CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

UN/ECE INTERNATIONAL CO-OPERATIVE PROGRAMME ON EFFECTS ON MATERIALS, INCLUDING HISTORIC AND CULTURAL MONUMENTS

Report No. 35 Draft

Results from the multipollutant programme: Corrosion attack on carbon steel after 1and 2 years of exposure (1997-1999)

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Summary

This report summarizes the results of evaluation of weight changes and corrosion losses of carbon steel after 1 and 2 years of exposure from 28 test sites in period 1997/98 and 1997/99 in frame of UN ECE ICP Effects on Materials. For selected test sites the analysis of amount of Cl^- in rust layer was performed.

Range of corrosion losses evaluated after exposure within the multipollutant programme is in general lower as within the original programme (1987-1995) caused by the lower corrosion impact on the industrial test sites.

For the results of the first year of exposure the correlation and regression analysis were performed. Results documents the remaining higher positive correlation of SO_2, high positive correlation between corrosion losses and conductivity and pH of precipitation, positive effect of NO_2 cannot be omitted. Negative correlation for O_3 was evaluated in accordance within the resluts for the original UN ICP Materials.

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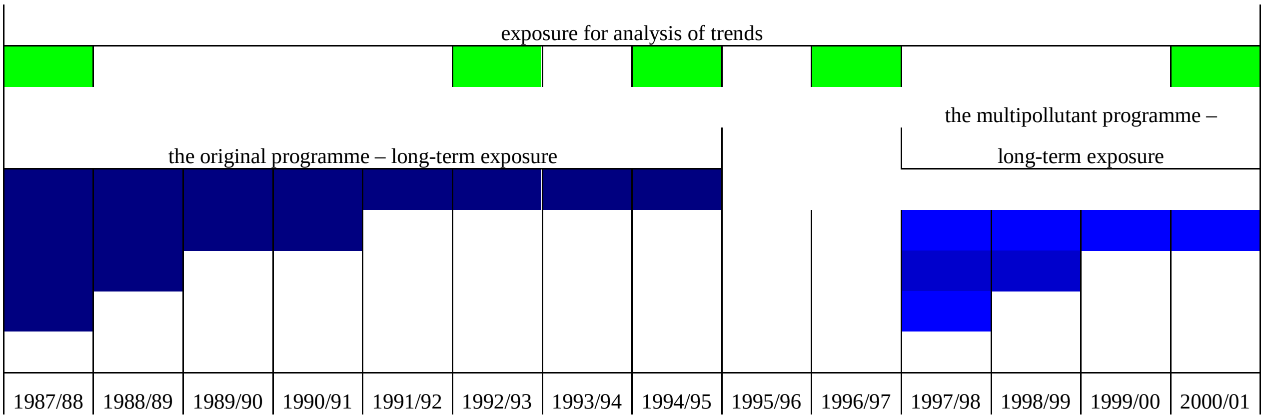
Introduction

The UN ECE International Cooperative Programme was performed to a quantitative evaluation of the effect of sulphur pollutants in combination with NO_x and other pollutants as well as climatic parameters on the atmospheric corrosion of important materials. The programme was based on an international field exposure on 39 test-sites in a wide geographical zone of the 13 Signatories to the Convention on Long-Range Transboundary Air Pollution. The original field exposure was performed in 1987-1995. The programme has formed a basis for derivation of D/R fuctions and for deeper knowledge of expected serviceability of materials and systems.

Within 1985 - 1995 the environmental situation have been changed. The SO_2 concentrations significantly decreased in many European countries whereas the concentration of NO_2 and O_3 are nearly remaining. In this changed pollution situation it is necessary to study corrosion effects of the complex atmospheric pollution. Thus in 1997 the multipollutant exposure programme has continued. Only selected test sites from the first stage of programme and new countries with new test sites were included in this programme. The new programme includes 18 countries with 29 test sites.

Time schedule of exposure programme



The new aims of programme are quantitative evaluation of multipollutant effects of combination of pollutants (SO_2, NO_x, O_3 and others) and run-off of metals from exposed material surfaces as a potential environmental risk.

SVUOM has been subcentre responsible of exposure and evaluation of samples of carbon steel in the multipollutant programme.

1. Methods

1.1. Characterization of material

Plates of unalloyed carbon steel (with C < 0.2 %, P < 0.07 %, Cr < 0.07 % according to CSN 11373) with dimentions 100 x 150 x 0,5 mm were used, triplicate per exposure period.

Before the exposure the samples were degreased in alcaline degreaser, rinsed with ethanol, dried and weighed. After exposure samples were weighed and the corrosion products were removed using pickling method according to ISO 8407 (Annex A - solution C 3.5.).

1.2. Exposure of samples

Methods prescribed in the Technical manual to the UN ECE International Cooperative Programme on Effects on Materials including Historic and Cultural Monuments (Report No 1, June 1988) were applied for exposure of samples on test sites.

The field exposure programme was started in autumn of 1997. Exposure periods were 1 and 2 years (exposure continues for 4 years). The list of test sites are in Table 1 and Figure 1.

Samples were exposed on racks in open atmosphere (unsheltered exposure) according to ISO 8565 and under special shelter protected samples against precipitation and dust depositions (shelter exposure).

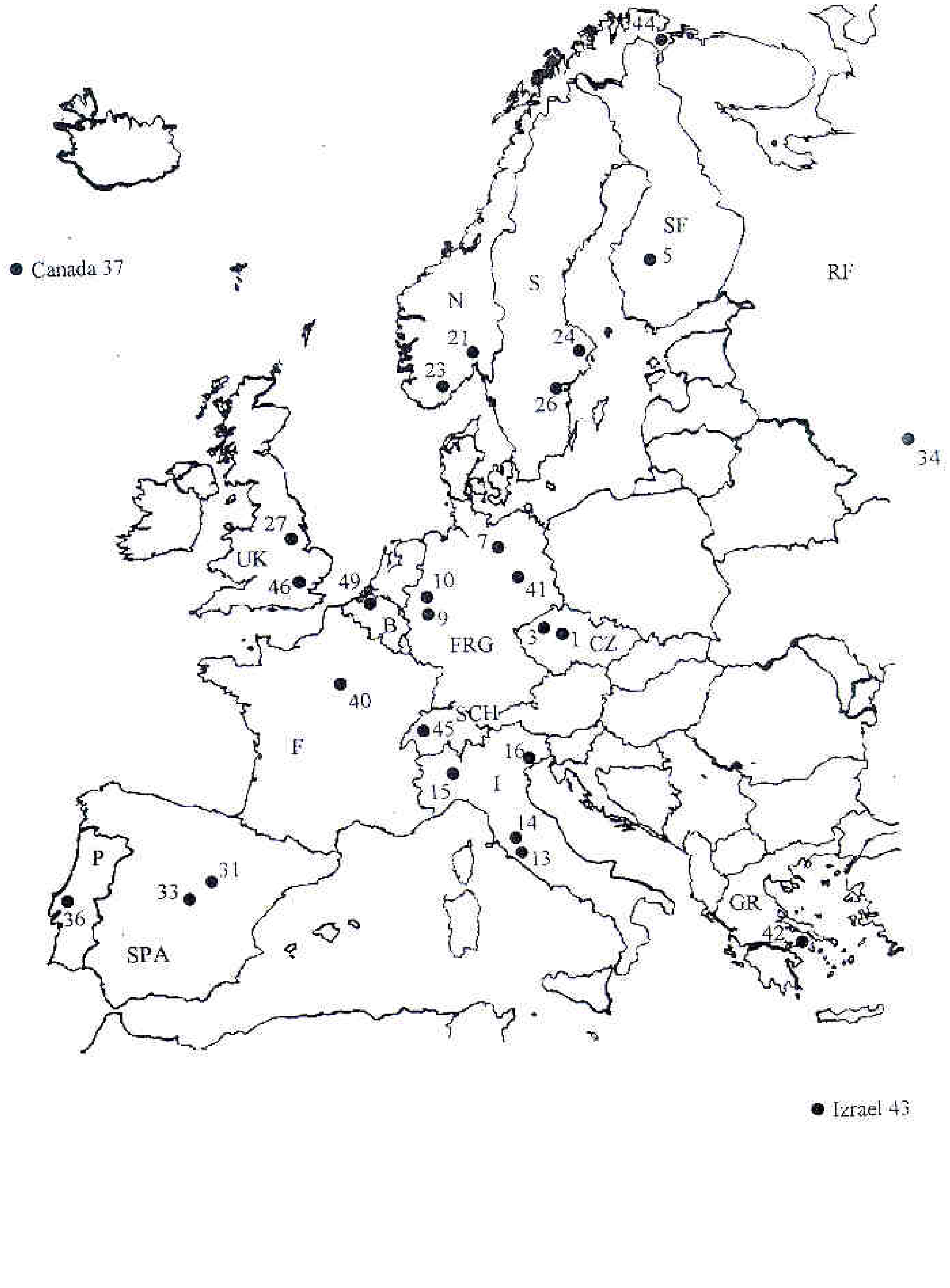
1.3. Measurement of environmental data

Basic climatic parameters (temperature, relative humidity and intensite of sunshine radiation), concentration of gaseous pollution (SO_2, NO_x, O_3) and precipitation (amount, pH, conductivity, amount of SO_4^{2-}, NO_3^-, Cl^-, and NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+} if available) were measured on each test site. All data were reported to, and completed by the Norwegian Institute for Air Research - NILU, which also checked the quality of the data. Environmetal data for period 1997-99 are in the Report No. 41.

Table 1 - The list of test sites

Country	Test site	Typ of atmosphere
the Czech Republic	1 Prague 3 Kopisty	urban-industrial industrial
Finland	5 Ahtari	rural
the Federal Rebublic of Germany	7 Waldhof-Langenbrugge 9 Langenfeld 10 Bottrop 41 Berlin	rural rural industrial urban
Italy	13 Rome 14 Casaccia 15 Milan 16 Venice	urban rural urban-industrial urban
Norwey	21 Oslo 23 Birkenes 44 Svanvik	urban rural rural
Sweden	24 Stockholm South 26 Aspvreten	urban rural
the United Kingdom	27 Lincoln Cathedral 46 London	urban urban
Spain	31 Madrid 33 Toledo	urban rural
the Russian Federation	34 Moskow	urban-industrial
Portugal	36 Lisbon	urban
Canada	37 Dorset	rural
France	40 Paris	urban
Greece	42 Athens	urban
Israel	43 Tel Aviv	urban
Switzerland	45 Chaumont	rural
USA	47 Los Angeles	urban-marine
Belgium	49 Antverps	urban-marine

Figure 1 – Map showing location of test sites



2. Results

2.1. Evaluation methods

The visual evaluation of layer of corrosion products were done. The character of rust layer can easely indicate nonuniform corrosion behaviour and nonuniform condition of exposure than layer of corrosion products of other exposed materials. So from this evaluation it was clear the samples under shelter on test site No. 35 (Lahemaa) were in contact each other and results from this test sites were excluded from treatment.

The basic informations about corrosion behaviour of carbon steel in different environmental condition give the value of weight change and mass loss.

For selected test sites the analysis of amount of soluble Cl^- in rust layer was performed.

2.2. Environmental characteristics

Average yearly climatic and pollution characteristics with expected influence for atmospheric corrosion of carbon steel for exposure periods 1997/1998 and 1997/99 are in Tables 2 and 3. The information about environment is not complete, some data are not avaliable yet.

Average yearly teperature is in range from –1.5^{0}C (44 Svanvik) to 25,4^{0}C (43 Tel Aviv). Test site 44 is the only one from the set with average yearly temperature below 0^{0}C. The mean value of average yearly temperature is about 10 ^{0}C. The mean of average yearly value of relative humidity is about 75%. Test sites with the significantly low value of relative humidity are 33 Toledo and 13 Rome.

The pollution situation is different on every test site. In comparison with the programme exposure in period 1987-95 the concentration of SO_2 was reduced significantly. The most of test sites (19) are in classified interval P_0 (according to ISO 9223) and test site with the highest concentration of SO_2 is 43 Tel Aviv – average value in period 1997/99 is 45,4 µg.m^{-3}. The other test sites with significant influence of SO_2 are 3 Kopisty, 10 Bottrop, 34 Moscow and 49 Antverps with mean value about 20 µg.m^{-3} of SO_2. On these test sites with relative high SO_2 concentration the decreasing of concentration continue. The concentration of NO_x various from about 1,0 µg.m^{-3} (23 Birkenes, 35 Lahemma, 44 Svanvik) to 70 – 90 µg.m^{-3} (15 Milan, 40 Paris).

The new mandatory and systematicaly measured pollution is concentration of ozone. Test sites with dominant effect of O_3 are 5 Athari, 7 Waldhof-Langenbrugge, 23 Birkenes, 26 Aspvreten, 33 Toledo, 35 Lahemaa, 37 Dorset, 44 Svanvik and 45 Chaumont. On these test sites the concentration of other gaseous pollutants are very low.

The concentration of H^+ in precipitation various from 4,25 to 6,71.

In this programme the concentration of chloride is measured only in precipitation. From these measurement it seems that test sites with significant influence of chloride are only 27 Lincoln Cathedral, 36 Lisbon and 46 London. Especially the test sites in U.K. show great amount of chlorides in precipitation – above 100 mg.l^{-1}.

Table 2 - Climatic parameters and pollution characteristics for period 1997-98 (mean year's values)

No Site	Temp [°C]	RH [%]	SO_2 [µg/m³]	NO_2 [µg/m³]	O_3 [µg/m³]	pH	Cl^- [mg/l]	Cond [µS/cm]
1 Prague	9,9	76	15,3	23,7	47,0	5,56	1,88	27,6
3 Kopisty	9,9	82	23,0	32,6	55,0	4,62	1,73	44,6
5 Ahtari	2,7	80	0,9	3,0	60,0	4,74	0,15	10,7
7 Waldhof-Langenbrugge	9,1	83	2,1	8,7	48,0	5,04	0,73	17,6
9 Langenfeld	10,9	80	8,3	33,5	33,0			
10 Bottrop	11,5	80	24,6	38,2	30,0	5,10	1,64	28,2
13 Rome	20,7	62	3,8	34,5	31,8			
14 Casaccia	14,5	74	5,2	19,3	28,6			
15 Milan	14,6	68	15,3	82,3	38,5			
16 Venice								
21 Oslo	6,4	78	4,0	27,9	35,4	5,66	1,03	23,0
23 Birkenes	5,9	79	0,2	1,1	55,0	4,50	1,50	25,5
24 Stockholm South	6,7	76	2,6	20,3	44,0	4,63	0,48	17,1
26 Aspvreten	5,8	87	0,6	2,9	51,0	4,59	0,57	17,3
27 Lincoln Cathedral	10,1		8,4	18,0	51,1	4,90	103,1	
31 Madrid			*4,8*	*26,9*		*5,83*	*1,05*	*12,5*
33 Toledo	*10,8*	*67*	1,6	11,2	88,5	5,95	0,89	14,8
34 Moskow	5,5	74	*31,5*	28,0	42,0	6,69	1,67	51,1
35 Lahemaa	5,0	82	*0,5*	0,7	58,0	5,16	0,78	19,4
36 Lisbon	17,1	62	17,8	40,8	11,2	6,48	13,44	68,9
37 Dorset	6,2	75	2,5	*9,4*	62,5	4,33	0,12	26,3
40 Paris	13,4	67	14,2	70,0	31,0	5,71	2,47	43,7
41 Berlin	10,1	77	10,9	37,7	47,0		2,33	
43 Tel Aviv	24,1	82	31,4	35,5	36,8			
44 Svanvik	- 1,8		7,5	0,9	54,0	4,77	1,87	20,7
45 Chaumont	6,9	77	1,3	7,7	86,0	4,99	0,18	9,4
46 London	12,5		*6,3*	44,9	37,6	*6,00*	*143,8*	
47 Los Angeles	17,2	64	0,6	22,5	50,4	*5,87*		
49 Antverps	11,8	75	24,2	53,7	29,7	*5,25*	*3,50*	*37,5*

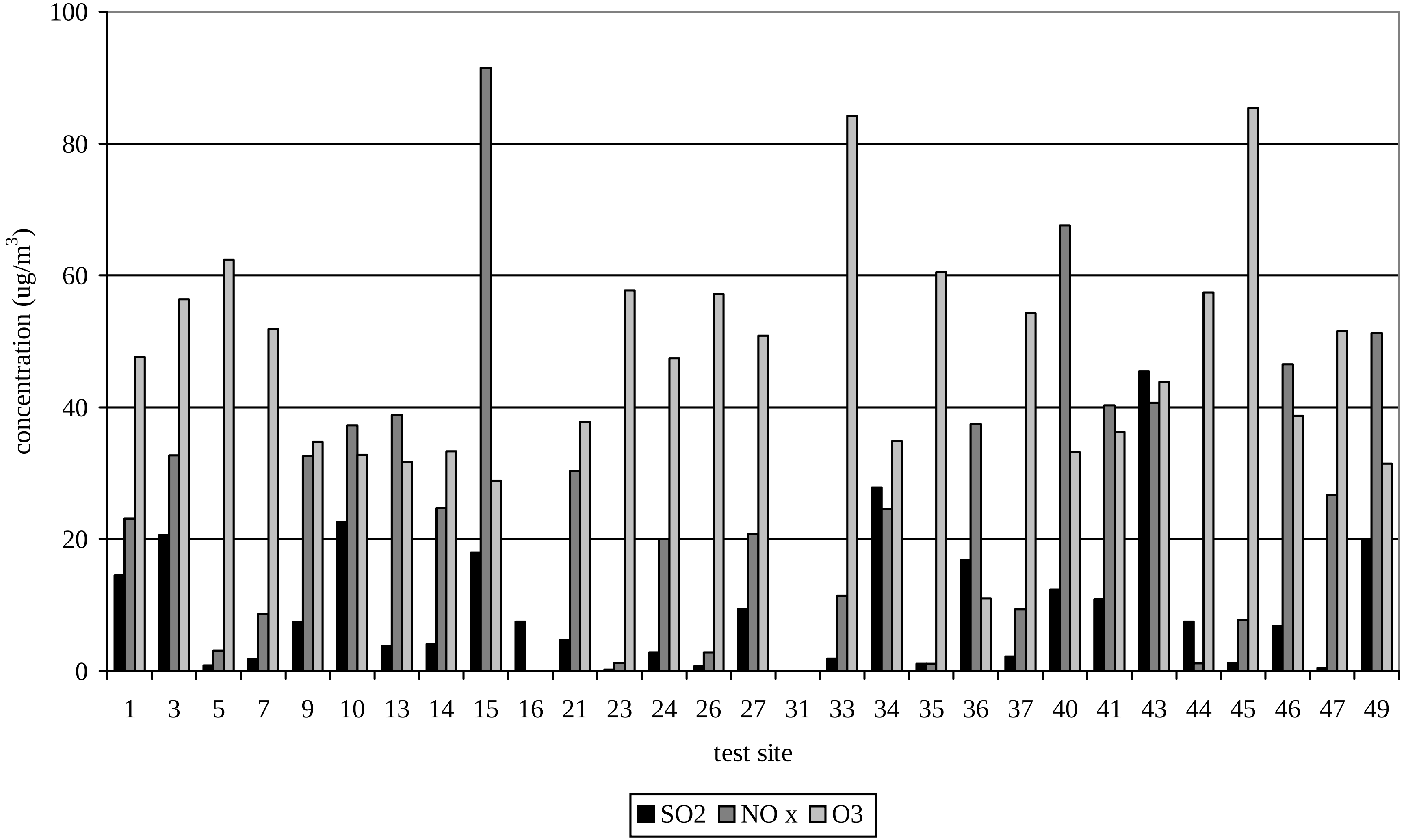
data written in Italic are from 6 – 9 months of measurents

Table 3 - Climatic parameters and pollution characteristics for period 1998-99 (mean year's values)

No Site	Temp [°C]	RH [%]	SO_2 [µg/m^3]	NO_2 [µg/m^3]	O_3 [µg/m^3]	pH	Cl^- [mg/l]	Cond [µS/cm]
1 Prague	9,6	76	13,6	22,5	48,2	5,38	2,81	28,4
3 Kopisty	9,4	78	18,4	32,7	57,7	4,84	1,93	59,6
5 Ahtari	2,8	79	0,9	3,2	64,8	4,63	0,16	14,4
7 Waldhof-Langenbrugge	9,4	78	1,5	8,7	55,7	5,29	1,41	20,2
9 Langenfeld	11,2	78	6,5	31,7	36,5			
10 Bottrop	13,5	81	20,5	36,1	35,5			
13 Rome	19,0	65		43,0	31,6			
14 Casaccia	16,1	67	3,0	30,1	38,0			
15 Milan	*11,3*	*77*	*20,6*	*100,6*	*19,3*			
16 Venice	13,5	82	7,5			*6,65*		
21 Oslo	6,5	78	5,3	32,8	40,2	4,68	0,82	22,3
23 Birkenes	5,9	81	0,3	1,4	60,5	4,57	1,90	16,2
24 Stockholm South	7,7	77	3,2	19,6	50,8	4,70	0,39	14,5
26 Aspvreten	6,0	84	0,8	2,7	63,2	4,57	0,71	19,2
27 Lincoln Cathedral	*6,0*		10,1	23,7	50,7	*5,66*	*68,7*	
31 Madrid								
33 Toledo	14,2	53	2,2	11,9	79,9	6,00	0,76	12,6
34 Moskow	6,3	70	24,2	21,2	*27,7*	6,61	1,34	*42,5*
35 Lahemaa	5,6	79	1,7	1,5	63,0	4,99	0,64	19,6
36 Lisbon	17,1	68	15,9	34,2	*10,9*	6.69	17.42	162.9
37 Dorset	6,6	73	*1,9*		*46,0*	*4,25*	*0,13*	*36,3*
40 Paris	13,6	*67*	*10,6*	*65,1*	35,4	6,66	2,64	46,6
41 Berlin	10,6	74	10,8	42,9	25,6		1,26	
43 Tel Aviv	26,6	88	59,4	45,9	50,9			
44 Svanvik	-1,1	77	7,5	1,4	60,8	4,80	1,35	19,8
45 Chaumont	6,2	80	1,2	7,7	84,9	4,91	0,20	10,3
46 London	*7,7*		7,4	48,1	39,8	*6,71*	*212,4*	
47 Los Angeles	15,8	65	0,4	31,0	52,8	*6,06*	*4,03*	*30,3*
49 Antverps	12,7	74	15,3	48,9	33,3	4,95	*4,70*	*44,2*

data written in Italic are from 6 – 9 months of measurents

Figure 2 – Gaseous pollutants – mean yearly average values in period 1997-99



2.3. Weight changes of carbon steel

Corrosion products of carbon steel exposed in atmosphere are the mixture of compounds depending on conditions of exposure and pollution situation. There is a difference between thickness and homogenity of layer formed under shelter and in open atmosphere.

unsheltered sample

sheltered sample

2 years of exposure on test site 3 Kopisty

Weight changes of exposed samples of carbon steel are summarized in Table 4 and 5 and Figure 3.

After 1 year of exposure the mass of unsheltered samples increased on all test sites except test site 43 Tel Aviv. After 2 years of exposure the weight changes were negative for samples exposed on test sites 10 Bottrop, 16 Venice, 27 Lincoln Cathedral, 36 Lisbon and 43 Tel Aviv.

After 1 year of exposure the weight changes of sheltered samples were negative for samples exposed on test sites 3 Kopisty, 10 Bottrop, 36 Lisbon, 44 Svanvik and 49 Antverps. After 2 years of exposure except these test sites negative weight changes were evaluated for samples exposed on test sites 1 Prague and 41 Berlin.

Mass increase of weight of exposed steel samples gives a complex and not well defined information. The measured value represents a differences of mass of corrosion products originated at impact of environment and part of them removed by run-off errosion and fall out of non adherent particles and sub-layers.

Table 4 – Weight increase ($g.m^{-2}$) of carbon steel after one year exposure – period 1997/98

Test site	open exposure	shelter exposure
	m_{un1}	m_{sh1}
1 Prague	55,7	1,0
3 Kopisty	79,7	-4,3
5 Ahtari	23,3	11,0
7 Waldhof-Langenbrugge	40,0	22,7
9 Langenfeld	23,0	21,3
10 Bottrop	18,7	-3,7
13 Rome	30,0	22,0
14 Cassacia	47,0	43,7
15 Milan	36,7	10,0
16 Venice	6,3	17,0
21 Oslo	38,7	11,3
23 Birkenes	33,3	9,0
24 Stockholm South	38,3	20,7
26 Aspvreten	-	6,3
27 Lincoln Cathedral	9,0	21,7
31 Madrid	33,7	12,0
33 Toledo	26,0	9,3
34 Moskow	34,0	17,7
35 Lahemaa	31,7	not evaluated
36 Lisbon	6,0	-10,7
37 Dorset	51,7	15,3
40 Paris	28,0	13,0
41 Berlin	57,3	57,3
43 Tel Aviv	-48,7	9,0
44 Svanvik	53,7	-0,3
45 Chaumont	30,3	13,0
46 London	10,0	12,0
47 Los Angeles	58,7	not exposed
49 Anvterps	51,7	-3,7

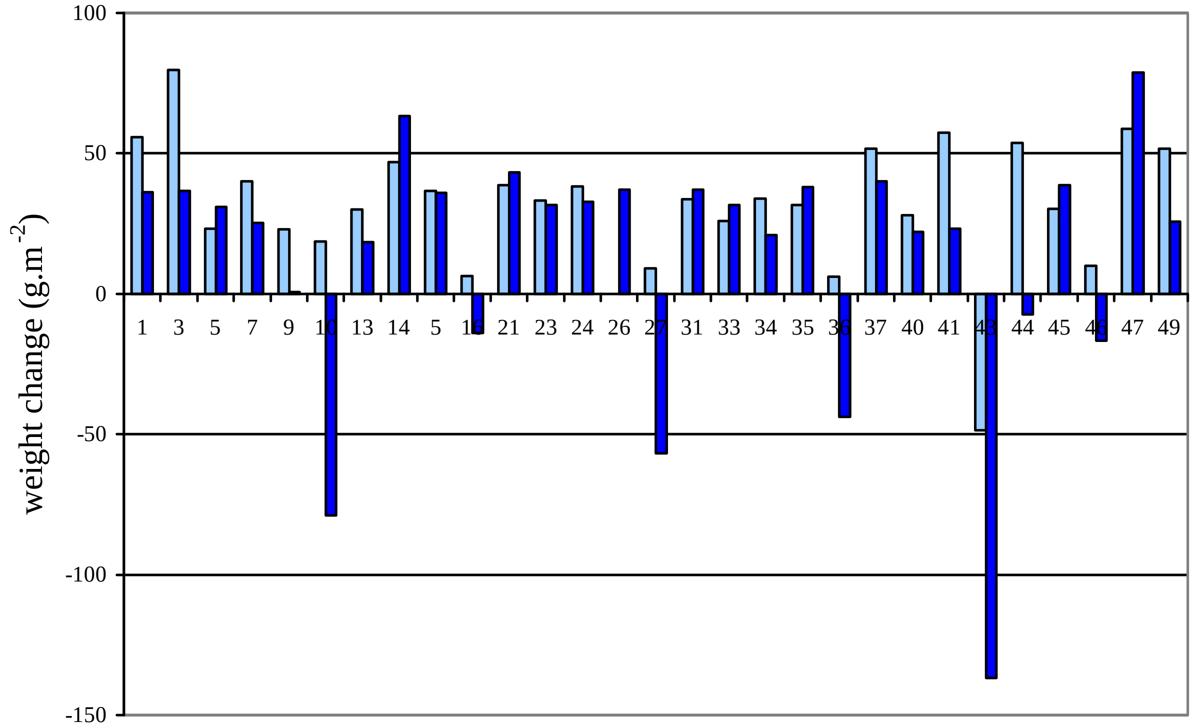
Table 5 – Weight increase ($g.m^{-2}$) of carbon steel after 2 years´ exposure – period 1997/99

$\Delta m = m_2 - m_1$

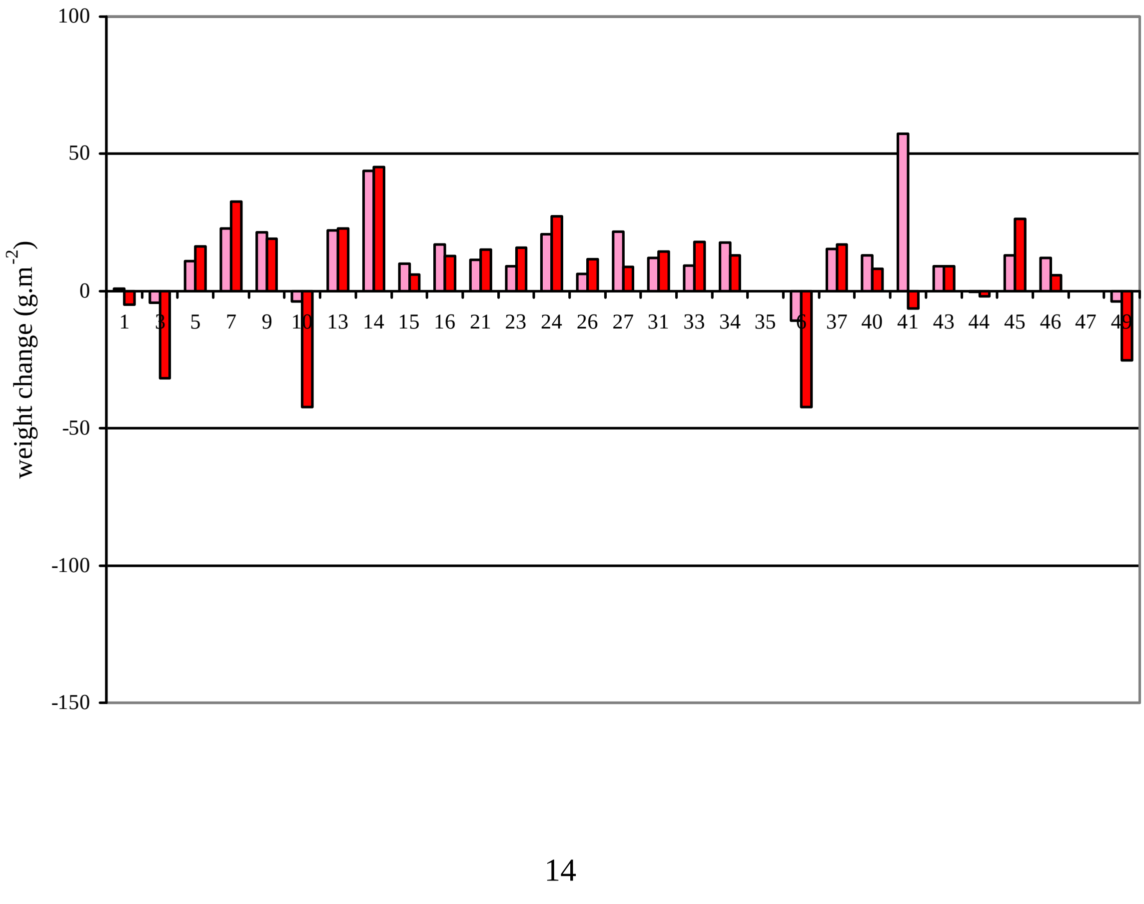
Test site	open exposure		shelter exposure	
	m_{un2}	Δm	m_{sh2}	Δm
1 Prague	36,3	- 19,4	-5,0	- 6,0
3 Kopisty	36,7	- 43,0	-31,7	- 27,0
5 Ahtari	31,0	+ 7,7	16,3	+ 5,3
7 Waldhof-Langenbrugge	25,3	- 14,7	32,7	+ 10,0
9 Langenfeld	0,7	- 22,3	19,0	- 2,3
10 Bottrop	-79,0	- 97,7	-42,3	- 38,6
13 Rome	18,3	- 11,7	22,7	+ 0,7
14 Cassacia	63,3	+ 16,3	45,3	+ 1,6
15 Milan	36,0	- 0,7	6,0	- 4,0
16 Venice	-14,0	- 20,3	12,7	- 4,3
21 Oslo	43,3	+ 4,6	15,0	+ 3,7
23 Birkenes	31,7	- 1,6	15,7	+ 6,7
24 Stockholm South	32,7	- 5,6	27,3	+ 6,6
26 Aspvreten	37,0	~	11,7	+ 5,4
27 Lincoln Cathedral	-56,7	- 65,7	8,7	- 13,0
31 Madrid	37,0	+ 3,3	14,3	+ 2,3
33 Toledo	31,7	+ 5,7	18,0	+ 8,7
34 Moskow	21,0	- 13,0	13,0	- 4,7
35 Lahemaa	38,0	+ 6,3	not evaluated	~
36 Lisbon	-43,7	- 49,7	-42,3	- 31,6
37 Dorset	40,0	- 11,7	17,0	+ 1,7
40 Paris	22,0	- 6,0	8,0	- 5,0
41 Berlin	23,3	- 34,0	-6,3	- 63,6
43 Tel Aviv	-136,7	- 88,0	9,0	0,0
44 Svanvik	-7,3	- 61,0	-2,0	- 1,7
45 Chaumont	38,7	+ 8,4	26,3	+ 13,3
46 London	-16,7	- 26,7	5,7	- 6,7
47 Los Angeles	78,7	+ 20,0	not exposed	~
49 Anvterps	25,7	- 26,0	-25,3	- 21,6

Figure 3 – Changes in weight of samples of carbon steel after 1 and 2 years of eposure

open exposure



shelter exposure



2.4. Corrosion losses of carbon steel

Corrosion losses for individual samples of unalloyed steel exposed in open atmosphere and under shelter evaluated for 1 and 2 years exposure (1997/98 and 1997/99) are summarized in Tables 6 - 9 and presented in Figure 4. There are not values of corrosion loss from shelter exposure for test sites No 35 (saples were in contact during exposure) and No 49 (samples were not exposed in this period).

After 1 year of exposure test sites with the greatest corrosion losses of unsheltered samples were 43 Tel Aviv and 10 Bottrop (over 300 $g.m^{-2}$). The same results were found after 2 years of exposure.

After 1 year of exposure test sites with the greatest corrosion losses of sheltered samples were 36 Lisbon and 27 Lincoln Cathedral (about 100 $g.m^{-2}$). The same results were found after 2 years of exposure. The results from test site 14 Cassacia after 1 year of exposure are not reliable, they are similar to corrosion loss from unsheltered exposure. Corrosion loss from test site 43 Tel Aviv from sheltered exposure is significantly lower than from unsheltered exposure. Corrosion loss from test site 10 Bottrop from sheltered exposure is relatively high.

2.5. Analysis of rust layer

For selected test sites the analysis of amount of Cl^- in rust layer from samples after 1 year of exposure was performed - the results are presented in Table 10. In the time of analyses environmental data were not available so test sites where the influence of salinity have been expected were choosen.

Table 10 – Amount of Cl^- in rust layer for selected test sites

test site	side	concentration of Cl^- (mg Cl^-/g rust)
16	upper	6,24
	ground	7,30
44	upper	7,16
	ground	3,11
47	upper	10,07
	ground	9,07

The amount of chlorides in these samples is not very high, but these test sites are not ones with the important influence of salinity – see 2.2.

Table 6 - Corrosion loss of carbon steel (g.m^{-2}) after one year´s exposure in open atmosphere

test site	exposure period		corrosion loss (g.m^{-2})			
			51	52	53	average
1 Prague	97-11-17	98-10-30	181,37	182,43	182,29	182,03
3 Kopisty	97-11-20	98-10-23	241,96	234,93	241,42	239,43
5 Ahtari	97-11-13	98-11-20	52,23	52,32	54,89	53,15
7 Waldhof-Langenbrugge	97-11-18		139,07	146,54	146,29	143,97
9 Langenfeld	97-11-17		204,91	203,00	204,93	204,28
10 Bottrop	97-11-19		311,76	307,97	314,55	311,43
13 Rome			131,01	130,77	139,32	133,70
14 Cassacia			124,81	126,56	123,43	124,93
15 Milan			175,11	171,14	173,07	173,11
16 Venice			205,18	214,55	212,87	210,87
21 Oslo	97-11-10	98-11-11	91,84	95,75	90,36	92,65
23 Birkenes	97-11-18	98-11-19	94,99	101,09	107,62	101,23
24 Stockholm South			122,12	126,95	124,79	124,62
26 Aspvreten			58,96	63,07	63,97	62,00
27 Lincoln Cathedral			265,99	270,54	272,40	269,65
31 Madrid	98-02-09	99-02-19	69,75	74,14	72,30	72,06
33 Toledo			51,89	56,22	54,65	54,25
34 Moskow			135,40	135,01	135,05	135,15
35 Lahemaa			104,74	108,82	105,27	106,27
36 Lisbon			216,92	212,52	212,64	214,03
37 Dorset	97-11-02	98-11-17	115,47	115,98	116,57	116,00
40 Paris	97-11-16	98-11-24	136,90	137,20	136,65	136,92
41 Berlin	97-11-19		174,51	180,41	181,15	178,68
43 Tel Aviv	97-11-10	98-11-11	322,24	321,11	327,19	323,51
44 Svanvik	97-11-12	98-11-05	166,69	163,22	166,88	165,60
45 Chaumont	97-10-23		65,91	68,20	65,92	66,68
46 London			180,32	173,97	175,92	176,73
47 Los Angeles			135,93	136,80	missing	136,36
49 Anvterps			176,60	170,39	165,53	170,84

Table 7 - Corrosion loss of carbon steel (g.m^{-2}) after two years´ exposure in open atmosphere

test site		exposure period		corrosion loss (g.m^{-2})			
				54	55	56	average
1	Prague	97-11-17	99-11-15	258,45	232,33	235,70	235,50
3	Kopisty	97-11-20	99-11-10	351,77	324,24	322,60	332,87
5	Ahtari	97-11-13	99-11-22	81,03	82,46	83,10	82,20
7	Waldhof-Langenbrugge	97-11-18	99-11-09	189,34	193,12	197,60	193,35
9	Langenfeld	97-11-17	99-11-08	231,30	236,31	233,68	233,76
10	Bottrop	97-11-19	99-11-09	442,64	460,78	481,33	461,59
13	Rome			missing	151,76	159,82	155,79
14	Cassacia			151,76	149,25	145,72	147,48
15	Milan			185,24	187,37	186,14	186,76
16	Venice			258,24	259,49	259,10	259,30
21	Oslo	97-11-10	99-11-08	135,58	137,45	137,67	136,90
23	Birkenes	97-11-18	99-11-09	155,46	169,25	167,62	164,11
24	Stockholm South			170,91	171,53	177,29	173,24
26	Aspvreten			95,87	102,27	102,60	100,25
27	Lincoln Cathedral			387,10	387,33	376,00	383,47
31	Madrid	98-02-09	00-02-08	126,23	106,64	111,46	114,78
33	Toledo			84,90	82,41	84,80	84,04
34	Moskow			157,11	158,36	156,14	157,20
35	Lahemaa		99-10-26	158,24	154,96	154,69	155,96
36	Lisbon			304,90	334,25	332,62	323,92
37	Dorset	97-11-02	99-11-10	164,47	165,73	167,48	165,89
40	Paris	97-11-16	99-11-22	155,91	158,79	163,15	159,28
41	Berlin	97-11-19	99-11-10	237,43	232,80	235,68	235,31
43	Tel Aviv	97-11-10	99-12-03	446,18	455,42	437,23	446,28
44	Svanvik	97-11-12	99-11-11	216,88	219,73	245,31	227,31
45	Chaumont	97-10-23	99-11-04	108,46	111,20	112,10	110,59
46	London			227,51	235,06	242,02	234,87
47	Los Angeles			186,14	187,58	175,23	182,98
49	Anvterps			230,37	234,28	236,99	233,88

Table 8 - Corrosion loss of carbon steel (g.m^{-2}) after one year´s exposure under shelter

test site	exposure period		corrosion loss (g.m^{-2})			
			61	62	63	average
1 Prague	97-11-17	98-10-30	52,79	51,42	52,00	52,52
3 Kopisty	97-11-20	98-10-23	85,98	82,15	78,97	82,37
5 Ahtari	97-11-13	98-11-20	20,03	19,41	19,12	19,52
7 Waldhof-Langenbrugge	97-11-18		44,75	45,22	44,85	44,94
9 Langenfeld	97-11-17		50,91	50,86	50,47	50,75
10 Bottrop	97-11-19		68,82	70,44	75,41	71,56
13 Rome			49,70	45,18	49,08	47,99
14 Cassacia			99,44	101,68	101,23	100,78
15 Milan			51,61	40,72	49,05	47,13
16 Venice			59,47	60,39	57,67	59,11
21 Oslo	97-11-10	98-11-11	24,42	20,76	20,71	21,97
23 Birkenes	97-11-18	98-11-19	19,84	17,42	19,06	18,77
24 Stockholm South			41,46	42,68	41,39	41,84
26 Aspvreten			13,78	12,29	13,83	13,30
27 Lincoln Cathedral			94,42	90,19	89,49	91,37
31 Madrid		99-02-19	25,63	23,30	21,27	23,40
33 Toledo			14,72	15,61	16,01	15,45
34 Moskow			38,40	37,41	37,78	37,86
35 Lahemaa			not evaluated			
36 Lisbon			105,96	100,76	102,79	103,17
37 Dorset	97-11-02	98-11-17	54,64	51,37	50,43	52,15
40 Paris	97-11-16	98-11-24	48,01	49,43	46,95	48,13
41 Berlin	97-11-19		56,08	61,03	60,06	59,05
43 Tel Aviv	97-11-10	98-11-11	56,24	44,29	43,14	47,89
44 Svanvik	97-11-12	98-11-05	34,99	29,15	45,35	36,50
45 Chaumont			23,58	24,36	25,58	24,51
46 London			77,06	73,34	70,99	73,80
47 Los Angeles			not exposed			
49 Anvterps			71,08	69,53	67,75	69,46

marked values are not reliable

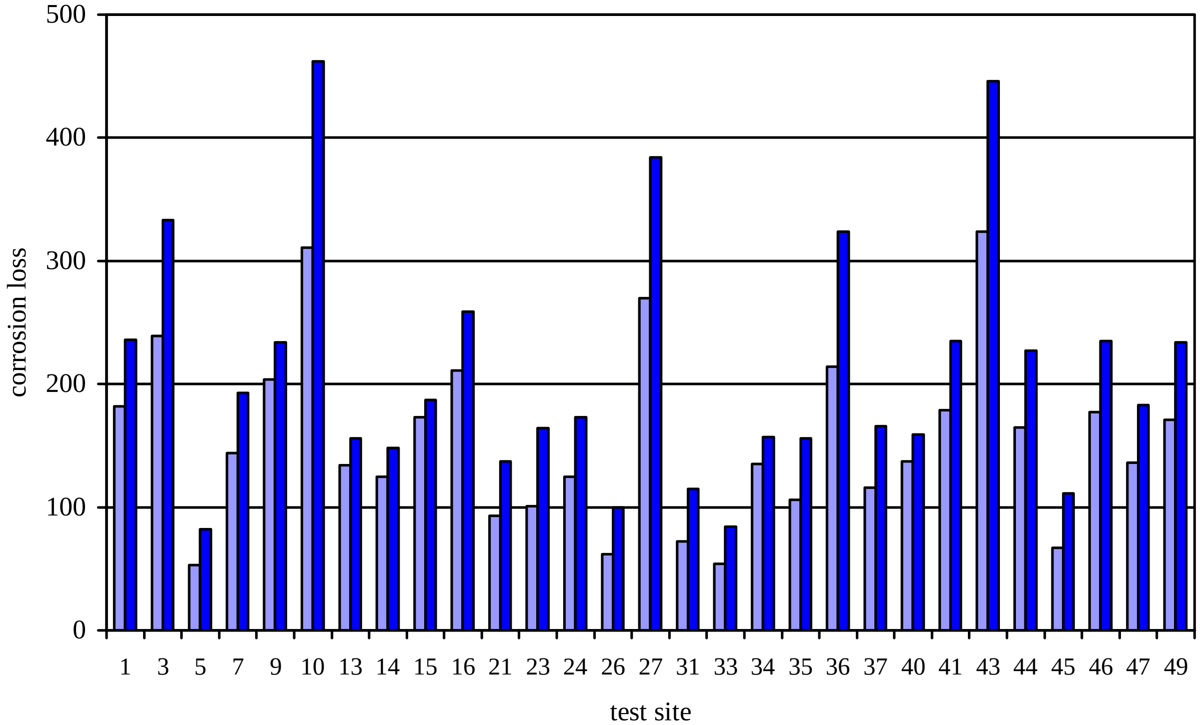
Table 9 - Corrosion loss of carbon steel (g.m^{-2}) after two years´ exposure under shelter

test site	exposure period		corrosion loss (g.m^{-2})			
			64	65	66	average
1 Prague	97-11-17	99-11-15	90,61	88,83	89,56	89,67
3 Kopisty	97-11-20	99-11-10	138,20	130,56	133,54	134,10
5 Ahtari	97-11-13	99-11-22	32,50	32,85	30,35	31,90
7 Waldhof-Langenbrugge	97-11-18	99-11-09	71,86	70,79	70,56	71,07
9 Langenfeld	97-11-17	99-11-08	91,17	91,87	89,48	90,84
10 Bottrop	97-11-19	99-11-09	144,04	147,89	151,45	147,79
13 Rome			70,08	67,08	66,85	68,00
14 Cassacia			114,27	104,65	97,61	105,51
15 Milan			72,76	60,97	62,26	65,33
16 Venice			81,90	82,11	85,14	83,05
21 Oslo	97-11-10	99-11-08	44,41	40,05	40,47	41,64
23 Birkenes	97-11-18	99-11-09	34,58	36,85	36,50	35,98
24 Stockholm South			64,93	63,75	62,65	63,78
26 Aspvreten			26,27	28,25	26,78	27,10
27 Lincoln Cathedral			157,03	153,05	171,71	160,60
31 Madrid		00-02-08	50,32	47,51	51,89	49,91
33 Toledo			33,00	33,59	32,85	33,14
34 Moskow			66,49	62,95	62,70	64,05
35 Lahemaa		99-10-26	not evaluated			
36 Lisbon			175,55	176,05	176,15	175,92
37 Dorset	97-11-02	99-11-10	78,55	78,66	76,30	77,83
40 Paris	97-11-16	99-11-22	70,99	68,42	68,27	69,23
41 Berlin	97-11-19	99-11-10	105,94	94,73	85,37	95,35
43 Tel Aviv	97-11-10	99-12-03	53,87	50,80	51,72	52,13
44 Svanvik	97-11-12	99-11-11	85,44	68,21	66,65	73,43
45 Chaumont		99-11-04	55,58	54,75	53,73	54,68
46 London			116,35	122,57	120,97	119,97
47 Los Angeles			not exposed			
49 Anvterps			105,17	102,54	102,95	103,55

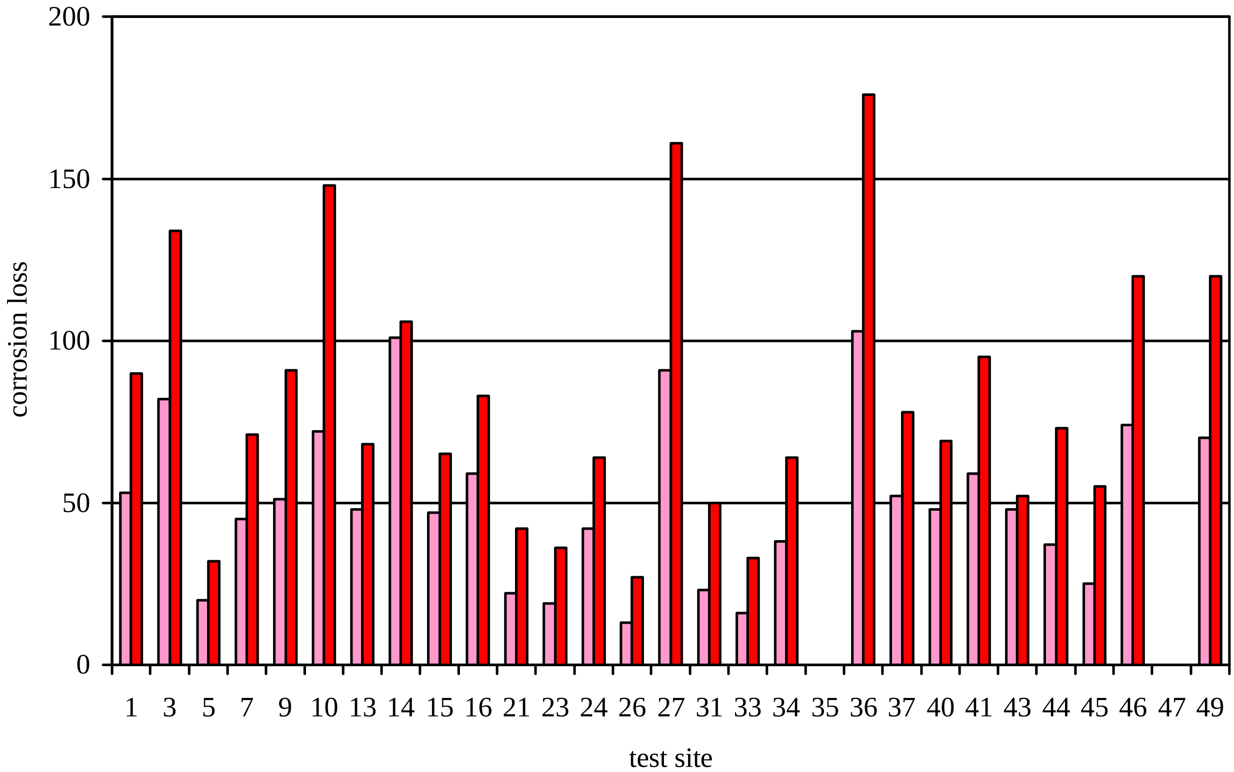
marked values are not reliable

Figure 4 – Corrosion loss of carbon steel ($g.m^{-2}$) after 1 and 2 years of exposure

open exposure



shelter exposure



3. Data treatment and evaluation of results

3.1. Comparison of corrosion behaviour of carbon steel

Samples of unalloyed carbon steel were exposed in the original programme only as standard specimen for determination of corrosivity of test sites (according to ISO 9226). During the programme the exposures of these samples were repeated to validate decreasing trend of corrosivity due to decreasing of SO_2 pollution. Results were published in Report No. 19.

Comparison of corrosion losses for different exposure periods are given in Tables 11 and 12 and Figures 5 and 6.

Range of corrosion losses evaluated after one year of exposure within the multipollutant programme is in general lower as within the original programme caused by the lower corrosion impact on the industrial test sites.

open exposure		
1987/88	557 $g.m^{-2}$ (Kopisty)	45 $g.m^{-2}$ (Toledo)
1997/98	311 $g.m^{-2}$ (Bottrop)	54 $g.m^{-2}$ (Toledo)

shelter exposure		
1987/88	254 $g.m^{-2}$ (Kopisty)	13 $g.m^{-2}$ (Toledo)
1997/98	103 $g.m^{-2}$ (Lisabon)	13 $g.m^{-2}$ (Aspvreten)

Figure 7 shows comparison of the relative effects of corrosion of unalloyed carbon steel in sheltered and unsheltered conditions in different exposure periods. The most significantly decreasing has been found for corrosion loss caused in unsheltered conditions, significantly decreasing has been also found for corrosion under shelter on test sites in past high polluted by SO_2 – 1 Prague and 3 Kopisty. It is interesting remaining high contribution of unsheltered corrosion loss on test site 10 Bottrop and increasing contribution of unsheltered corrosion on test site 27 Lincoln Cathedral. Only test site where corrosion loss in shelter exposure increased in period 1997/98 was 36 Lisbon (test site moved on the othere side of building).

Comparison of the individual corrosion losses and weight increases can be interpereted as a general characteristic for the adhesion, chemical stability and consistency of the rust layers – Figures 8 and 9. Principal differences can be expected between the rust layers from the polluted or wet test sites Kopisty, Bottrop and Lincoln Cathedral:

test site	mass loss ($g.m^{-2}$)	mass increase ($g.m^{-2}$)
03 Kopisty	239	79,7
10 Bottrop	311	18,7
27 Lincoln Cathedral	269	9,0

Table 11 - Comparison of corrosion losses (g/m^2) of unalloyed steel after one-year´s exposure in open atmosphere in different exposure periods

Test site	period 1987-88	period 1992-93	period 1994-95	period 1996-97	period 1997-98
1 Prague	438 C4	271 C3	241 C3	232 C3	182 C2
3 Kopisty	557 C4	350 C3	352 C3	293 C3	239 C3
5 Ahtari	132 C2	48 C2	59 C2	54 C2	53 C2
7 Waldhof-Langenbrugge	264 C3	231 C3	166 C2	156 C2	144 C2
9 Langenfeld	293 C3	231 C3	210 C3	206 C3	204 C3
10 Bottrop	373 C3	347 C3	294 C3	296 C3	311 C3
13 Rome	178 C2	-	124 C2	109 C2	134 C2
14 Casaccia	235 C3	-	148 C2	135 C2	125 C2
15 Milan	366 C3	-	198 C2	149 C2	173 C2
16 Venice	245 C3	-	212 C3	188 C2	211 C3
21 Oslo	229 C3	135 C2	101 C2	99 C2	93 C2
23 Birkenes	194 C2	132 C2	109 C2	114 C2	101 C2
24 Stockholm South	264 C3	120 C2	103 C2	104 C2	125 C2
26 Aspvreten	147 C2	75 C2	81 C2	69 C2	62 C2
27 Lincoln Cathedral	315 C3	308 C3	237 C3	-	270 C3
31 Madrid	222 C3	162 C2	151 C2	159 C2	72 C2
33 Toledo	45 C2	26 C2	36 C2	36 C2	54 C2
34 Moskow	181 C2	141 C2	121 C2	123 C2	135 C2
35 Lahemaa	185 C2	-	-	-	106 C2
36 Lisbon	224 C3	308 C3	204 C3	289 C3	214 C3
37 Dorset	149 C2	110 C2	104 C2	120 C2	116 C2
40 Paris	-	-	-	-	137 C2
41 Berlin	-	-	-	174 C2	179 C2
43 Tel Aviv	-	-	-	-	324 C3
44 Svanvik	-	-	-	160 C2	166 C2
45 Chaumont	-	-	-	94 C2	67 C2
46 London	-	-	-	-	177 C2
47 Los Angeles	-	-	-	-	136 C2
49 Antverps	-	-	-	-	171 C2

C2, C3 , C4 ..... corrosivity categories according ISO 9223

Table 12 - Comparison of corrosion losses (g/m^2) of unalloyed steel after one-year´s exposure under shelter in different exposute periods

Test site	period 1987-88	period 1992-93	period 1994-95	period 1997-98
1 Prague	195 C2	15 C2	81 C2	53 C2
3 Kopisty	254 C3	203 C3	206 C3	82 C2
5 Ahtari	37 C2	12 C2	16 C2	20 C2
7 Waldhof-Langenbrugge	106 C2	95 C2	53 C2	45 C2
9 Langenfeld	83 C2	44 C2	54 C2	51 C2
10 Bottrop	86 C2	81 C2	98 C2	72 C2
13 Rome	49 C2	-	36 C2	48 C2
14 Casaccia	98 C2	-	51 C2	101 C2
15 Milan	101 C2	-	52 C2	47 C2
16 Venice	71 C2	-	22 C2	59 C2
21 Oslo	46 C2	21 C2	28 C2	22 C2
23 Birkenes	27 C2	19 C2	26 C2	19 C2
24 Stockholm South	93 C2	35 C2	36 C2	42 C2
26 Aspvreten	43 C2	23 C2	29 C2	13 C2
27 Lincoln Cathedral	155 C2	83 C2	61 C2	91 C2
31 Madrid	52 C2	34 C2	30 C2	23 C2
33 Toledo	13 C2	17 C2	13 C2	16 C2
34 Moskow	48 C2	37 C2	27 C2	38 C2
35 Lahemaa	89 C2	-	-	-
36 Lisbon	69 C2	89 C2	81 C2	103 C2
37 Dorset	47 C2	24 C2	17 C2	52 C2
40 Paris	-	-	-	48 C2
41 Berlin	-	-	-	59 C2
43 Tel Aviv	-	-	-	48 C2
44 Svanvik	-	-	-	37 C2
45 Chaumont	-	-	-	25 C2
46 London	-	-	-	74 C2
47 Los Angeles	-	-	-	-
49 Anvterps	-	-	-	69 C2

marked values are not reliable

C2, C3 .... corrosivity categories according ISO 9223

Figure 5 – Comparison of corrosion losses (g.m^{-2}) of unalloyed carbon steel in different exposure periods - open exposure

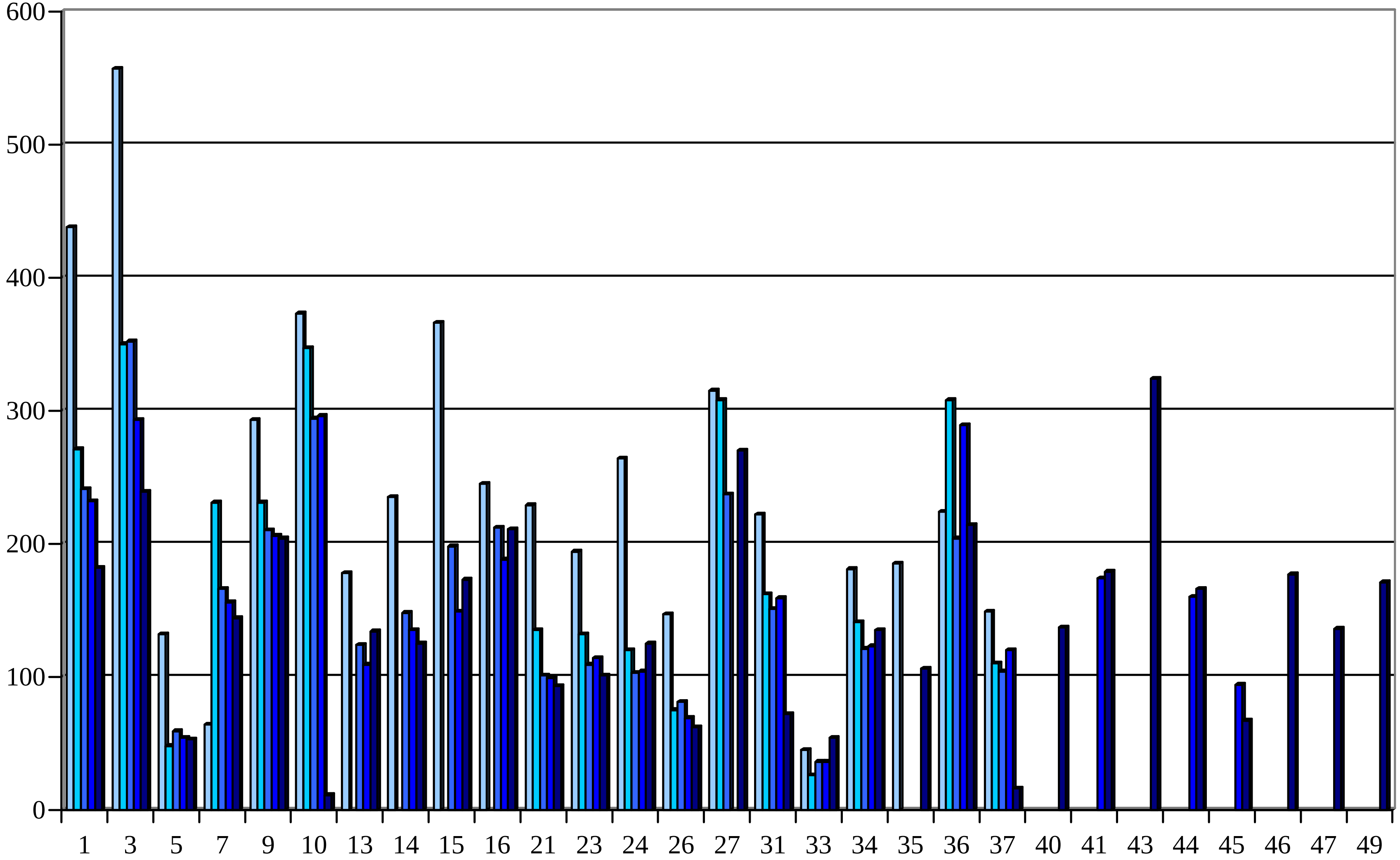


Figure 6 – Comparison of corrosion losses (g.m^{-2}) of unalloyed carbon steel in different exposure periods - shelter exposure

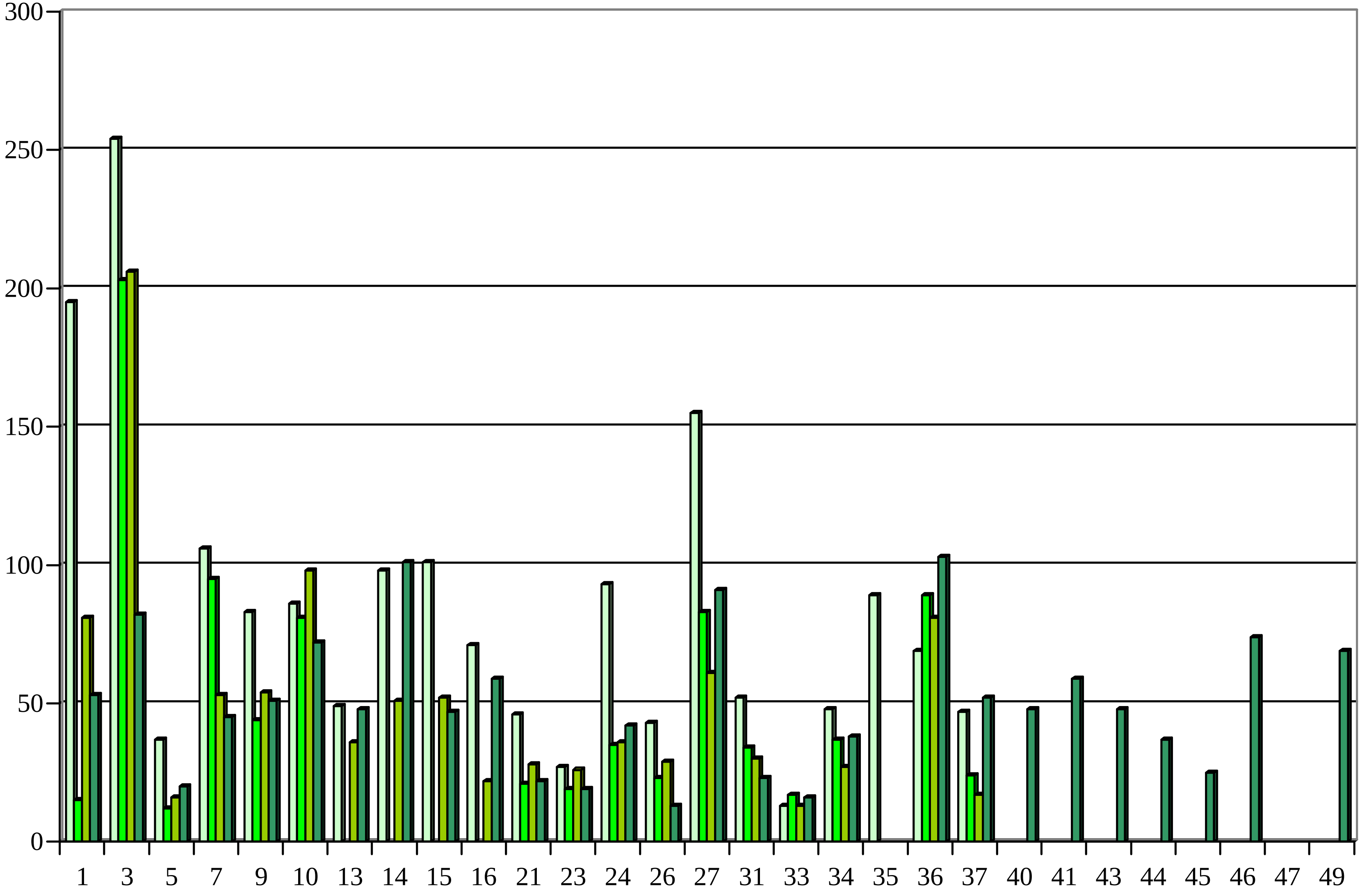


Figure 7 – Comparison of corrosion losses in different exposure conditions (unsheltered x sheltered) in period 1986/87 and 1997/98

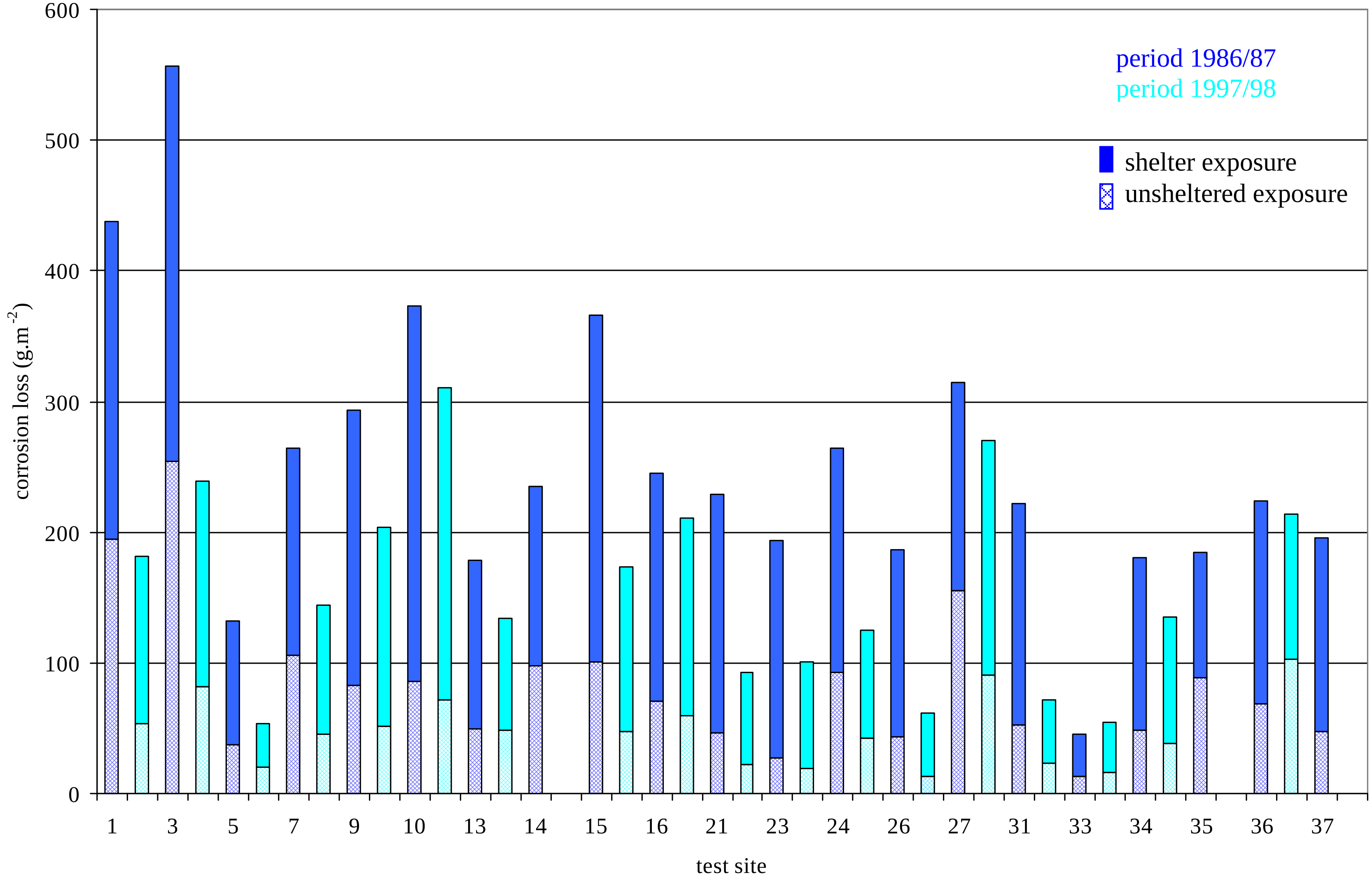
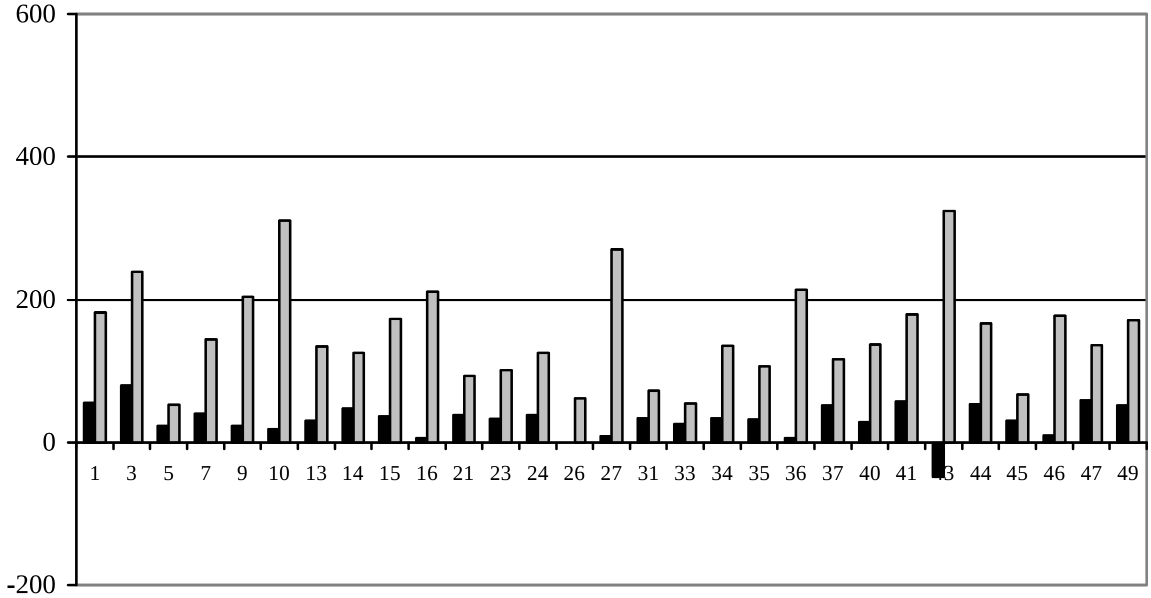


Figure 8 - Comparison of weight increases and corrosion losses (g.m^{-2}) of samples exposed in open atmosphere

1 year exposure



2 years of exposure

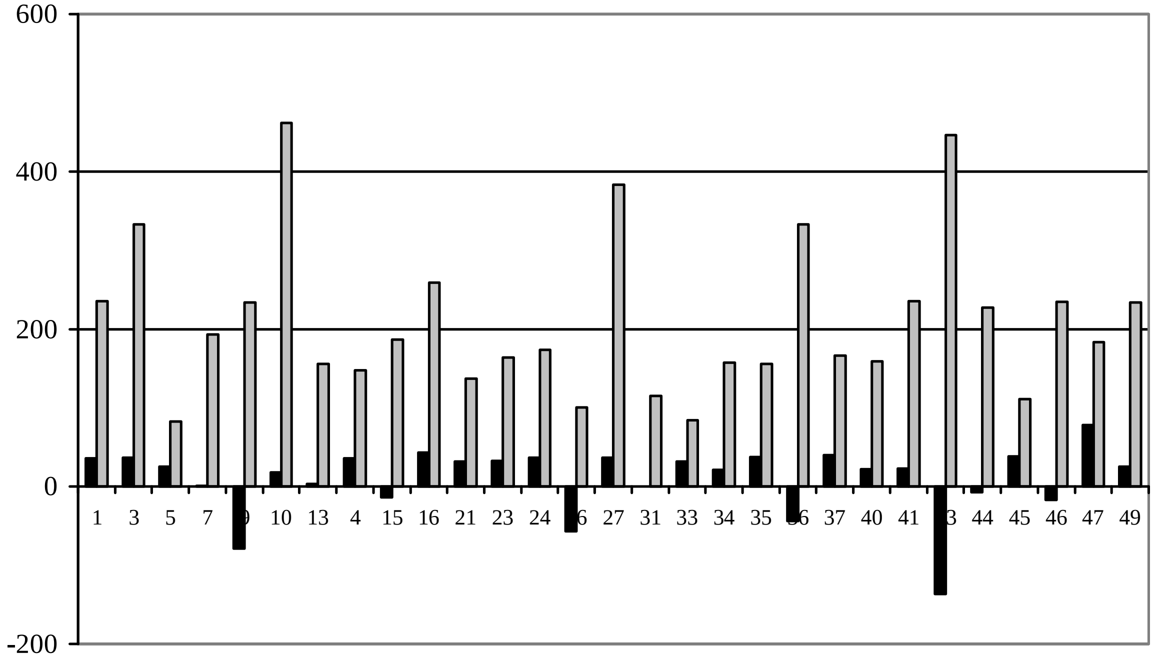
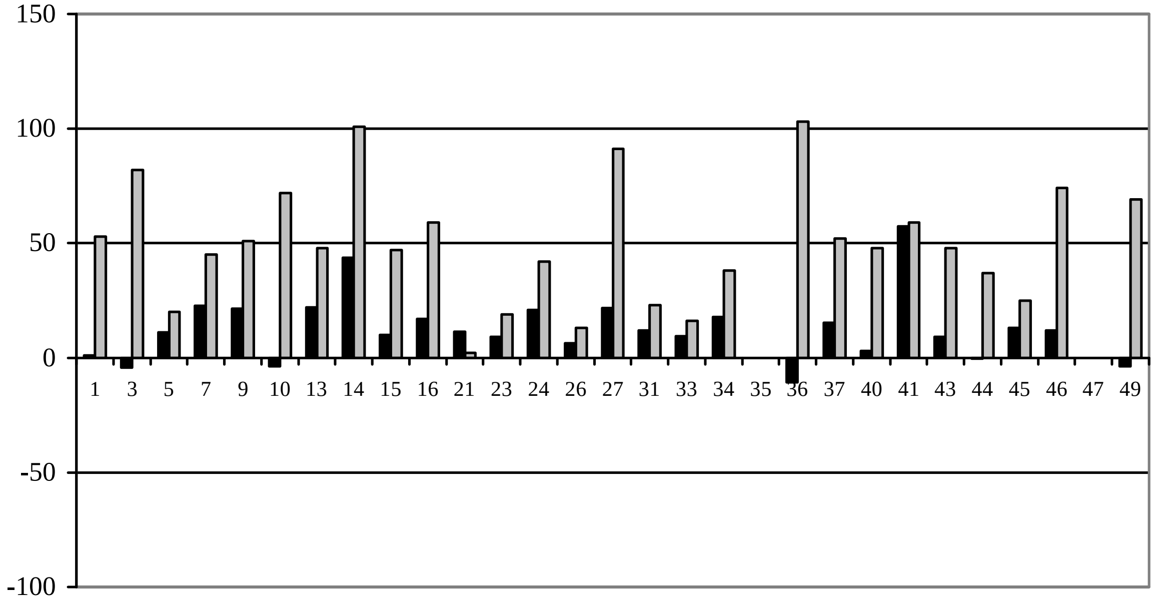
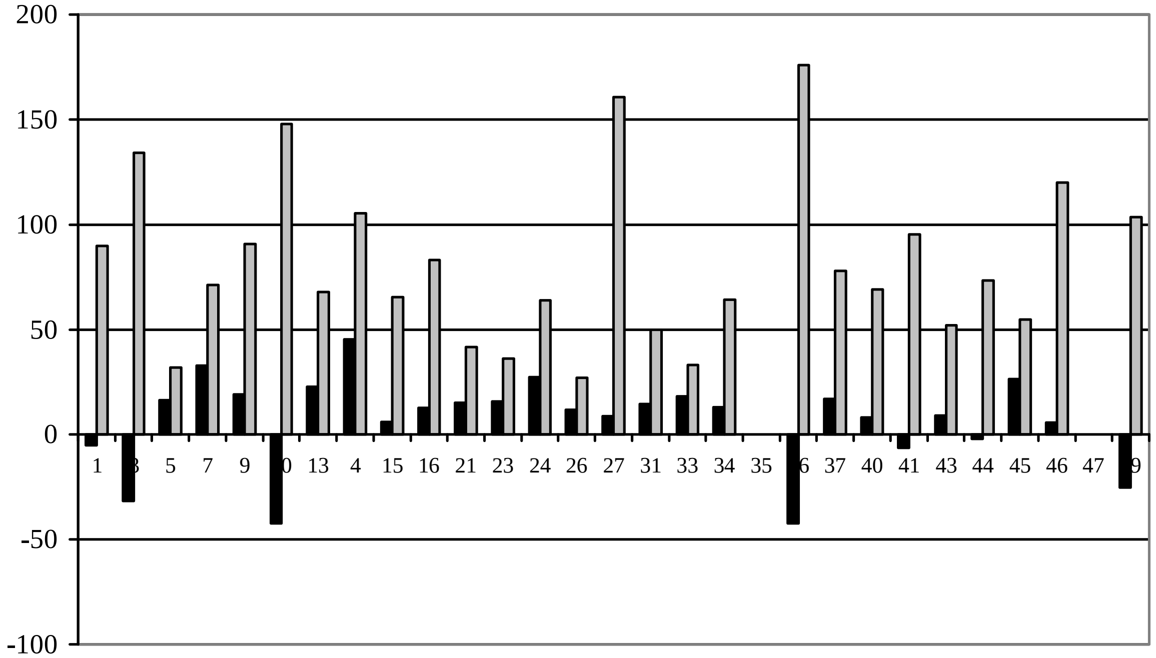


Figure 9 - Comparison of weight increases and corrosion losses (g.m^{-2}) of samples exposed under shelter

1 year exposure



2 years of exposure



There was performed comparison of decreasing of corrosion losses and the main environmental parameters affecting corrosion of carbon steel – Table 13. Figure 10 presents comparison of corrosion losses of carbon steel versus SO_2 pollution for years 1987/88 and 1997/98. The decreasing of concentration of SO_2 and its corrosion effect is evident.

Table 13 – Decreasing of corrosion losses and selected environmental parameters (%) in periods 1986/87 and 1997/98

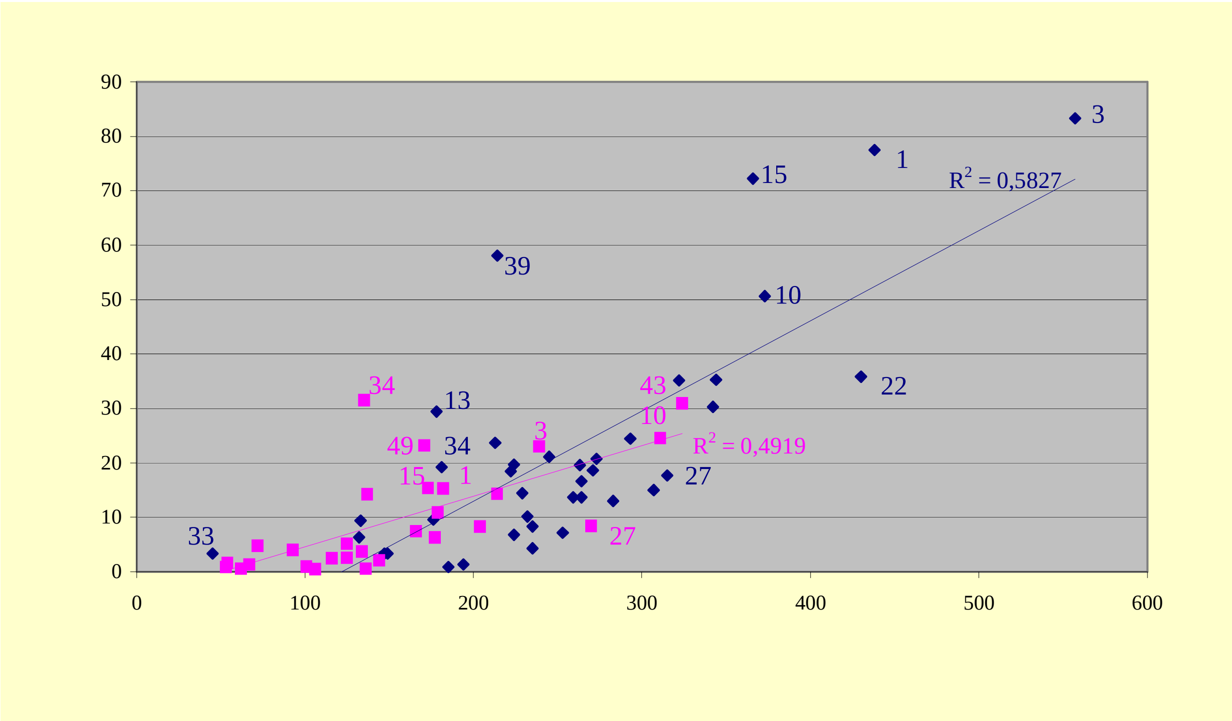
$$\Delta K = 100 - 100\frac{K_{98}}{K_{88}}$$

Test site	corrosion loss		environmental parameters	
	unsheltered	sheltered	SO_2	pH
1 Prague	58	73	80	28
3 Kopisty	57	68	72	7
5 Ahtari	60	46	86	4
7 Waldhof-Langenbrugge	45	58	85	15
9 Langenfeld	30	39	66	-
10 Bottrop	17	16	51	10
13 Rome	25	2	87	-
14 Casaccia	47	-	37	-
15 Milan	53	53	79	-
16 Venice	14	17	-	-
21 Oslo	59	52	72	21
23 Birkenes	48	30	85	6
24 Stockholm South	53	55	84	6
26 Aspvreten	58	70	82	7
27 Lincoln Cathedral	14	41	53	1
31 Madrid	68	56	*74*	10
33 Toledo	-	-	*52*	11
34 Moskow	25	21	-	8
35 Lahemaa	43	-	*44*	10
36 Lisbon	4	-	-	6
37 Dorset	22	-	*24*	1
average	40	46	67	9

Table 14 presents the corrosion losses on test sites after 1 year of exposure arranged in order to mass loss value for open exposure together with relevant environmental characteristics.

Figure 10 –Comparison of corrosion loss of carbon steel versus pollution of SO_2 in period 1987/88 and 1997/98

unsheltered exposure



shelter exposure

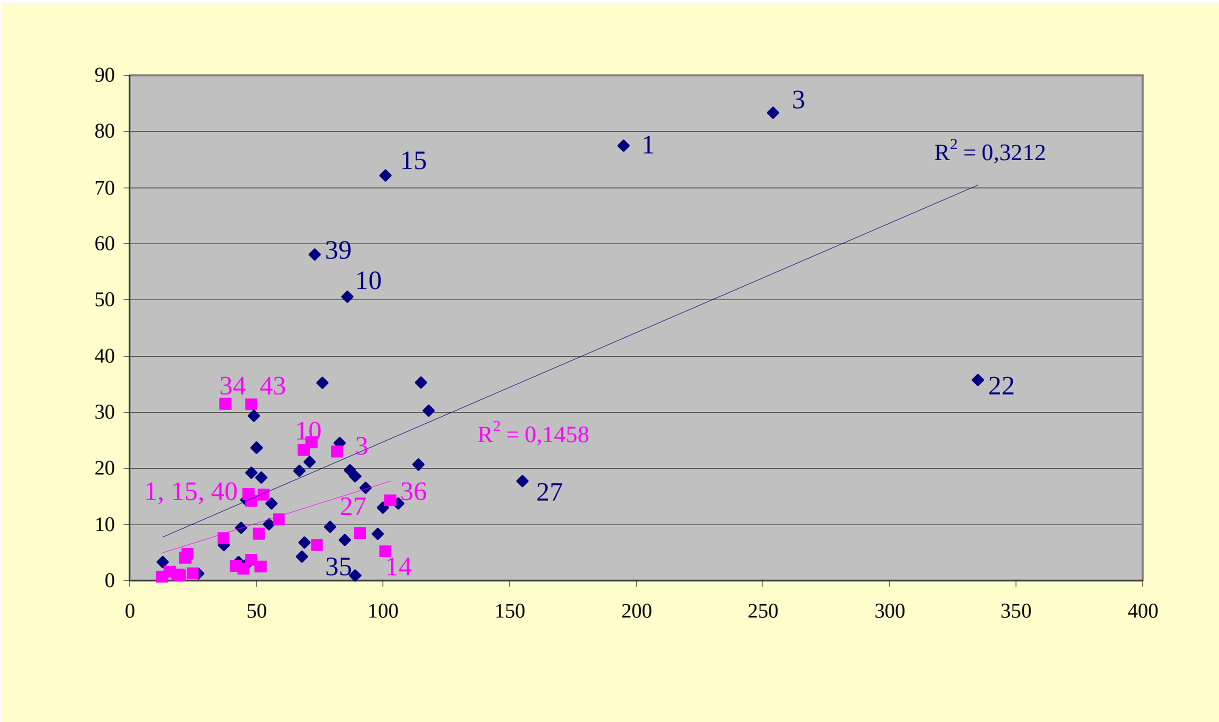


Table 14 – Comparison of corrosion losses after 1 year exposure and important pollution characteristics

test site	corrosion loss ($g.m^{-2}$)		gaseous pollution ($\mu g.m^{-3}$)			Conductivity of precipitation	Cl^- (mg/l)
	open	shelter	SO_2	NO_2	O_3		
43 Tel Aviv	324	48	31,4	35,5	37		
10 Bottrop	311	72	24,6	38,2	30	28,2	1,6
27 Lincoln Cathedral	270	91	8,4	19,1	51		103,1
3 Kopisty	239	82	23,0	32,6	55	44,6	1,7
36 Lisbon	214	103	14,3	47,8	7	*76,3*	*11,5*
16 Venice	211	59					
9 Langenfeld	204	51	8,3	33,5	33		
1 Prague	182	53	15,3	23,7	47	27,6	1,9
41 Berlin	179	59	10,9	37,7	47		2,3
46 London	177	74	6,3	45,3	36		*143,8*
15 Milan	173	47	15,4	83,9	38		
49 Anvterps	171	69	23,2	52,8	29	37,5	3,5
44 Svanvik	166	37	7,5	0,9	54	20,7	1,9
7 Waldhof-Langenbrugge	144	45	2,1	8,7	48	17,6	0,7
40 Paris	137	48	14,2	70,0	31	43,7	2,5
47 Los Angeles	136		0,6	21,7	48		
34 Moscow	135	38	31,5	28,0	42	51,1	1,7
13 Rome	134	48	3,7	37,8	33		
14 Cassacia	125	101	5,2	21,0	30		
24 Stockholm South	125	42	2,6	20,3	44	17,1	0,5
37 Dorset	116	52	2,5	*9,4*	63	26,3	0,1
35 Lahemaa	106		0,5	0,7	58	19,4	0,8
23 Birkenes	101	19	0,2	1,1	55	25,5	1,5
21 Oslo	93	22	4,0	27,5	35	23,0	1,0
31 Madrid	72	23	4,8	26,9		*12,5*	*1,1*
45 Chaumont	67	25	1,3	7,7	86	9,4	0,2
26 Aspvreten	62	13	0,6	2,9	51	17,3	0,6
33 Toledo	54	15	1,6	12,1	89	14,8	0,9
5 Ahtari	53	20	0,9	3,0	60	10,7	0,7

marked values are not reliable.

3.2. Statistical analysis

The correlation coefficients are the measure for the expresion of the stochastic liason between variables. The correlation coefficients computed according to Pearson and Spearman are listed in Tables 15 and 16. Coefficients give degree of covariance between two variables.

Table 15 – Pearson´s correlation analysis of results

	unsheltered exposure				sheltered exposure			
	1 year		2 years		1 year		2 years	
	r-Pears.	p-value	r-Pears.	p-value	r-Pears.	p-value	r-Pears.	p-value
Temp	**0,48**	0,006	**0,41**	0,015	**0,46**	0,011	0,24	N.S.
RH	0,07	N.S.	0,28	N.S.	-0,26	N.S.	-0,16	N.S.
SO_2	**0,71**	0,000	**0,70**	0,000	**0,41**	0,018	**0,30**	0,065
NO_2	**0,42**	0,014	**0,30**	0,063	**0,38**	0,028	**0,29**	0,074
O_3	**-0,50**	0,004	**-0,39**	0,022	**-0,57**	0,001	**-0,50**	0,005
pH	0,03	N.S.	0,11	N.S.	0,17	N.S.	0,26	N.S.
Cl	**0,35**	0,055	**0,28**	0,100	**0,46**	0,018	**0,41**	0,033
cond	**0,59**	0,004	**0,52**	0,009	**0,77**	0,000	**0,76**	0,000

correlation is significant

The correlation matrix of corrosion losses of unalloyed carbon steel and environmental parameters was performed after the first year of original programme (1987/88). The selected environmental parameters were: concentration of SO_2, NO_x, TOW, amount of SO_4^{2-} and Cl^- in precipitation, pH and conductivity of precipitation.

The comparison of correlation coefficients in both programmes are below (Table 17). The depentance of corrosion loss on environmental parameters are similar except pH of precipitation and NO_x for sheltered exposure. The significance of SO_2 partial decreased.

Table 17 – Comparion of correlation coefficient for environmental parameters

Parameter	Correlation coefficients			
	period 1987/88		period 1997/98	
	unsheltered	sheltered	unsheltered	sheltered
SO_2	0,76	0,56	0,71	0,41
NO_x	0,42	0,15	0,42	0,38
Cl^-	0,43	-	0,35	-
pH	0,29	-	0,03	-
cond	0,69	-	0,59	-

Table 16 – Correlation analysis of results – Spearman´s rho

1 year of exposure									
		Environmental parameters							
		Temp	RH	SO_2	NO_2	O_3	pH	Cl	cond
Cor_{unsh}	r	0,48	0,07	0,71	0,42	-0,50	0,03	0,35	0,59
	p-value	0,01	0,37	0,00	0,01	0,00	0,44	0,06	0,00
Cor_{sh}	r	0,46	-0,26	0,41	0,38	-0,57	0,17	0,46	0,77
	p-value	0,01	0,12	0,02	0,03	0,00	0,24	0,02	0,00
Temp	r	1,00	-0,52	0,35	0,57	-0,48	0,52	0,29	0,60
	p-value		0,00	0,03	0,00	0,01	0,01	0,10	0,00
RH	r	-0,52	1,00	0,00	-0,45	0,22	-0,68	-0,63	-0,55
	p-value	0,00		0,50	0,01	0,15	0,00	0,00	0,01
SO_2	r	0,35	0,00	1,00	0,56	-0,46	0,38	-0,04	0,75
	p-value	0,03	0,50		0,00	0,01	0,04	0,43	0,00
NO_2	r	0,57	-0,45	0,56	1,00	-0,60	0,50	0,21	0,65
	p-value	0,00	0,01	0,00		0,00	0,01	0,18	0,00
O_3	r	-0,48	0,22	-0,46	-0,60	1,00	-0,36	-0,15	-0,70
	p-value	0,01	0,15	0,01	0,00		0,05	0,26	0,00
pH	r	0,52	-0,68	0,38	0,50	-0,36	1,00	0,17	0,52
	p-value	0,01	0,00	0,04	0,01	0,05		0,22	0,01
Cl	r	0,29	-0,63	-0,04	0,21	-0,15	0,17	1,00	0,77
	p-value	0,10	0,00	0,43	0,18	0,26	0,22		0,00
cond	r	0,60	-0,55	0,75	0,65	-0,70	0,52	0,77	1,00
	p-value	0,00	0,01	0,00	0,00	0,00	0,01	0,00	
2 years of exposure									
		Environmental parameters (average for 1997/99)							
		Temp	RH	SO_2	NO_2	O_3	pH	Cl	cond
Cor_{unsh}	r	0,41	0,28	0,70	0,30	-0,39	0,11	0,28	0,52
	p-value	0,01	0,08	0,00	0,06	0,02	0,31	0,10	0,01
Cor_{sh}	r	0,24	-0,16	0,30	0,29	-0,50	0,26	0,41	0,76
	p-value	0,12	0,23	0,06	0,07	0,01	0,13	0,03	0,00
Temp	r	1,00	-0,34	0,48	0,56	-0,45	0,61	0,09	0,55
	p-value		0,05	0,01	0,00	0,01	0,00	0,34	0,01
RH	r	-0,34	1,00	0,15	-0,30	0,19	-0,57	-0,47	-0,42
	p-value	0,05		0,23	0,07	0,18	0,00	0,02	0,04
SO_2	r	0,48	0,15	1,00	0,50	-0,43	0,35	0,00	0,58
	p-value	0,01	0,23		0,00	0,01	0,05	0,50	0,00
NO_2	r	0,56	-0,30	0,50	1,00	-0,67	0,56	0,27	0,52
	p-value	0,00	0,07	0,00		0,00	0,00	0,10	0,01
O_3	r	-0,45	0,19	-0,43	-0,67	1,00	-0,45	-0,16	-0,73
	p-value	0,01	0,18	0,01	0,00		0,02	0,23	0,00
pH	r	0,61	-0,57	0,35	0,56	-0,45	1,00	0,33	0,52
	p-value	0,00	0,00	0,05	0,00	0,02		0,06	0,01
Cl	r	0,09	-0,47	0,00	0,27	-0,16	0,33	1,00	0,91
	p-value	0,34	0,02	0,50	0,10	0,23	0,06		0,00
cond	r	0,55	-0,42	0,58	0,52	-0,73	0,52	0,91	1,00
	p-value	0,01	0,04	0,00	0,01	0,00	0,01	0,00	

Athough the data set is relatively small and the set of environmental data is not completed, the stepwise regression analysis for 4 sets show the similar results (Table 18). Corrosion loss of carbon steel is affected by concetration of SO_2, temperature, pH of precipitation and amount of chlorides in precipitation.

Table 18 – Regression analysis of results

Dependent Variable: St_{1uns}	R Square	0,723			
	B	Std. Error	Beta	t	p-value
(Constant)	317,256				
SO_2	5,641	1,161	0,783	4,857	0,000
pH	-51,951	17,909	-0,484	-2,901	0,011
Temp	4,436	2,847	0,260	1,558	0,140
Cl	0,763	0,276	0,405	2,769	0,014
Dependent Variable: St_{1sh}	R Square	0,768			
	B	Std. Error	Beta	t	p-value
(Constant)	82,077				
SO_2	1,600	0,414	0,589	3,865	0,002
pH	-15,421	6,315	-0,390	-2,442	0,028
Temp	3,168	1,001	0,495	3,164	0,007
Cl	0,301	0,096	0,432	3,120	0,008
Dependent Variable: St_{2uns}	R Square	0,660			
	B	Std. Error	Beta	t	p-value
(Constant)	489,981				
SO_2	8,390	1,859	0,733	4,513	0,000
pH	-86,324	28,697	-0,617	-3,008	0,008
Temp	9,962	4,079	0,444	2,443	0,027
Cl	1,061	0,378	0,447	2,804	0,013
Dependent Variable: St_{2sh}	**R Square**	**0,704**			
	B	Std. Error	Beta	t	p-value
(Constant)	146,888				
SO_2	3,344	0,902	0,641	3,708	0,002
pH	-27,452	12,741	-0,439	-2,155	0,049
Temp	4,901	1,936	0,452	2,531	0,024
Cl	0,555	0,168	0,534	3,297	0,005

4. Conclusions

Results document the remaining high positive correlation of SO_2 with corrosion losses, high positive correlation between corrosion losses and conductivity, positive influence of NO_2 cannot be omitted. Negative correlation for O_3 was evaluated in accordance with the results for the first stage of UN ICP Materials although this parameters were not measured on all test sites. The results confirm the using of temperature and RH as individual parameters not in combination expressed as TOW.