Traffic-induced salt deposition on facades

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Abstract

Air pollution has been reduced significantly in Germany during the last decades. Nevertheless high pollution levels are recorded in urban environment along heavy-trafficked roads. Relevant portions of the emissions of nitrogen oxides and fine particulate matter are caused by car traffic. The change of environmental conditions has led to a change of salt deposition on facades. Actual research results show high concentrations of nitrogen oxides at historical facades but contradictory data for the deposition of nitrates. Additionally, the redispersion of de-icing salt by car traffic leads to a deposition of chlorides on facades not only in the base zone.

Keywords: air pollution, traffic emissions, salt input

1. Introduction

In the last decades air pollution in Germany has been reduced significantly. Most notably the emission of sulphur dioxide and its secondary products are reduced now to a level of less than 10%, compared to the amount emitted in the late 1980ies. Consequently the pH value of rain increased to a level being inherent by the saturation with carbon dioxide. Also the emissions of nitrogen oxides and particulate matter could be reduced, but they still are at problematic levels, regarding the actual threshold values. High proportions of these pollutants originate from road traffic. Besides their negative health effects, their impact on materials has to be considered. This is particularly true for historic buildings, because they have been exposed to air pollutants for decades or even centuries and they also will be affected by future emissions.

The paper will report some results from recent research on the impact of the actual pollution regime on historic stone buildings.[1-3] Especially results regarding the input of salts and salt-forming substances are presented.

2. Methods

In the research project presented here, the impact of traffic-related immissions on the building materials of historic monuments in several German cities was studied. Data on traffic volume and air quality, numerical modelling of the dispersion of air pollutants in the surrounding of selected monuments, exposure experiments, as well as laboratory and onsite measurements were combined to allow for an evaluation of the traffic-related pollution and its effects on stone buildings. More details are given in a specific paper.[2]

Data from five cities, Bamberg, Würzburg, Mainz, Essen, and Munich were used to obtain an overview of the situation in these cities being characterized by different climate and traffic volume *(Tab. 1)*.

Maps of traffic volume were drawn and superimposed to maps of the stock of cultural heritage buildings. Then the

City	Area (km2)	Inhabitants	Motor vehicles
Bamberg	54.62	70,863	44,168
Würzburg	87.63	124,577	71,342
Mainz	97.74	202,756	107,004
Essen	210.30	566,862	307,943
Munich	310.70	1,388,308	771,625

Table 1: Size, inhabitants and number of registered motor vehicles of the selected cities. Data from 2011, from [3]

Rack code	City	Building	Average traffic volume (vehicles/d)	Rack: Horizontal distance from street	Rack: Height above street
BA	Bamberg	Nonnenbrücke 1	15,000	1 m	5 m
WÜ, up	Würzburg	Residenz	15,000	4 m	up: 23 m
WÜ, low	-	-	-	-	low: 4 m
MZ, up	Mainz	-	-	up: 35 m	up: 31 m
MZ, low	-	Christuskirche	27,000	low: 24 m	low: 13 m
E	Essen	Wasserturm	30,000	2 m	6 m
M	Munich	Bayer. National-museum	52,000	5 m	3 m

Table 2: Exposure sites (2 racks at Mainz and Würzburg - up: upper rack, low: lower rack; from [3]

number of buildings positioned along main road and thus exposed to increased traffic emissions was determined.

Using data from air quality survey and meteorological stations, the dose-response functions from the MULTI ASSESS program[4] were calculated for the recession of the reference material Portland Limestone.

In each of the five cities one historic building in a heavy-trafficked road was chosen for the exposure of several passive samplers *(Tab. 2)*. Special exposure racks were constructed and fixed at the facades of the buildings in traffic-near positions. In Würzburg and Mainz, racks were mounted at two different heights of the buildings. Then slabs of three stone varieties, Portland limestone, Baumberger sandstone, and Carrara marble were exposed in sheltered and unsheltered positions. Only in Mainz a Mank's carousel was additionally installed to allow for a comparison with literature data.

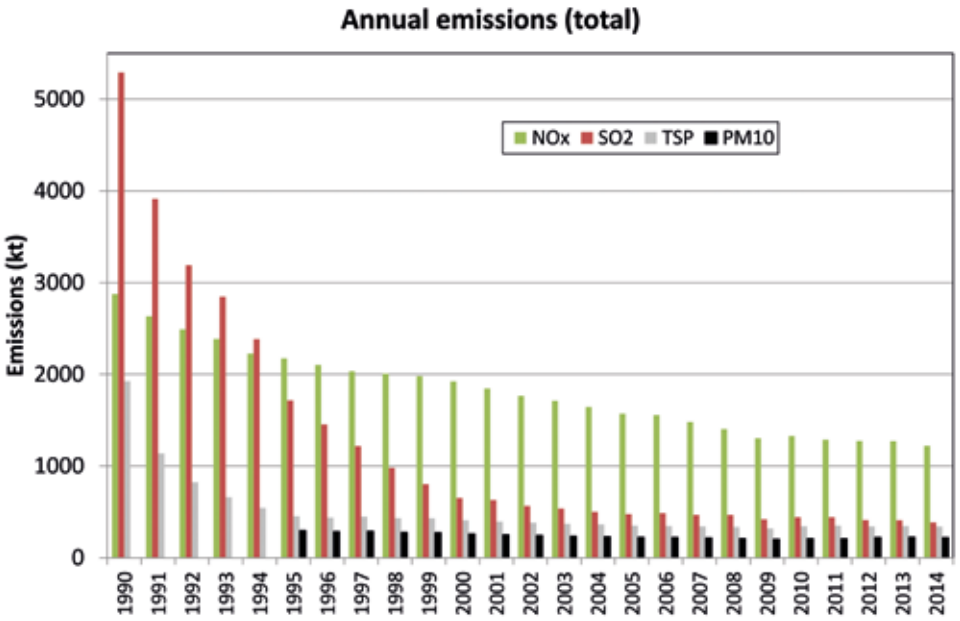

Figure 1: Development of pollutant emission from 1990 to 2010. Data: Umweltbundesamt [8] TSP: Total suspended particles.

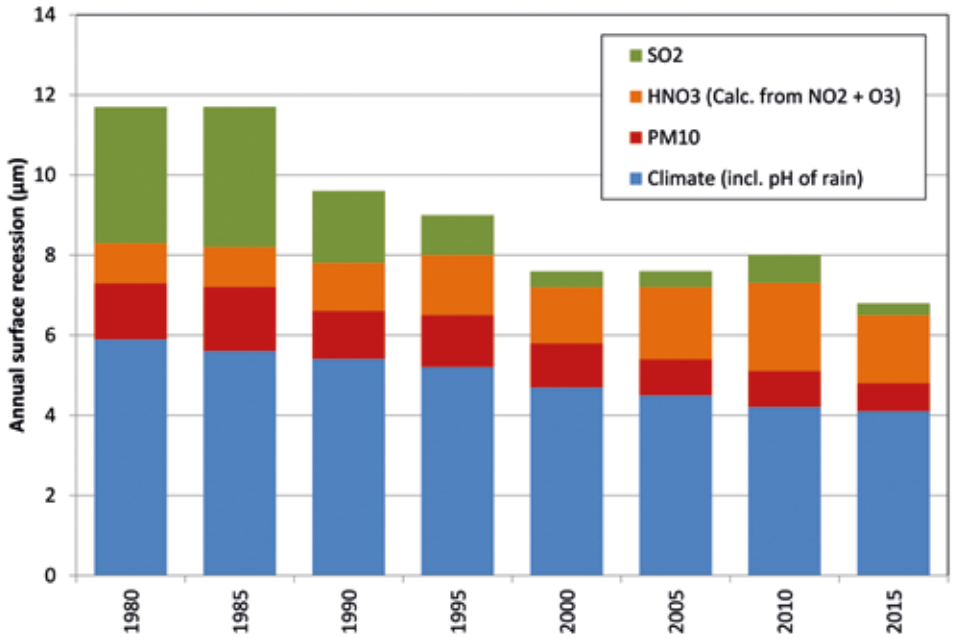

Figure 2: Calculated recession rates of Portland limestone under environmental conditions of the Karlsplatz at Munich during the last 35 years. Modified from [9]

On all racks horizontally oriented rain-sheltered passive samplers made of a boron substrate were used to collect particulate matter for ESEM studies and EDX-analyses. The samplers were replaced bi-monthly during one year. The collected particles were characterized morphologically and chemically by semi-automated individual-particle analysis with a Scanning Electron Microscope (FEI Quanta 200 Feg) combined with energy-dispersive microanalysis (EDX). Due to the large spectrum of particle sizes, 2,000 particles from each sample were analysed within two size classes (300 nm to 3 µm and > 3 µm). Image analysis showed that the coverage of the substrates was mainly due to the deposition of particles of the > 3 µm fraction. Therefore so far only the larger fraction is evaluated. Altogether approximately 42,000 particles were analysed. Each particle was checked visually and after elimination of artefacts and particles with poor images about 31,000 particles were evaluated. Based on chemical composition and morphology, the particles were classified into 14 groups, and the relative particle number, as well as the relative and absolute area coverage of each group was calculated.

Surface Active Monitors (SAM) according to Rumpel[5] are filter papers impregnated with a solution of alkali carbonate. They were used to sample sulphate, nitrate and chloride and were replaced bi-monthly.

The contents of chloride, sulphate and nitrate of the SAM filters and of the stone slabs that were exposed under sheltered conditions were analysed by ion chromatography (Dionex ICS 1000).

Diffusive samplers NO_2 and HNO_3[6,7] were provided and analysed by courtesy of the IVL Swedish Environmental Research Institute.

3. Results

Air pollution by sulphur dioxide has been diminished drastically. The emissions of nitrogen oxides and fine particulate matter were also reduced, but to a much lower extent *(Fig. 1)*.

Calculation of recession rates of the reference material Portland limestone via the MULTI ASSESS formulae[5] show a decline of recession caused by the lowering of pollutant concentrations with time. These calculations need an input of data of air pollution (SO_2, HNO_3, pH of precipitation) and climate (relative humidity). Missing HNO_3 data are calculated from the concentrations of NO_x (calculated as NO_2) and ozone, and relative humidity[5]. By setting one of these factors after another to zero, the influence of the single pollutants on the calculated recession rate can be evaluated. *Fig. 2* shows the result of such calculations with environmental and meteorological data from Munich. Although the recession rate decreases continually with time, an increasing impact of HNO_3 (more precisely NO_2 and O_3) on the weathering of limestone is observed.

The question of the role of the actual pollutant regime on historic buildings is not an academic one. In several German cities the stock of historic buildings being positioned along main roads and thus being exposed to increased traffic emissions caused by more than 5,000 vehicles daily is considerable *(Tab. 3)*.

Single particle analysis of deposited particulate matter shows a distinct increase of chloride particles in the winter months at several buildings *(Fig. 3)*. This proves, that the deposition of chloride particles is mainly due to the redispersion of de-icing salt by car traffic. Calculating annual surface coverage rates from the bi-monthly samples, it can be shown, that at Munich and at Bamberg about 10 % of a horizontal, rain-sheltered surfa-

	Listed monuments	Therefrom at main roads	Number of historic buildings along main roads				Exposed to traffic volume > 5,000 vehic./d	
Traffic volume (x10³ vehicles/d			< 5	5-20	20-40	>40	Total	%
Bamberg (city)	1348	507	163	344	-	-	344	26
Würzburg (city)	802	318	222	87	9	-	96	12
Mainz (part of city)	866	328	167	94	57	10	161	19
Munich (part of city)	4882	1212	90	702	344	76	1122	23

Table 3: Stock of listed historical buildings and monuments exposed to elevated traffic volume. Data from [10]

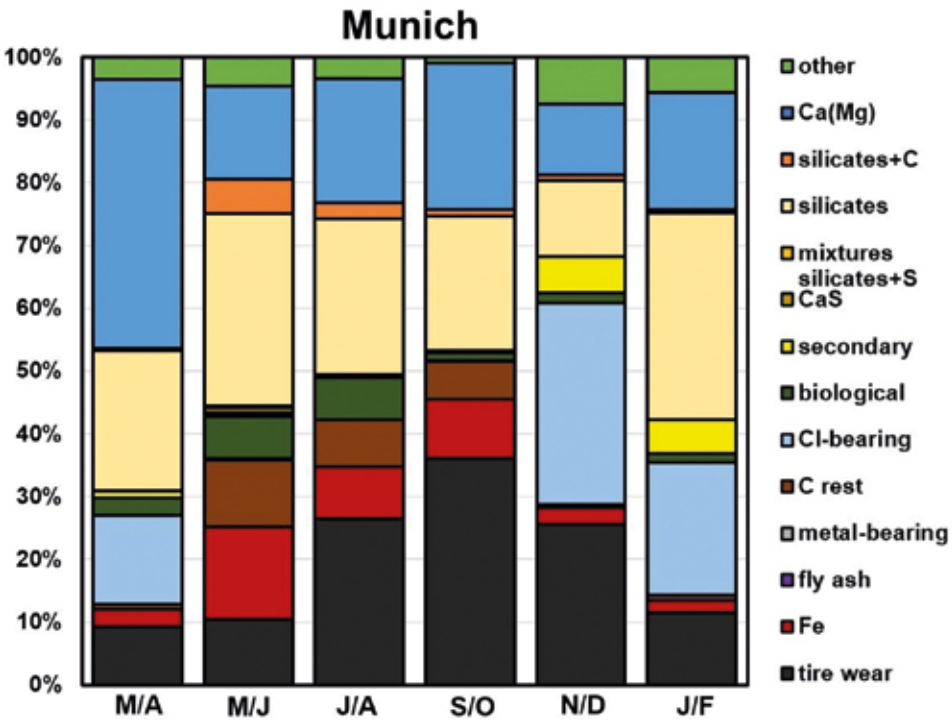

Figure 3: Variation of the composition of deposited particles > 3 µm at Munich (relative surface coverage). Letters on x-axis indicate bi-monthly sampling. From [11]

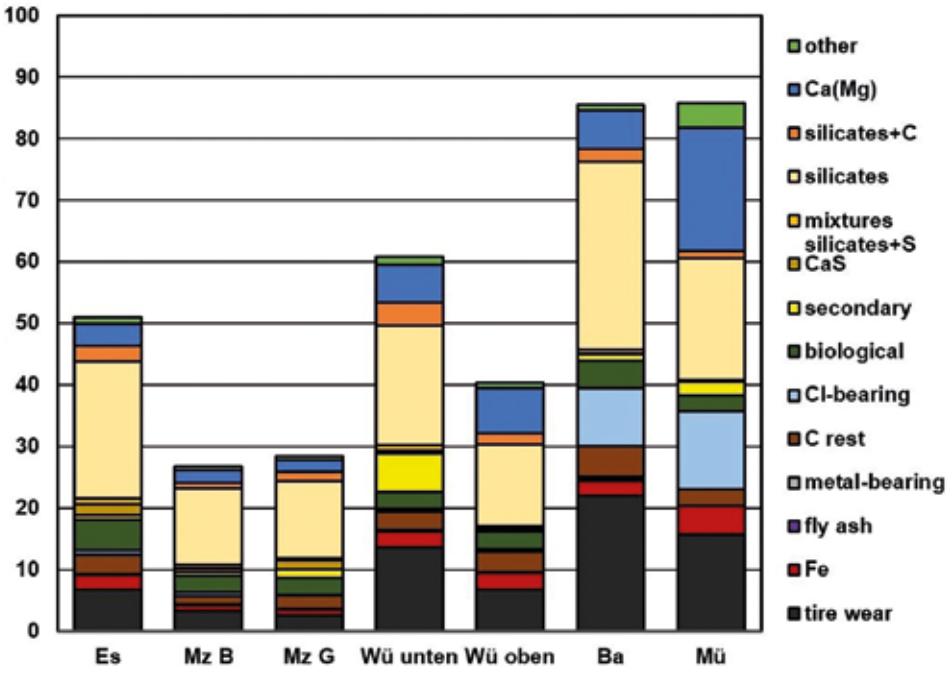

Figure 4: Calculated annual area coverage by particle deposition. From [11]

ce might be covered by chloride particles after one year *(Fig. 4)*. The deposition of redispersed chloride particles predominantly affects the lower parts of buildlings, but in two cases it was found also in remarkable height (MZ, low and WU, up). Using surface activated monitors (SAM), the deposition rates are higher for nitrate compared to sulphate. Using however natural stone samples, more sulphate than nitrate is deposited *(Fig. 5)*. It is supposed, that this is caused by the ability of the SAM-samplers to bind nitrogen oxide gases chemically, while at stone surfaces gaseous substances are only bound physically by weak adhesion forces. So the input on SAM-filters seems to depend on the concentration of NOx-gases, while the input on stone samples is assumed to be determined by the formation of HNO_3.

The measurements of HNO_3-concentrations in air directly at the facades show two trends: First there is a distinct seaso-

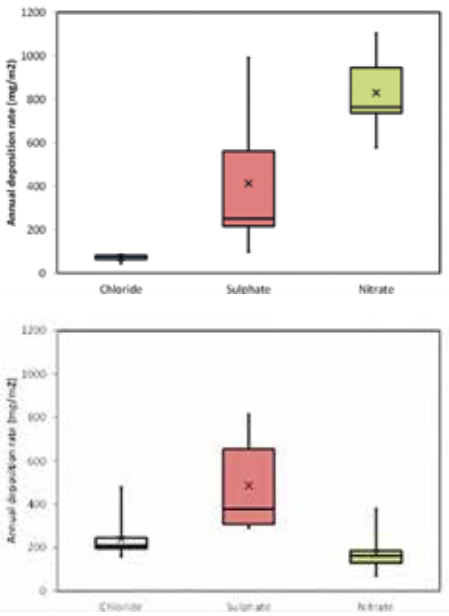

Figure 5: Annual deposition of salts on passive samplers from historic buildings situated at traffic-rich urban main roads. Left: Results for SAM-samplers. Right: Results for stone samples.

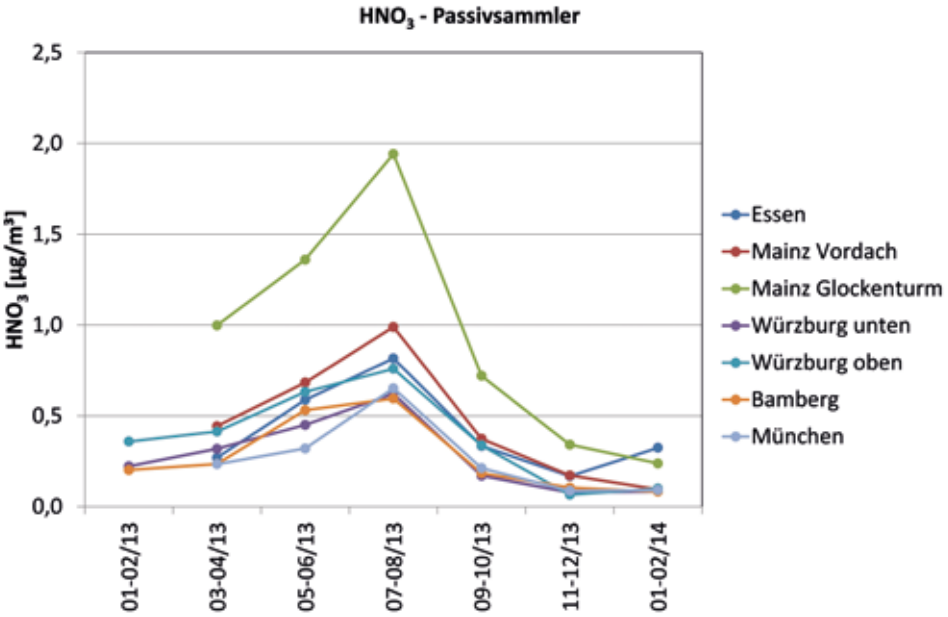

Figure 6: Results from passive sampling of HNO_3 at various facades. Data by courtesy of the Swedish Corrosion Institute, from [12]

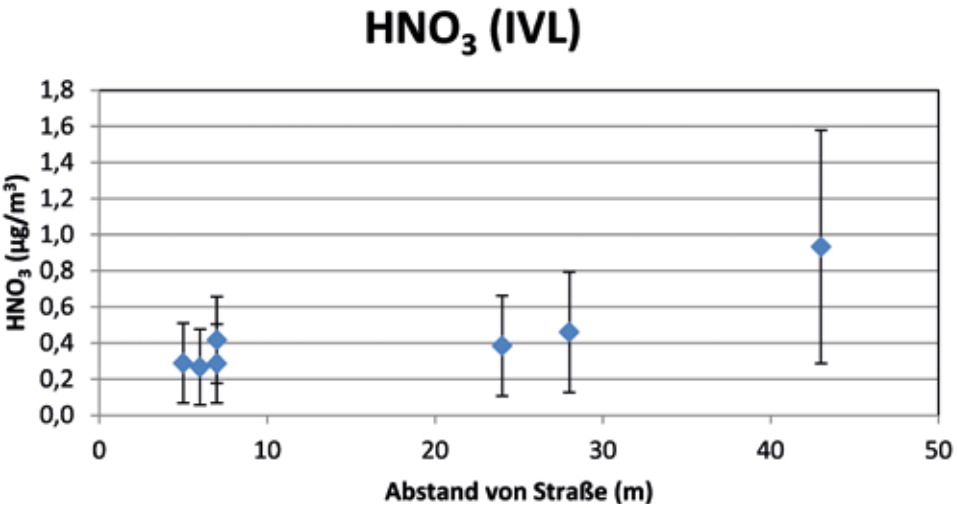

Figure 7: Mean values of the data from figure 6, ordered by the distance from the emission source. Data: see fig. 6

nal dependency with maximum values in summer *(Fig. 6)* and secondly a positive correlation with the height of the exposure racks at the building is indicated *(Fig. 7)*.

4. Discussion

While the observation from *figure 6* is due to the intensity of solar insolation, the dependency on building height could indicate that the formation of HNO_3 from NO_2 takes some time. HNO_3 seems not to be formed immediately at the source of the exhaust emissions, but in some distance. This could indicate that the reaction of nitrogen oxides with water and oxygen might to be rather slow. A consequence of this time-dependency might be a formation of HNO_3 not only at the higher parts of the buildings but also in the side roads, where it is superimposed by the dilution effect due to the minor NO_x emission rates.

5. Conclusions

Although the environmental conditions have been significantly improved over the last decades, there is still an impact of pollutants on many historic buildings. Besides accelerated soiling by tire wear and others the deposition of salts takes place on facades. Chloride and sulphate is deposited in form of fine particulate matter. Chlorides originate mainly from the redispersion of de-icing salt.

The role of nitrogen oxides and HNO_3 is not very clear up to now. The contradictionary results obtained by using different sampling materials need further research on the formation and deposition of nitrates.

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