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Modeling the calcareous stone sulphation in polluted atmosphere after exposure in the field

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Two series of 8 samples of calcareous stone (Parisian Lutetian limestone and Richemont limestone) were exposed sheltered and unsheltered from rain for up to 3 years to the atmospheric pollution on real sites in Paris (Saint Eustache Church) and Tours (Saint Gatien Cathedral). The samples of each series were withdrawn after 1, 2, 4, 6, 12, 18, 24 and 36 month.

The distribution of Sulphur concentrations under the surface of stones was measured by pyrolysis and Infra Red elemental analysis on powder obtained by milling the stone in subsequent steps of 0.1 mm on 2.5 mm depth.

The main results can be summarized as follows:

1-On the samples *exposed to rain*, the Sulphur concentration remains equal to stone background concentration that means the Sulphur deposited between two rain events is leached by the next event.

2-The Sulphur concentration in the first step under the stone surface *exposed sheltered from rain* increases with time, but not linearly, pointing out an accumulation phenomenon.

The Sulphur concentration is 10 times higher in the Parisian limestone than in the

Richemont limestone, reflecting the lower concentration in $SO_2$of the atmosphere of Tours (3 to 4 times) and the different properties of the stones (Ca concentration, porosity…)

The sulphation does not penetrate more than 0.2 mm despite of the increasing concentration in the first step (0.1 mm).

Under 0.2 mm depth the Sulphur concentration is that of the mean natural background concentration in the stone: 0.06% for Parisian limestone and 0.04% for Richemont limestone.

A logistic model fitted well the measured Sulphur concentrations in time; it should be tested if it allows predicting the forthcoming evolution:

P(t) = K / (1 + e $^{-\alpha t}$)

where: P is the Sulphur concentration in the first step at time t, K the carrying capacity of the atmosphere in SO_2andα the intrinsic rate of growth. For the Parisian limestone: P = 1.479 / (1 + 29.4 e $^{-0.0041t}$) and for the Richemont limestone: P = 0.14 / (1 + 3.53 e $^{-0.004t}$).

This model indicates a starting phase (0 to 200 days in average), a rapid increasing phase (200 to 1000 days in average) and a phase of stabilization (after 1000 days) and it is in a good agreement with many natural phenomena tending generally to saturation