldehyde

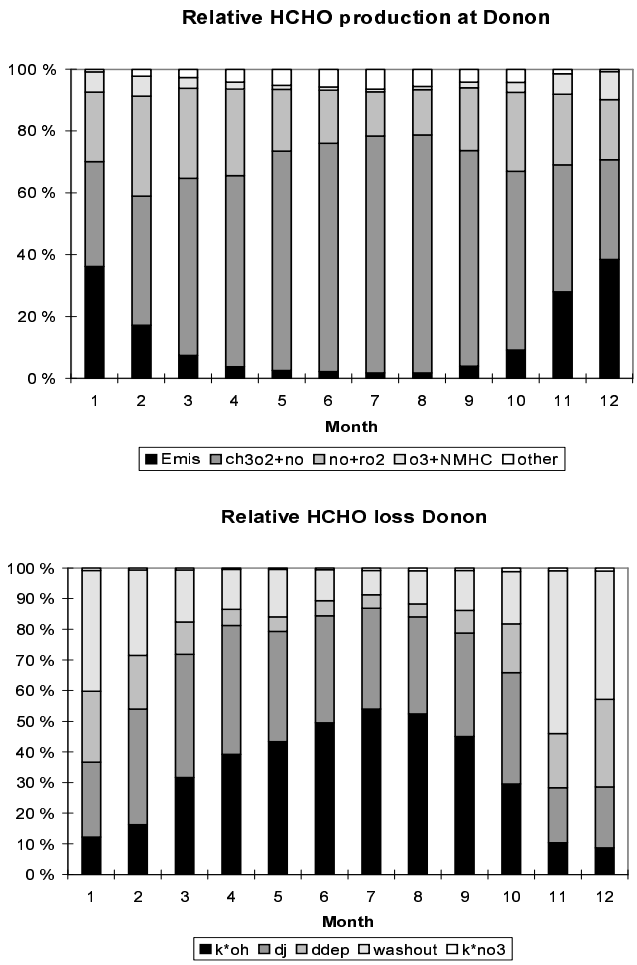

Figure 6.3: Monthly average production and loss terms for formaldehyde averaged along 4 days' back trajectories arriving at EMEP VOC site Donon, using the years 1993-1996.

6.3 Production and loss processes

Formaldehyde and acetaldehyde are both formed and lost by a large number of reactions and processes, and it is thus difficult to link the differences seen in the two models to certain model parameterisations. The Lagrangian model has been used to estimate the processes that control the formaldehyde concentrations. The model's individual loss and production terms for formaldehyde were integrated along the 4 days back trajectories, taking into account the effect of dilution, identical to the trajectory averaged quantities as defined by Simpson et al. (1995). Trajectory averages of the production and loss terms of HCHO were calculated through the years 1993-1996 and were subsequently averaged on a monthly basis, taking all years together. Figure 6.3 shows the calculated relative contribution from the different production and loss terms for Donon on the monthly basis.

It turned out that the four dominant production processes, as calculated by this procedure, were the primary emissions and chemical formation by the reactions NO + CH_3O_2, NO + RO_2 (R higher than methoxy) and O_3 + alkenes. The contribution from the other processes

were lumped together and are referred to as 'other' in the figures. The results furthermore indicate a marked seasonal cycle in the relative contribution for the individual production terms. The direct emissions contributed 25%-40% in winter (Nov.-Jan.) and almost negligible in the summer half year (April-Sep.). This reflects the reduced chemical degradation in winter combined with generally less efficient mixing.

Washout was the dominant loss process of formaldehyde in winter whereas the loss by OH is calculated to increase from less than 10% in winter to 40%-50% in summer. The relative of formaldehyde due to photolysis is calculated to have a smaller seasonal cycle than the HCHO + OH reaction, indicating a larger seasonal cycle in the OH concentration than compared with the photolyse rates of HCHO.

6.4 Discussion and Conclusions

The carbonyl calculations of the 3D Eulerian and the Lagrangian EMEP oxidant models have been compared to each other and to EMEP measurements.

The study has revealed substantial differences in the calculated concentrations and in model performance. In general the Lagrangian model gave a better agreement with the measurements than the Eulerian model, both with respect to correlations and absolute level for formaldehyde and acetaldehyde for the year as a whole. The Eulerian model gave substantially higher concentrations than the Lagrangian model in general, particularly for the aldehydes in winter. However, in winter the Lagrangian model underestimates the VOC relative to the measurements, possibly beacuse of vertical concentration gradients which are better simulated by the Eulerian model.

It is firstly important to stress that this comparison is very preliminary and more work is needed to check the data presented from both the models and measurements. The large discrepancy found so far between the Eulerian model and both the measurements and Lagrangian model is disturbing, but rather hard to explain, so at this stage coding errors cannot be ruled out. Perhaps most importantly, this comparative study has shown the importance of the VOC measurements for model validation. Some possible explanations for the differences are:

Vertical resolution differences The one-layer Lagrangian model is obviously not suited for describing vertical concentration gradients that may occur in winter with a stable thermal stratification. This will in particular affect compounds emitted at the ground, and, as shown by Figure 6.3, direct emissions are important for the formaldehyde concentration in mid-winter.

However, the two models' aldehyde concentrations also differ the rest of the year when vertical concentration gradients are of less importance and direct emission of formaldehyde is insignificant (Figure 6.3).

Vertical resolution can clearly explain why the Eulerian model has higher concentrations than the Lagrangian, especially in winter, but not why the modelled concentrations are much greater than the observations. In contrast, the earlier comparison of both models against measured NO_2 showed that the Eulerian model was much better able to simulate this compound than the Lagrangian.

VOC speciation and chemistry The VOC emission rates are based on the same data-base in the two models and should not cause any major differences. However, the VOC split, i.e. the relative distribution of the VOC emission on the individual species differ between the two models due to the different reaction schemes. Some harmonisation of the VOC emission splits have been attempted in the two models, but with different chemical schemes the effect for secondary VOC (as the aldehydes) is still unclear.

Both models used so-called condensed chemical mechanisms. It has been shown by Andersson-Sköld and Simpson (1999) that the HCHO predictions of the Lagrangian model scheme match very well those of a much more comprehensive (2000 reaction scheme) of Andersson-Sköld (1995), but no such evaluation is available for the Eulerian model mechanism. Furthermore, the reaction scheme for isoprene, which is an important contributor to formaldehyde during summer is much more simplified in the Eulerian model compared to the Lagrangian model (Simpson et al., 1995).

Deposition terms As shown in Figure 6.3, dry and wet deposition terms are important loss processes for formaldehyde in the Lagrangian model. These deposition terms are missing in the Eulerian model chemical scheme and this will probably explain to a some extent the higher Eulerian model concentrations, especially in wintertime.

Averaging times The measured values represent 8-hour averages centered around noon. The model values used here are daily averages. There is very little data available giving the diurnal variation of aldehydes in Europe, but the possibility exists that some systematic error is introduced into our comparison by this inconsistency.

Coding errors Usually, large discrepancies between modelled and observed values can be traced to errors in the model codes, reported measurements, or some inconsistencies between the two sets of data. As this comparison is very preliminiary we have not had time to completely rule out this type of problem.

Future work will concentrate on addressing as far as possible the above issues, in order to explain the discrepancies between especially the Eulerian model and the measurements. In order to aid future comparisons it will be useful to merge the different chemical reaction schemes into one common mechanism for use in EMEPs oxidant models, taking the best from the two present models. The handling of the VOC emissions (with respect to VOC speciation etc) should also be harmonised, and a common isoprene reaction scheme implemented. In this work the EMEP VOC measurements should have an essential role for evaluating the model development and performance. When a harmonised model chemistry has been implemented, a similar study between the Lagrangian and Eulerian model should be performed to identify differences induced by differences in parameterisation of e.g. mixing, advection and deposition.

6.5 Acknowledgements

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Chapter 7

The VOC speciation profile. Long-term measurements and model calculations.

S. Solberg

7.1 Introduction

The EMEP VOC measurement programme started with grab sampling of hydrocarbons in the middle of 1992, and 8-hours sampling of carbonyls in 1993. In this period the number of VOC stations have been in the range 4-10. In addition VOC measurements from a few monitoring sites outside the EMEP VOC network have shared their data with EMEP. Thus, regular VOC measurements (through the year) are available for several sites and years, providing an unique data base for model validation.

The VOC are particularly useful for oxidant model validation for several reasons. Firstly, they include a large number of components with chemical lifetimes ranging from a few hours to months. Secondly, most compounds are not strongly influenced by wet removal or dry deposition to surfaces, which is often difficult to calculate accurately in models due to local effects, and which thereby could complicate the comparison between model results and measurements. Thirdly, VOC are essential for the oxidant formation and therefore useful for testing both the emission rates and the intermediate compounds. On the other hand, using VOC for model validation is also difficult in several ways. The organic chemistry in the atmosphere is very complex and must inevitably be substantially simplified, e.g. by using lumped reaction schemes, in numerical models used for long-term calculations. Thus, a comparison between modeled and measured single components are not straight forward.

Both the total VOC emission and the VOC speciation (the relative distribution of the individual compounds) are important for the atmospheric chemistry and the oxidant formation. As the potential for ozone formation (POCP) varies substantially for the different VOC, the knowledge of the VOC speciation is essential for the modeling which makes the basis for abatement strategies related to VOC, ozone and NO_x.

In the following, some key points from a detailed model comparison with the VOC measurements are presented.

7.2 Procedure

The Lagrangian model (EMEP, 1998) was used to calculate a large number of individual hydrocarbons, following the procedure described by Hov et al. (1997). The modeling of the additional hydrocarbons was carried out by defining the individual emission rates for each individual species as well as their reaction rate constants with OH, ozone and NO_3. These species were then integrated along the trajectories, similar to the main components of the model, but without any feedback to the main model. Some modifications relative to the procedure presented

by Hov et al. (1997). were used for the initialization and emission estimates, but the basic principles were the same.

The emissions of the individual hydrocarbons were based on a recently updated VOC NAEI (National atmospheric emissions inventory) emission profile for the UK (PORG, 1997). That report gives the annual emissions (in ktonnes/year) for 1995 for about 50 individual VOC for each of the UN-ECE source categories except agriculture and nature. In the model the emission rates of the extra individual hydrocarbons were calculated by scaling the main model's total VOC emission for each source category and grid square by the VOC emission profile adopted from the PORG report.

The seasonal cycle of the emissions of the individual hydrocarbons was taken care of through the standard model's seasonal cycle of the emissions of the sum of VOC for each UN-ECE category (EMEP, 1998). This gives a day-to-day variation for the emission rates for each country and each sector through the year, and is based on data from the EUROTRAC subproject GEN-EMIS. This, however, introduces an uncertainty as the seasonal cycle of individual VOCs may differ from the seasonal cycle of the sum of VOC for the corresponding UN-ECE category. The traffic emission is e.g. made up of exhaust and evaporative compounds with different seasonal cycles due to changes in ambient temperature and in fuel composition. Another uncertainty in this procedure is the assumption that the VOC emission profile for each source category is equal to the UK profile everywhere, independent of country and location. Without more detailed emission speciation, however, this is the best approach available.

The initialization procedure is important as the chemical lifetime for certain compounds and at certain times of year may be longer than the integration time along the trajectories (4 days). Thus, a considerable part of the initial concentration may exist after 4 days of integration. The assimilated fields, i.e. pre-calculated concentration fields to parts of the EMEP grid (EMEP, 1998), were used also for initialization of the extra, individual VOC by scaling the pre-calculated sum of VOC with a fraction reflecting the emission fraction of that single VOC. In areas outside the assimilated region, monthly 25-percentiles of VOC measurements from the Arctic station at the Zeppelin Mountain, Spitsbergen were used for initialization.

7.3 Model results compared with the observations

The model was run for the years 1993-1996 for the six receptor sites Pallas, Utø, Birkenes, Waldhof, Kosetice and Starina, shown in Figure 7.1 Time series of modeled vs observed concentrations for all sites for the components ethane, ethene, acetylene, propane, propene, n-butane, i-butane, n-pentane, i-pentane and benzene are given in Appendix C.

In general the results show good agreements between the model calculations and the measurements. The linear correlation coefficients, r, are mostly above 0.7 for ethane, ethene, acetylene and propane. For the other components the correlation is more variable. However, for many of the time series the agreement is very good, with correlation coefficients above 0.8. With respect to the concentration level, the results show a general underestimation by the model compared to the measurements. This could be explained by the lack of vertical resolution in the Lagrangian model. Episodes with stable thermal stratification, and even inversions, often occurring in winter is not reproduced in the one-dimensional Lagrangian model which inevitably mixes the emitted VOC in the whole planetary boundary layer (PBL). Many of the episodic spikes in the measurements are reproduced by the model but at a lower concentration. This could be explained by the lack of complete mixing through the PBL, and also by an underestimation of the emission rates.

The VOC speciation, i.e. the relative concentration levels of the VOC is important with respect to the potential for ozone formation. The VOC speciation measured and given by the model calculations is indicated by Figure 7.2. This Figure shows the agreement between the measured and modelled VOC medians and 75-percentiles, respectively, during the six-months winter period October-March for the six stations, separately. The medians indicate the long-term seasonal concentration, whereas the 75-percentiles are more indicative of the elevated episodes. Figure 7.2 indicates that the calculated median VOC speciation profile at Pallas, Birkenes and Utø agrees very well with the measured median profile, although ethane at the two Finnish sites are slightly higher in the calculations than measured. As these sites are far away from the large continental European source regions, this indicates that the model assumptions regarding initial concentrations and the VOC emission profile are reasonable in an overall

Figure 7.1: Map showing the six monitoring sites for VOC used in the study.

perspective. The other sites are presumably more exposed to urban areas and emission areas within the nearest 50-100 km and thus more sensitive to the vertical mixing and meteorological situations. These sites all show considerably higher measured median concentrations of ethene and acetylene than modelled, whereas the median of n-butane is lower in the measurements than modelled. The same was noted in the results of the VOC modeling from 1994 reported by Solberg et al. (1995).

Both ethene and acetylene are tracers of vehicle exhaust and originates almost exclusively from that source. Without a more detailed meteorological evaluation of the Lagrangian model results or calculations with another model with a finer vertical resolution, it is difficult to judge whether the discrepancy for ethene and acetylene is due to lack of vertical resolution in the model or an underestimation of exhaust emissions. The comparison of the modelled and measured 75 percentiles (right column of Figure 2) gives much the same results as for the medians, except that the discrepancy for ethene and acetylene is also seen for Birkenes, indicating episodes of traffic emissions at this site as well. Another difference is that the measured 75 percentile of ethane is significantly higher than the model calculations as well.

7.4 Conclusions and recommendations

Some key points of a detailed comparison between the Lagrangian model and the VOC measurements have been shown. In general the results were very satisfactory. Results from the sites most remote from the main emission areas in Europe, Birkenes, Utø and Pallas, indicated a good agreement between modelled and measured concentrations on an overall basis. This indicates that the model assumptions regarding initial concentrations and the VOC emission profile (for the 10 components shown) are reasonable in an overall perspective. However, for

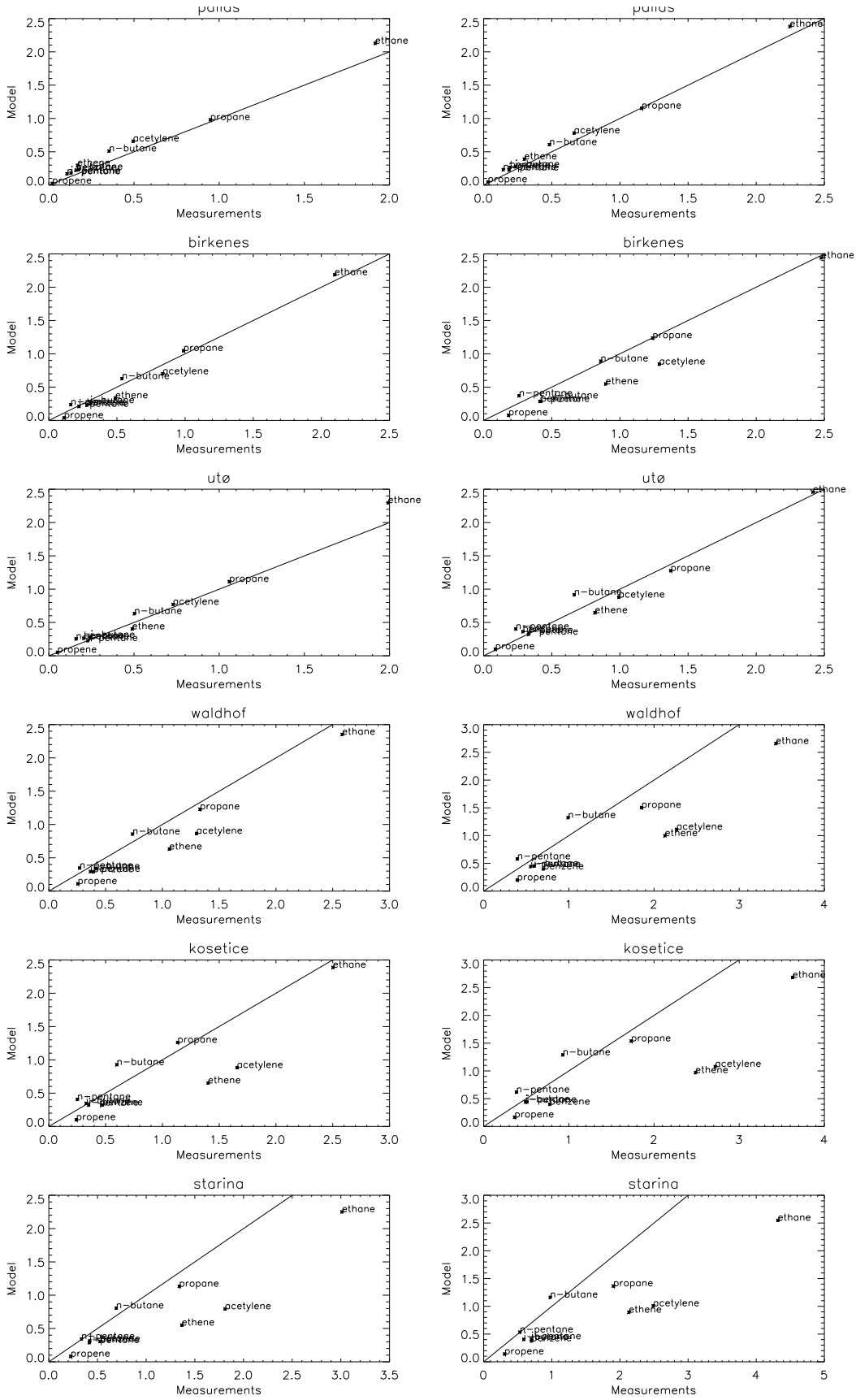

Figure 7.2: The median (left column) and 75-percentile (right column) of the measured vs the modelled concentrations of the 10 hydrocarbons ethane, ethene, acetylene, propane, propene, n-butane, i-butane, n-pentane, i-pentane and benzene based on the six-months period October - March for all years together. All values are in ppb. The straight line indicates the 1:1 relationship between measured and modelled values.

some periods there are discrepancies which need to be studied in more detail. For the other, continental, sites and when looking at peak values at the Nordic sites, the results show a systematic underestimation of vehicle exhaust compounds (ethene and acetylene) by the model compared to the measurements. This could either reflect the lack of vertical resolution in the one-layer Lagrangian model or a underestimation of this type of emissions. n-Butane on the other hand is somewhat overestimated by the model (in winter) when compared with the measurements, possibly reflecting a different seasonal cycle in the emissions of evaporative compounds than applied in the model.

This study, although rather limited as presented here, has shown the importance of the VOC measurement data for validation of models and emission estimates. Based on this experience it is recommended to explore further the importance of a finer vertical resolution for the model performance - either by studying the Lagrangian model results in a closer meteorological perspective or by performing similar VOC calculations by the Eulerian model, preferably both. Secondly, the effects of introducing an improved simulation of the seasonal variation for each of the individual VOC could be studied with respect to the model performance. Thirdly, VOC speciated emissions on a country-basis would be advantageous to incorporate in the calculations wherever such data are available. Lastly, the uncertainty introduced by comparing model calculations with grab samples with a sampling time of only 20 min, has not been discussed here, but is clearly a matter which need to be evaluated. This could e.g. be done by comparing the VOC calculations with measurements from sites having a continuous VOC monitor.

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Chapter 8

Conclusions and outlook

8.1 Summary and conclusions

This summary report for photochemical oxidants has covered a number of topics related to the Lagrangian and Eulerian photo-oxidant models and their evaluation. In general, work in 1998-1999 has concentrated on application, comparison and evaluation of the two oxidant models, against each other and in a preliminary study against measurements.

The emissions scenarios covered in this years' report are rather different in character to those presented previously. Many scenarios have been designed to support the EU's acidification and ozone strategy and for the continued negotiations for the 2nd NO_x Protocol, especially in order to verify and provide more detail concerning emissions strategies worked out with the IIASA RAINS-ozone model (Amann et al., 1999). Additionally, the work under the INFOS project has looked at each emission-sector in turn, especially for the road-transport sector. These scenarios made use of very detailed information (e.g. about VOC speciation in different vehicle types), to attempt as realistic a simulation as possible of the different sectors.

Some of the main conclusions to be drawn from this report are:

- In spite of some differences in absolute levels, both MSC-W oxidant models arrive at similar conclusions for air quality in the year 2010. Despite significant improvements as a result of currently planned measures, levels of AOT40 will still lie significantly over the critical thresholds for vegetation damage. AOT60 levels are also predicted to be above zero in 2010 in most parts of Europe, suggesting continued risk of health effects. These statements still seem to be true after application of even more stringent control measures than currently planned, as shown by the G5/2 calculations.

- Model results with joint runs of the global University of Oslo CTM2 model and the 3-D EMEP model have shown that despite increased ozone levels in the free troposphere by the year 2010, ozone levels (and especially AOT60) in the boundary layer over Europe are expected to decrease significantly as a result of European control measures.

- Both EMEP models showed rather similar behaviour when comparing the effects of NO_x control against that of VOC control in sector-specific scenarios and the country-to-grid calculations. Indeed in the sector-specific scenarios the changes in AOT40 and AOT60 were very similar in the two models for about 95% of grid squares. However, the exceptions occurred at the 5% of grid squares with highest AOT values. These extreme grid cells sometimes showed very different or even opposite behaviour between the two models. In the country-to-grid calculations the two models showed reassuringly similar patterns of ozone increases and decreases in response to reduced NO_x and NMVOC emissions, but the 3-D model consistently showed larger responses than the Lagrangian.

- The effects of eliminating emissions from different SNAP sectors has been investigated for the year 2010 INFOS scenario. The sectors contributing most to AOT levels in this analysis were found to be road transport, solvents and industrial processes. Within the

road transport sector, heavy-duty vehicles and evaporative emissions are predicted to make the largest contributions, followed by passenger car exhaust.

- A preliminary comparison of country-to-grid calculations with the Lagrangian and Eulerian models showed rather similar results, except for a general pattern that the 3-D model predicts stronger changes than the Lagrangian model. Thus, both the negative and positive NO_x effects are stronger in the 3-D model, as are the always-positive VOC effects. However, given the known uncertainties of modelling in general and AOT predictions in particular, the large similarity between the results of the two models is encouraging and gives continues confidence in the usage of source-receptor matrices in emission control analysis.

- The comparison of the models results against measured carbonyl concentrations produced the most surprising result of the studies presented, that the Lagrangian model performed significantly better than the Eulerian in predicting both HCHO and CH_3CHO concentrations. This result is in contradiction to the better performance of the Eulerian model for other species such as NO_2, and should be regarded with caution until a more thorough examination of the data is possible. The result does underline the need to continuously evaluate models against species other than ozone though, and indeed the combination of carbonyl and hydrocarbon measurements at EMEP sites is extremely valuable for such model evaluations.

- Comparison of calculated versus observed hydrocarbon concentrations using the Lagrangian model has shown very satisfactory agreement. Some discrepancies were found, but the Lagrangian model's lack of vertical resolution complicates a detailed comparison.

Most of the differences in response to NO_x and NMVOC control between the two EMEP models can be readily explained by the greater resolution of the Eulerian model, even though other factors such as chemistry differences may play a role. Especially surface NO_x sources are much better resolved in the 3-D model (with a lower-layer thickness of ca. 40m) than in the 1-layer Lagrangian model. This gives greater NO_x concentrations in the lowest layer, which increases the model's sensitivity to NMVOC over urban areas, and gives a greater 'negative' effect when the NO_x emissions are reduced because of the pronounced titration reaction $NO + O_3 \rightarrow NO_2$.

Detailed discussions of the relationships between resolution and NOx-NMVOC sensitivity can be found in for example Sillman et al. (1990a,b) or Pleim and Ching (1993). However, as pointed out by Pleim and Ching (1993) it is not always clear when improved resolution gives more realistic ozone formation. The basic feature of this is that NO_x molecules in a clean (low-NO_x) area will generate ozone more efficiently than in a high-NO_x area, indeed the latter may even act as a sink for ozone. As an example of the difficulties though, consider a large NO_x source (e.g. an industrial plant) which may emit NO_x emissions into a relatively clean area. With a large grid size (e.g. in the Lagrangian model), the grid-average NO_x concentrations may be low, and thus the emissions will generate ozone probably more effectively than in the real atmosphere. Other emissions within the grid square (e.g. scattered small towns, roads) may be treated fairly realistically in this resolution though. In contrast, with a 50 km grid, and fine (e.g. 40m) vertical resolution, this large NO_x source will give very high NO_x concentrations within the grid, probably suppressing ozone formation. This may or may not be realistic for the large point source, but is probably an incorrect description of the atmosphere for other sources within the same 50 km grid.

Ideally of course, model resolutions would be so fine that such problems were made irrelevant, but with today's computers the problem is unavoidable. Thus, it is probably true that in most situations the 3-D model gives a more realistic description of the European atmosphere than the Lagrangian, but this is an assumption and may not always be correct. Thus, for the time being, there is great value in comparing the results of the two models, especially in assessing control strategy predictions.

8.2 Outlook

Development work will continue with the 3-D Eulerian model as this offers the most potential for improving the science within the EMEP models.

An immediate task is to improve the deposition modules in order to tackle the calculation of fluxes to vegetation. This will build upon the work of Emberson et al. (1996) and initial progress will be reported in an EMEP Note shortly. Aerosol modules also need to be added to the oxidant model's capabilities.

The measurements of carbonyls and hydrocarbons available within EMEP have proven extremely valuable in evaluating both the models and the emission inventories. It is recommended that such measurements continue and if possible be enhanced with other key-species for evaluating photochemical models, for example NO_y constituents and radical concentrations.

Apart from the problem with $HCHO/CH_3CHO$ predictions mentioned in this report, the 3-D model has much greater potential for evaluating measurements than the Lagrangian model, because of its much greater vertical and horizontal resolution. The model is particularly useful for dealing with pollutants which are know to have large vertical gradients much of the time (e.g. NO_2). Serious gaps in the measured species do exist, however, and efforts should be made to make use of data available from other activities, for example EUROTRAC or national programmes. By actively pursuing such comparisons both the model and its inputs (emissions, land-use) can be continuously evaluated and improved.

As noted in chapter 6, it will be useful to merge the different chemical reaction schemes into one common mechanism for use in EMEP's oxidant models, taking the best from the two present models. In this work the EMEP VOC measurements should have an essential role for evaluating the model development and performance.

Such changes should be made such that in future it is much easier to change chemical mechanisms than at present, and to add such components as aerosol chemical schemes in a flexible and efficient manner.

8.3 References

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Appendices