

## EFFECTS OF ACID DEPOSITION ON CONSTRUCTION MATERIALS

J.C.Th. HOLLANDER and R.W. LANTING  
TNO Division of Technology for Society, P.O. Box 217, 2600 AE Delft  
(The Netherlands)

### ABSTRACT

Deposition of anthropogenic air pollutants causes significant incremental damage to economically important construction materials in addition to that caused by natural weathering. This paper discusses the effects of acidic air pollutants as well as those of other factors, such as material properties and climate, which are strongly interrelated with the former. Sulphur dioxide and its transformation products have been identified as the major active anthropogenic species in the atmospheric corrosion of metal structures and building stones.

Economic damage due to increased costs of maintenance, repair and replacement are discussed in the second part of this paper. Estimates of the costs for painted and galvanized steel, which have been identified as the economically most important technical materials, range from an annual US \$6 per capita in earlier studies to US \$2 in more recent ones.

### INTRODUCTION

Materials exposed to the ambient atmosphere are subject to many environmental influences inducing deteriorative effects, such as corrosion of metals, decay of building stones, and erosion and discoloration of paints. It has long been recognized that, in addition to natural environmental factors, air pollutants can accelerate the degradation of many types of materials.

Recently, the role of acid and acidifying air pollutants has become the cause of considerable concern at both national and international level. This concern has centred mainly round the harmful effects associated with acid deposition, such as the die-back of forests, freshwater acidification in environmentally sensitive areas resulting in loss of fish stock, changes in the chemistry and biology of soils resulting in loss of elements essential for healthy vegetation, and serious damage to historical monuments involving the loss of aesthetic appearance. Corrosion and erosion of construction materials apparently plays a less noticeable role in the concern on acid deposition.

Today, there is a considerable body of literature describing the effects of air pollutants on construction materials as determined by both laboratory and field experiments. In most of these studies sulphur dioxide figures prominently and is at the same time the most frequently discussed representative of the acidifying air pollutants.

If one considers the sheer volume of constructions exposed to the deteriorative effects of (acidic) air pollutants it is clear that even a small incremental damage to a unit material has a significant economic impact resulting from increased costs of maintenance, repair, replacement, or substitution by less sensitive (more expensive) materials. This line of thought has already led to considerable effort in the quantification of air pollution damage to construction materials in terms of economic costs. A reliable estimation of costs, setting the price of air pollution control against the benefit of the damage avoided, might be a powerful instrument in the hands of national and international policy makers.

This paper discusses the effects of anthropogenic acidic air pollutants on construction materials known to be economically important. Thus, the discussion will be limited to the effects of the primary air pollutants,  $\text{SO}_2$  and  $\text{NO}_x$  and the sulphates and nitrates formed from them, together with the associated  $\text{H}^+$  ion.

Although it is recognized that the complex atmospheric (photo-) chemistry of oxides of nitrogen and hydrocarbons plays a key role (as the driving force) in the conversion of the primary pollutants  $\text{SO}_2$  and  $\text{NO}_x$  into their acids through the production of hydroxyl radicals, hydrogen peroxide, and ozone, this point is only mentioned here and not further elaborated.

#### IMPACT MECHANISMS

The atmospheric deterioration of materials is a very complicated process that involves the physical and chemical nature of the materials as well as environmental factors. The latter can be divided in climatological factors or more specifically the micro-climate around objects and factors related to the chemical and physical properties of both natural and anthropogenic air pollutants.

#### Deposition processes

The first step in the mechanism of deterioration of any material by air pollutants obviously is their transport to the surface of the material,

a process generally described as deposition of air pollutants. This process includes both the supply to a surface of pollutants dissolved in rain water (wet deposition) and of gaseous and particulate air pollutants in the absence of rain (dry deposition).

The effects of wet deposition are difficult to assess owing to the event-like nature of precipitation, its varying intensity, and complex interaction with the exposure history of materials. With respect to the event-like nature of precipitation it must be noted that the data on concentrations of acidic species in rain water have been and still are mainly based on long term integrating sampling techniques and do not provide data on the flux of pollutants during a single event. The main acidic species supplied by wet deposition are hydrogen ion and sulphates and nitrates formed by atmospheric transformation of the primary pollutants  $\text{SO}_2$  and  $\text{NO}_x$  during their transport through the atmosphere from their source areas to remote and relatively unpolluted areas. Wet deposition is therefore considered to contribute substantially to the deposition flux of pollutants in such areas.

Dry deposition of air pollutants is a complicated process described by a parameter called deposition velocity, which is simply the ratio between the flux of a pollutant to a material surface and its concentration. This parameter is by no means a constant, but depends on material properties, climate factors, and chemical and physical properties (particulate, gaseous) of the individual pollutant. It has been emphasized recently that these dependencies must be borne in mind when relating deterioration rates obtained from field experiments to concentrations of air pollutants (ref. 1,2). In theory, monitoring of air pollutant deposition fluxes would be more relevant with respect to atmospheric deterioration rates, however, especially in the case of gaseous air pollutants, data for air pollutant concentrations are more abundantly available and can be readily used to derive necessary emission reductions. The main acidic species in dry deposition are the primary gaseous air pollutants  $\text{SO}_2$  and  $\text{NO}_x$  and their conversion products, gaseous nitric acid and particulate sulphuric acid, sulphates and nitrates. Since, unlike wet deposition, dry deposition is continuous and since the deposition flux is proportional to the concentration of an air pollutant, it is the dominant process in areas close to sources of primary air pollutants. These are generally urban and industrial areas where the majority of sensitive and economically important materials are located.

The relative importance of wet and dry deposition of air pollutants containing sulphur is illustrated in Table 1 by data taken from Beilke (ref. 3).

TABLE 1

Relative importance of wet and dry deposition of sulphur containing species (long-term average), simplified from Beilke (ref. 3).

Degree of air pollution	% Removal of sulphur by wet deposition	% Removal of sulphur by dry deposition
Polluted regions SO <sub>2</sub> : 50 µg.m <sup>-3</sup>	10-20	80-90
Moderately polluted	15-25	75-85
Unpolluted regions SO <sub>2</sub> : 1-10 µg.m <sup>-3</sup>	35-65	35-65
Large European region	35-50	50-65

Although these data are uncertain (ref. 3), mainly owing to the difficulty of quantifying dry deposition processes, they illustrate the general trend that dry deposition is responsible for the removal from the atmosphere of some two thirds of the sulphurous pollutants it contains, at least in the long term and over a large budget area. For NO<sub>x</sub> and its conversion products the relative contributions of wet and dry deposition are less easy to ascertain, partly because less is known about their dry deposition behaviour, and partly because the relationships between the concentrations of NO and NO<sub>2</sub> and their conversion products are more complex (ref. 3).

The problem of the relative contributions of wet and dry deposition is closely related to that of contributions from local or remote sources to the deposition of air pollutants. Its solution is very important for the design of an effective abatement strategy that mitigates the effects of acid pollutants on materials as well as other harmful effects of acid deposition.

#### Material properties

The size of an object or structure, as well as its shape and surface roughness determine the characteristics of the boundary layer of air surrounding it, and with it the aerodynamic resistance to transfer of pollutants from the atmosphere to its surface (ref. 2, 4, 5). Lipfert and Wyzga (ref. 4) calculated deposition velocities for structures (or structural elements) of various sizes, and found a dramatic increase by more

than one order of magnitude for objects of small diameter (fence wire, 0.001 m diameter) as compared to objects of large diameter (storage tank, 10 m diameter). They also point to the pitfalls in applying corrosion rate data from small test plates to objects that are greatly different in size leading to possible biases of a factor of 3 or more in the estimated physical damage.

Design and geometry of objects determine to a large extent the degree of accumulation of water and salts, and thus the degree of interaction with the material of adsorbed and absorbed pollutants.

Surface texture is also an important factor in the absorption of pollutants. Porous materials absorb more pollutants and moisture than do non-porous materials. Weathering may increase the roughness of a surface, and so cause deposition and absorption of pollutants to increase.

The chemical properties of an exposed surface can increase the resistance to transport of pollutants beyond that governed by aerodynamic factors. Slow reaction rates or diffusion processes on the surface decrease the dry deposition rate. Important porous materials such as concrete, mortar, and cement rendering all have alkaline properties which make them perfect sinks for acid pollutants. Sulphur dioxide is rapidly adsorbed on wet zinc surfaces, but on a perfectly dry zinc surface the deposition rate falls to zero when a monolayer of adsorbed  $\text{SO}_2$  has built up (for which an exposure dose of roughly  $25 \mu\text{g}\cdot\text{m}^{-3}\cdot\text{h}$  is needed) (ref. 6).

#### Climatic factors

Relative humidity is the most important variable affecting atmospheric corrosion. Temperature and relative humidity govern the formation of a water film on the material surface. Since water is necessary as a medium for chemical reactions to proceed, the time the surface is wet is an important factor. Moreover, the example of the deposition of  $\text{SO}_2$  on zinc in the previous section shows that surfaces only absorb pollutants in significant amounts when the surface is wet. The time during which a water film is present on a surface is called the time of wetness, operationally defined as the time a certain critical value of the relative humidity is exceeded. This critical value depends slightly on the nature of the material (ref. 1). The presence of hygroscopic corrosion products and hygroscopic salts deposited on the surface may decrease this critical value, and so lengthen the time of wetness. The presence of water on a surface is a more or less periodic event causing the corrosion process to proceed discontinuously. The time of wetness depends on the prevailing climate and may range from 20% to well over 60% of real time.

A specific case is the formation of dew. Compared to rain precipitation the amounts of water deposited during a dew period are only minor (about  $0.05 \text{ ml.cm}^{-2}$ , corresponding to 0.5 mm of precipitation). In combination with the preceding dry deposition of acids and acid forming species this can lead to a very strongly acidic environment on the receptor surface. Under these conditions an almost stoichiometric reaction of the acid with the surface material will take place (ref. 6, 7).

Rain also causes wetting of surfaces, but the effects of precipitation on the corrosion process go much further. They depend both on its intensity and its chemical composition. In the case of non-porous surfaces, a drizzle or light shower of short duration promotes attack of materials by wetting the surface and bringing corrosive agents dissolved in the precipitation to the surface. Heavy and longer lasting precipitation events may have the main effect of diluting and washing away such agents and corrosion products, so decreasing the corrosion rate. When the corrosion resistance depends on the presence of protective layers of corrosion products, as in the case of metals, dissolution of these products increases the corrosion rate. The solubility of metal oxides, hydroxides and carbonates increases strongly at pH values less than 4. Metals like zinc and copper are therefore sensitive to precipitation with a low pH. The processes described above show that the effects of rain strongly interact with those of the deposition preceding the rain event.

The main damage caused by rain to porous materials arises from the uptake of large amounts of (polluted) water by capillary action, and the ensuing secondary effects due to hydration and crystallization of migrating salts with moisture as the driving force.

The microclimate in the immediate vicinity of objects can differ markedly from the general macroclimate. Changes of the wind velocity profile have already been mentioned. In densely built-up urban areas, the so-called heat island effect strongly influences the time of wetness.

#### Air pollutants

Natural pollutants of importance in material deterioration are carbon dioxide, chloride ion, small amounts of naturally occurring other ions, and dust from natural sources.

The main effect of carbon dioxide on cementitious stone material is the formation of calcium carbonate (carbonation), with simultaneous reduction of the alkalinity of the stone matrix. It also reacts with metal hydroxides to form metal carbonates, which are more easily leached out of the protective corrosion layer by highly acidic rain. Although a natural

constituent of the atmosphere, significant anthropogenic emissions of carbon dioxide occur in urban and industrial areas.

Chlorides contribute to the electrochemical corrosion of painted and bare metals. When penetrating the carbonation zone of concrete, they may initiate corrosion of the reinforcing steel. Chlorides can also react with sulphuric or nitric acid to form highly corrosive hydrochloric acid. In coastal and marine environments the main source of  $\text{Cl}^-$  is sea spray. The Dutch marine climate and the concentration of large industrial and urban centres with a significant stock of materials in a narrow zone parallel to the coast make chloride one of the dominating factors in atmospheric corrosion of economically important groups of materials. Another important source of chloride is the intensive use of de-icing salts on roads. Hydrochloric acid and chlorides are also emitted in the burning of household refuse and coal with a high chloride content.

The main effect of re-entrained dust from natural sources is the soiling of surfaces. However, interaction of gaseous air pollutants and deposited dust may lead to local formation of aggressive species causing pit corrosion, as in the case of aluminium. On the other hand, a dust deposit may shield a surface from acid pollutants, especially when it is (slightly) alkaline (ref. 8).

The major anthropogenic air pollutants causing damage to construction materials are  $\text{SO}_2$ ,  $\text{NO}_2$ , and their oxidation products  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ , and the  $\text{H}^+$  ion. These acidic air pollutants accelerate the natural weathering processes. Damage from deposition of acidic air pollutants either dry or wet is generally indistinguishable from damage caused by the natural phenomena, although this is precisely what is needed if an effective abatement strategy is to be undertaken. In addition, the strong interaction between natural factors and anthropogenic air pollution makes it very difficult to demonstrate and quantify the specific contributions of (individual) anthropogenic acidic air pollutants.

The effects of acidic deposition on materials have been studied under both laboratory and field conditions.

Controlled exposure of materials in the laboratory offers the advantage that the specific effects of a single pollutant or environmental factor can be isolated. However, to produce measurable damage in a reasonable time, the material often has to be exposed continuously to extremely high pollutant concentrations. Moreover, the complex synergism or antagonism of environmental factors and pollutants makes it difficult to reproduce realistic conditions. Nevertheless, laboratory experiments are of great help in elucidating the mechanisms of deterioration, and establishing

fundamental data on reaction stoichiometry and deposition velocities. Edney et al. (ref. 6, 7), in a series of very elegant experiments with galvanized steel samples, have recently demonstrated the power of experiments in advanced computer-programmed environmental chambers, allowing for more realistic conditions, combined with chemical analysis of dew produced by chilling test samples, instead of measuring physical damage.

Field studies involve the exposure of materials to ambient atmospheres representing various combinations of pollutant concentrations and other environmental factors. Statistical analyses of physical damage as a function of pollutant concentrations and other environmental factors afford functional relationships between these factors, and may be used to predict damage from known concentrations and environmental factors for other locations. The major disadvantages of this approach include the difficulty of isolating the effect of a single pollutant owing to its interaction with other variables (covariance of pollutant concentrations), and the restricted number of variables that can be monitored, which may lead to effects being attributed to the wrong variables. Monitoring that does not cover all relevant variables is likely to be responsible for much of the unexplained variance in statistical data and, consequently, for the lack of predictive power of the exposure-response relationships derived from them.

In order to judge the results of statistical analyses of field data, it is necessary for the functional forms of the exposure-response relationships (relating physical damage to pollutant concentrations) to be so formulated that they reflect the mechanism of the deterioration process. In this way at least some of the coefficients obtained can be compared to theoretical values calculated on the basis of dry deposition velocities, reaction stoichiometry, and other known details of the deterioration process. Several samples of this type of analyses have recently been published (ref. 1, 5, 8).

Sulphur dioxide figures prominently among damaging acidic air pollutants, followed by the wet and dry deposition of secondary pollutants ( $H^+$ ,  $SO_4^{2-}$ ,  $NO_3^-$ ). Field studies have revealed no effects of sulphate aerosols. Their deposition velocity is much lower than that of  $SO_2$ . For  $NO_2$ -interactions ( $NO$  is estimated to play a less important role) only limited evidence is available. However, there is a significant lack of  $NO_2$  ( $NO_x$ ) measurements in most exposure programmes. Laboratory experiments (ref. 7) have shown that the dry deposition velocity of (gaseous) nitric acid, which is highly corrosive, on zinc is comparable to that of  $SO_2$ . Its presence and formation in the atmosphere are still not well known.

## DAMAGE TO SPECIFIC CONSTRUCTION MATERIALS

From an economic point of view, the most important construction materials at risk from acidic deposition are painted and unpainted metals. Building stones on a cementitious base, like concrete and mortar, come second, but their susceptibility to attack by pollutants crucially depends on their formulation and quality.

### Unprotected carbon steel

Unprotected carbon steel is very sensitive to corrosion and has been well studied. It is, however, normally given a protective coating, and so is not further discussed in this paper.

### Zinc

Zinc is technically very important and mainly used to protect steel (galvanized steel). Zinc corrosion has been studied in numerous field exposure programmes, and a wide variety of exposure-response or damage functions is available. In a recent study Lipfert et al. (ref. 1) tried to reconcile damage functions by analysing the data bases of 8 exposure programmes in the US, Canada, Western and Eastern Europe with a total of 72 exposure sites. A damage function was derived not only from a purely statistical treatment of the data, but also from current knowledge of the corrosion mechanism. The result of the statistical analysis was judged by comparing the derived coefficients with theoretical ones calculated from the stoichiometry of the  $\text{SO}_2$ -Zn reaction and deposition velocity during time of wetness (RH above 85%). Hence the function reflects typical mechanistic effects of the corrosion process. This damage function makes it possible to look at the effect of acid precipitation versus dry deposition of  $\text{SO}_2$ . Figure 1a shows the calculated corrosion rates as a function of the concentration of  $\text{SO}_2$  for various pH values (fraction of time wet: 0.35; precipitation:  $1000 \text{ mm year}^{-1}$ ).

There is a strong increase of corrosion for pH values below 4.5, corresponding to the increasing solubility of the protective layer of natural corrosion products (zinc hydroxide and carbonate). Note also the fourfold increase of the corrosion rate at zero  $\text{SO}_2$  concentration when the pH decreases from 5.6 to 3.5. The slight increase of the slope with decreasing pH points to a synergism between the effects of  $\text{SO}_2$  and  $\text{H}^+$ . Due to the removal of the protective layer by  $\text{H}^+$  the  $\text{SO}_2$  deposition will attack zinc more directly, increasing the corrosion rate.

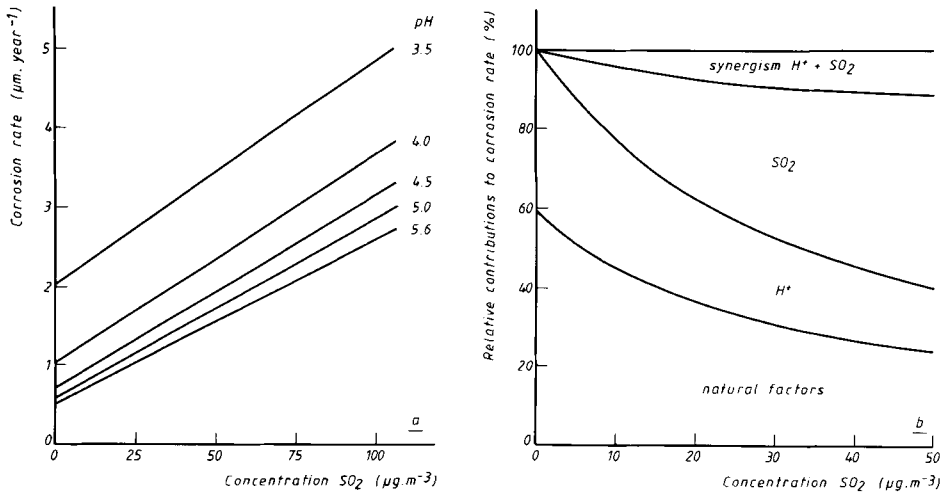


Fig. 1a. Corrosion rates of zinc (4-year average) for different pH values as a function of ambient  $\text{SO}_2$  concentrations.

Fig. 1b. Relative contributions of  $\text{SO}_2$  and  $\text{H}^+$  (pH = 4.2) to the corrosion rate of zinc as a function of ambient  $\text{SO}_2$  concentration.

Figure 1b shows the relative contributions, calculated from the same damage function, of  $\text{H}^+$ ,  $\text{SO}_2$  and natural factors (including effects of unspecified pollutants) to the corrosion rate of zinc at a rain pH of 4.2 (representative for Central Europe). It shows that at an annual average  $\text{SO}_2$  concentration of  $10 \mu\text{g}\cdot\text{m}^{-3}$ , the combined effect of  $\text{SO}_2$  and  $\text{H}^+$  is of the same magnitude as that of natural factors. At higher  $\text{SO}_2$  concentrations the effects of  $\text{SO}_2$  rapidly begin to dominate those of  $\text{H}^+$ . Hence, the overall effect in unpolluted areas is mainly due to wet deposition of  $\text{H}^+$ , whereas in polluted areas the main effect is caused by dry deposition of  $\text{SO}_2$ .

The role of  $\text{NO}_2$  was not assessed in Lipfert's study (ref. 1). A conservative estimate on basis of its deposition velocity and stoichiometry (ref. 5, 7) indicates that its effect on zinc is at least a factor four less than that of  $\text{SO}_2$ .

### Painted steel

Reliable damage functions for present day paint formulations are not available.

Apart from natural factors such as chloride and UV radiation,  $\text{SO}_2$ ,  $\text{O}_3$ ,  $\text{NO}_2$  and dust are suspected of interacting. Shortened lifetimes are often attributed to deposition of pollutants between the pretreatment and painting of an object resulting in undercutting by rust and loss of adhe-

sion. Other processes are the penetration of the paint film by corrosive agents at sites where the layer is thin. Existing damage functions for paint address only erosion of the paint film as physical damage parameter. This can only partly explain the observed damage in real life.

#### Cementitious materials

There is much concern about a type of damage to reinforced concrete that is commonly referred to as concrete disease. The high alkalinity of the cement matrix protects the reinforced steel from corrosion in well made concrete. The main process leading to loss of alkalinity and hence to corrosion of steel involves reaction with  $\text{CO}_2$  and is called carbonation. When the surface layer, covering the steel, is thick enough, acid pollutants will only cause the formation of soluble and insoluble salts, which are leached out or washed away by acid precipitation causing the surface to become etched. If, on the other hand, the surface layer is not thick enough, the carbonation layer will reach the reinforced steel and the passivation of the steel will be lost. Chlorides, and to some extent nitrates as well, are able to penetrate the carbonation zone, so initiating corrosion of the steel. The voluminous corrosion products of steel cause cracks, which lay the embedded steel more open to attack by  $\text{SO}_2$ .

Reinforced concrete is a typical example of an otherwise rather durable material that can be rendered very susceptible to attack by air pollutants through faulty building practices. It will be clear that for such complex processes no damage functions are yet available, and are very difficult to develop.

#### Structures immersed in soil or water

Effects of deposition of acidic air pollutants are noted in all three compartments of the ecosystem, viz. air, soil and water. Structures embedded in soil or immersed in water will suffer indirect effects of acidification of these media.

Buried concrete foundations, ducts, steel tanks and lead jacketed cables are at risk when the pH of soil falls below 5.

A well-known problem caused by acidification of ground and surface water is the internal corrosion of pipes transporting drinking water in areas where this water is not treated to adjust the pH. The resulting contamination with salts of lead, copper, cadmium or zinc may eventually lead to health effects.

## ASSESSMENT OF ECONOMIC DAMAGE

Cost-benefit analysis, in which the total costs of proposed scenarios for emission abatement are set against the benefits (in terms of avoided damage), is increasingly used to assist in making policy decisions. For assessing economic damage from air pollution there are essentially three basic approaches.

The first, or physical damage approach, is based on established exposure-response relationships or damage functions.

The second, or market approach, assesses for areas with different pollution levels the demand for services and goods, sensitive to damage by air pollutants, or the costs of maintenance and repair.

The third, or non-market approach, estimates the willingness of the public at large to pay for any improvements in the environment.

The two last mentioned approaches fail to single out the damage caused by an individual pollutant. The physical damage approach does have this capability, and is the most straightforward for construction materials. On basis of critical damage levels, the physical damage derived from the damage functions is translated into changes in useful lifetime of materials at risk. The cost of incremental maintenance, repair and replacement is used to convert physical damage into monetary terms. This approach requires a great deal of data, which may not always be available. To mention some of the problems:

Most dispersion models for predicting air quality and deposition fluxes are not able to assess the actual exposure regime of objects in built-up areas such as cities.

Damage functions are derived from exposure of small specimens in the field and may not be representative for what actually happens to a real building (e.g. the microclimate is quite different). Moreover, no reliable damage functions are available for many important materials such as painted metals and stone. Considerable uncertainty is also introduced by assumptions on maintenance, repair and replacement strategies (ref. 9).

Hence, most estimates of economic damage are subject to large errors which, regrettably, are not always taken into account when making cost-benefit analyses.

However, if economic damage is expressed as a probability distribution, an approach now adapted by the United Nations Economic Commission for Europe, such estimates are valuable tools in decision making.

The economically most important construction materials are painted and galvanized steel considering the widespread use and sensitivity to attack by air pollutants. Many assessments of damage by  $\text{SO}_2$  were made in the seventies for these materials (Table 2).

TABLE 2

Estimates of total costs from damage to metals and painted metals (corrosion and protection) due to air pollution.

Country	Base year	Materials <sup>a)</sup>	Pollutant	Cost <sup>c)</sup> US \$/capita. annum (1980)	Ref.
USA	1970	1, 2	all	13.8	10
USA	1970	1, 2	$\text{SO}_2$	3.8	11
USA	1970	1	$\text{SO}_2^+$	3.1	12
UK <sup>b)</sup>	1970	1, 2	all	5.4	13
Sw <sup>b)</sup>	1970	1, 2, 4	$\text{SO}_2$	9.0	14
NL	1970	1, 2, 3	$\text{SO}_2^+$	4.5	15
OECD (11) <sup>b)</sup>	1974	1, 2	$\text{SO}_2^+$	5.2	16
				mean 6.4, sd 3.7	
NL	1978	1, 2, 3	$\text{SO}_2$	1.0	17
N	1979	1, 2	$\text{SO}_2^+$	3.1	18
USA (case study)	1985	1	$\text{SO}_2$ & $\text{H}^+$	2.6 (1985)	19
				mean 2.2, sd 1.1	

- a) 1 = galvanized steel, 2 = painted steel, 3 = painted galvanized steel  
4 = painted wood
- b) on basis of costs savings for maximum reduction of emissions
- c) approximate conversion on 1980 prices based on consumer price index for the USA.

None of these studies has afforded fully satisfactory results, mainly owing to the lack of reliable damage functions for painted steel. The estimates around 1970 vary by a factor of 3 to 4 with an overall average value of US \$ 6 (1980) per capita per annum. Studies around 1980 indicate a damage of roughly \$ 2 (1980) per capita per annum. This apparent decrease in damage costs is due on one hand to the concurrent reduction of ambient  $\text{SO}_2$  concentration levels and, on the other, to a more critical use of damage functions for paint. For other construction materials, such as stones and painted surfaces, damage estimates are highly unreliable, because the effects of acid deposition can not be separated from natural phenomena.

## ABATEMENT STRATEGIES

Abatement of the adverse effects of air pollutants on materials is usually effected along two independent routes, with different control levels. At the administrative level, improvement of air quality by reduction of emissions is the major route, whereas at the manufacturing or consumer level, the improved pollution resistance of the material itself, the aim of many industrial exposure programmes, is pursued. With respect to pollution control,  $\text{SO}_2$  and its transformation products are the main anthropogenic agents in the atmospheric corrosion of metal structures and building stones.

The contribution of  $\text{NO}_2$  has not been resolved for all technical materials. At current concentrations of  $\text{SO}_2$  and  $\text{NO}_2$  the dry deposition of  $\text{NO}_2$  is estimated to be responsible for far less than 20% of the damage caused to metals by  $\text{SO}_2$  (ref. 5, 7, 20).

The interaction between wet acid deposition and the film of passivating corrosion products on a metal suggests that the relationships between exposure and effect are not linear, even for a relatively simple system like zinc.

However, it is obvious that any reduction in acid generating pollutants will reduce the economic damage to materials.

In deciding control strategies distinction must be made between long-range transport of air pollutants and local pollution. Figure 1a shows that any further increase in rain acidity is likely to increase the corrosion rate of zinc, particularly in rural areas, where the relative contribution of  $\text{H}^+$  is high. On the other hand, urban and industrial areas contain a high density of sensitive materials, so most of the harm will be done by local pollution sources provided that these have low emission heights. Control of local emissions is therefore likely to yield the highest benefits in terms of avoided damage to materials.

The desirable degree of abatement depends on what physical or economic damage is considered acceptable. Cost-benefit analyses based on damage functions may assist in deriving the optimum solution.

Whereas the above mentioned line opts for a better environment the second line seeks to improve the pollution resistance of materials. This approach can be quite cost-effective.

Educated guesses supported by construction engineers state that roughly one quarter of the damage caused by atmospheric corrosion could be avoided by simple actions such as better design, proper material selection and application of improved quality control.

Apparently the use of new materials and building practices caused to a large extent the loss of century old awareness and craftsmanship with respect to avoidance of atmospheric degradation processes of building materials.

Since the present knowledge, often acquired by bitter experience, is only fragmentary disseminated in the building and construction trade, effort should be made to integrate this knowledge, leading to better guidance with respect to the prevention of atmospheric corrosion by receptor oriented measures.

#### CONCLUSIONS AND RECOMMENDATIONS

The process of atmospheric degradation is a complex phenomenon governed by many interrelated factors of which air pollution is only one.

Attempts to quantify the individual contributions of acidic air pollutants were initially centred on the dry deposition of  $\text{SO}_2$ , and have more recently included the role of wet deposition of  $\text{H}^+$  ions. For lack of field monitoring data, the contribution of  $\text{NO}_x$  cannot as yet be fully quantified.

Damage functions are still highly empirical, with a recent tendency to incorporate physico-chemical relationships in the design of the functional form. There is not enough knowledge available to develop purely theoretical damage functions from first principles, using the appropriate physico-chemical relationships only.

The use of air pollutant concentrations as the main variables in damage functions requires a large research effort in monitoring and investigation of factors affecting the relationships between concentration and deposition flux. An alternative would be to improve methods of measuring deposition fluxes of acidic air pollutants.

Current damage functions for painted objects and stone are only partly able to describe the total damage processes. They do not take into account influences such as variations in thickness and permeability of the paint layer and in the porosity of stone.

Most estimates of economic damage show large uncertainties, accumulated from several factors such as: the extrapolation of damage observed on small test samples to real objects or constructions, absence of reliable damage functions describing the full damage, uncertainties in assumptions on maintenance, replacement and substitution strategies.

It is obvious that any reduction in acid generating pollutants will reduce the economic damage to materials. However, more knowledge is needed

on the relative contributions of local versus remote sources and related topics, to judge the effectiveness of emission control scenarios.

Since much damage to structures can be avoided by design modification, proper material application and improved quality control, these options should also be incorporated in cost-benefit analysis.

The assessment of the effectiveness of protective treatment, when damage is already done or when protection cannot wait for improvement of air quality remains still an important research area.

#### REFERENCES

1. F.W. Lipfert, M. Benarie and M.L. Daum, Derivation of metallic corrosion functions for use in environmental assessment, Brookhaven National Laboratory report, Brookhaven, 1985.
2. F.H. Haynie, in P.J. Sereda and G.G. Litvan (Eds.), Durability of Building Materials and Components, ASTM STP 691. American Society for Testing and Materials, 1980, pp. 157-175.
3. S. Beilke, Acid deposition, an updated review on atmospheric physico-chemical aspects of the acid deposition problem in Europe, Commission of the European Communities, 1985.
4. F.W. Lipfert and R.E. Wyzga, Application of theory to economic assessment of corrosion damage, presented at ACS Symposium on Degradation of Materials due to Acid Rain, Arlington, Virginia, 1985.
5. F.H. Haynie, in S.W. Dean, Jr. and E.C. Rhea (Eds.), Atmospheric Corrosion of Metals, ASTM STP 767, American Society for Testing and Materials, 1982, pp. 286-308.
6. E.O. Edney, D.C. Stiles, J.W. Spence, F.H. Haynie and W.E. Wilson, Atmospheric Environment, 20 (1986) 541-548.
7. E.O. Edney, D.C. Stiles, J.W. Spence, F.H. Haynie and W.E. Wilson, in press.
8. F.H. Haynie, Environmental factors effecting the corrosion of weathering steel, presented at ACS symposium on Degradation of Materials Due to Acid Rain, Arlington, Virginia, 1985.
9. A.R. Stankunas, E.F. McCarthy and J.E. Yocom, Uncertainties in estimating the effects of air pollutants on materials, Proceedings of the 75th Annual Meeting of the Air Pollution Control Association, New Orleans, 1982.
10. F.W. Fink, F.H. Buttner and W.K. Boyd, Technical-economic evaluations of Air Pollution corrosion costs on metals in the US., Batelle Memorial Institute Columbus, OH, report PB 198453, 1971.
11. D.G. Gillette, Sulphur dioxide standards and material damage, J. Air Pollut. Control Assoc., 25 (1975) pp 1238.
12. B.C. Liu and E.S. Yu, Physical and economic damage functions for air pollutants by receptors, Report EPA 600/5-76-011, 1976.
13. P.M.C. Jones, An economic and technical appraisal of air pollution in the United Kingdom, Programme Analysis Unit, PAU, Berks, 1972.
14. V. Kucera, *Ambio*, 5 (1976) 243-248.
15. H.M.A. Jansen and A.A. Olsthoorn, Economische waardering van de nationale schade door luchtverontreiniging. Mozaïek van de milieuproblematiek. Report Instituut voor Milieuvraagstukken, VU, Amsterdam, 1982.
16. OECD, The cost and benefits of sulphur oxide control, OECD, Paris, 1981.

17. A.A. Olsthoorn, Schade aan materialen door SO<sub>2</sub>, Werknota 117, Instituut voor Milieuvraagstukken, VU, Amsterdam, 1975.
18. J.F. Henriksen, S. Haagenrud and F. Gram, The effect on atmospheric corrosion costs of a requirement for oils with a low-sulphur content, report NILU, 1979.
19. F.W. Lipfert, L.R. Dupuis, R.G. Malone, J. Schaedler and M.L. Daum, A case study of materials damage due to air pollution and acid rain in New Haven, CT, presented at Second U.S. Dutch International Symposium: Aerosols, Williamsburg, VA, 1985.
20. R.W. Lanting, Deterioration of materials by NO<sub>x</sub> (in Dutch), Ministerie van Volkshuisvesting, Ruimtelijk Ordening<sup>x</sup> en Milieubeheer, Publicatie reeks Lucht 12, 1983.