PAVE 92

COLOUR CHANGES RESULTING FROM THE WEATHERING OF PIGMENTED CONCRETE

Dr. Peter Kresse
Bayer Ag, Krefeld-Uerdingen, Germany

Not all pigments are suitable for colouring concrete. German standard DIN 53237 [1] describes laboratory methods which can be used to test the cement resistance and light-fastness of pigments for colouring concrete building materials although these same pigments then prove to be unsuitable in practice. This is illustrated in Fig. 1 which compares concrete roof tiles weathered for 12 months with unweathered tiles. Although all the pigments performed well in the laboratory tests, only the iron oxides withstood outdoor conditions.

It is a well-known fact that carbon black pigments fade when weathered outdoors [2]. Curiously, these pigments are affected less by an industrial climate than by a non-polluted atmosphere. This indicates that the weathering location is of great significance. However, not all pure inorganic pigments are suitable, as shown by the problems experienced with ultramarine blue. In order to assess weather stability and lightfastness, therefore, outdoor weathering tests are essential, if possible at various locations.

Bayer has a number of weathering stations used specially for testing the behaviour of coloured concrete. The largest of these is near our production plant at Krefeld and is pictured in Fig. 2. Most of the test samples are extruded concrete roof tiles exposed to the south at an angle of 45°. The tiles are manufactured from concrete with a cement/sand ratio of 1 : 3. The coarsest grains of sand are 4 mm in diameter. When extruded, the concrete has a water/cement factor of 0.39. We normally use a PZ 45 portland cement and sometimes a white portland cement. The smooth and very homogeneous tile surface produced allows the use of colorimetric methods to measure colour changes during the course of weathering. This is more difficult when testing concrete pavers, although we carefully test these too, as shown in Fig. 3. What is missing in tests of concrete pavers is the exposure to pedestrians or vehicles.

As the concrete roof tile formulation is similar in composition to the facing used on concrete pavers, the results obtained for roof tiles can easily be applied to pavers. Table 1 provides an overview of a major test series, the results of which I will discuss in greater detail later.

Higher quality requirements have meant that even such slight colour changes have gained significance in a field where the pigments used are generally regarded as stable, such as those in Table 1. What are the causes of such slight colour changes? Colorimetry has proved extremely useful in the examination of this question. In the following paragraphs I will attempt to briefly explain the principles of this method.

The phenomenon of colour is best represented in a three-dimensional system known as the colour space in which the light/dark axis is the vertical in the colour plane. Fig. 4 shows a diagram of this system and Fig. 5 a colour plane in this system. The further one moves along the light/dark axis towards black, the less differentiated the colours are.

The same thing happens if one moves the other way along the axis until all the colours merge into white. Fig. 6 shows a number of pigments in concrete blocks plotted as the a,b coordinates in a colour plane. This is a simplification insofar as the various samples do not all possess the same lightness and should therefore not all be plotted in the same plane.

If concrete is weathered outdoors, the colour change can be described in terms of a change in lightness and position in the colour plane, in other words, a shift of the a,b coordinates. Weathering therefore results in a three-dimensional change in the position of the sample in the colour space.

In our tests we worked with a Hunterlab or a Minolta, using the CIELAB system and Standard Illuminant C. All the measurements are differential and not absolute, thus eliminating equipment-related factors.

Let us first turn our attention to the change in lightness. Fig. 7 shows some of the concrete roof tiles from the test series mentioned earlier, both in their weathered and unweathered states. The unweathered tiles in the top row are arranged in order of their initial lightness, which is marked on them. The lightness after five years' exposure in a marine climate is also marked on the samples in the bottom row. It can be seen that the concrete which was lightest at the start is now the darkest. Very dark concrete may even become lighter during weathering. It is immaterial whether the concrete contains a pigment or not. The samples marked "0 mixture" are made from only white or grey cement and contain no pigments. Fig. 8 shows the entire series. Plotting the change in lightness ΔL^* against the initial lightness (Fig. 9) illustrates what I have just described. The number of samples which become slightly lighter during weathering is very small and seems to be a coincidence more than anything else. Under these weathering conditions concrete samples will darken by 12 lightness points at the most. Fig. 10 shows later results which correlate well and are only shown separately to prevent the confusion of a large number of measurements.

It is not just the use of different pigments which give the concrete higher or lower lightness. Using the same pigment in different concentrations will have the same effect. Fig. 11 shows a test series containing concentrations as high as 25 %, calculated on the cement content. Such an amount is, of course, excessive and is neither economically or technically justifiable. Nevertheless, comparison of the unweathered and weathered samples shows clearly that the concrete containing less pigment, i.e. with a higher initial lightness, undergoes a greater colour change as the result of weathering. Fig. 12 plots measurements taken during a number of such test series. The samples containing higher concentrations of colour pigments which are darker than the concrete itself (the exception being the white pigment) always have a lower initial lightness and therefore the change in lightness during the course of weathering is smaller. In other words, the higher the concentration of pigments with a lower initial lightness than the concrete itself, the smaller the colour change in the concrete during the course of weathering. It should be pointed out here that the human eye is particularly sensitive to changes in lightness.

What effect does weathering have on the position of coloured concrete in the colour plane, i.e. in the a,b diagram? Fig. 13 shows a section of the colour plane discussed earlier, one which is typical for the characterization of concrete. Fig. 14 shows the colour changes caused by weathering in a number of samples. The box at the end of a line represents the colour after weathering. It can be seen that, in most cases, there is a shift towards yellow-orange.

If concrete coloured with the pigments shown in Table 1 changes colour in a way which can be described by the two rules - that it becomes darker, the lighter it was to start with and that it shifts towards yellow-orange in the colour plane - then it can be assumed that there is a common reason for this change. As the greatest colour change is seen in unpigmented concrete, it is reasonable to assume that the cause is a discoloration of the hardened cement paste which is masked by the pigment. In all probability this is due to a reaction of the largely colourless iron-bearing cement phase, brownmillerite ($4CaO.Al_2O_3.Fe_2O_3$), with atmospheric carbon dioxide to produce decomposition products, some of which are yellow-red. If this is the case, then it should be possible to simulate the colour change in pigmented concrete. We carried out a purely empirical test in which a slightly dark-tinted, transparent film was laid over an unpigmented concrete sample. Looking at the concrete through the film fairly accurately represented what the concrete would look like after four to five years' weathering (Fig. 15).

Colorimetry could be used to examine exactly how good the foil represents the changes in lightness and position in the colour plane caused by weathering. However, this would be too complicated. Let us recall what has already been said, namely that the total colour change in the colour space is three-dimensional, in terms of both the colour plane a,b and the lightness coordinate L*. This colour change, expressed as $\Delta E^*(a,b)$, can be calculated using the following equation:

$$\Delta E^*(a,b) = (\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2$$

Fig. 16 shows a model representing this change.

Fig. 17 shows bar charts plotting the actual overall colour change $\Delta E^*(a,b)$ measured after weathering and the values recorded through a film for a number of concrete samples. The correlation is very satisfactory.

This result should not lead us to assume that the same correlation will exist in concrete weathered over different periods of time. During the first two years of outdoor weathering efflorescence can generally be expected to occur. This makes the surface of the concrete appear lighter than it is and makes it impossible to evaluate the actual colour change. After five years' outdoor weathering the aggregate at the surface of the concrete is exposed by erosion of the hardened cement paste. The colour of the pigmented cement paste becomes less dominant as the natural colour of the aggregate becomes more visible. The overall colour of the weathered concrete then tends more towards the natural colour of the aggregate. Ultimately, algae, lichens and mosses infest the surface of the concrete. We described the time scale of these process in a letter to our American subsidiary Mobay [3].

Conclusion

Coloured concrete, although it is usually pigmented with lightfast, weather-stable and cement-resistant products, still undergoes certain colour changes as the result of weathering. These can be classified in a uniform system in terms of lightness and colour change. The common cause of this colour change is the alteration in the colour of the hardened cement paste as the concrete weathers. The expected colour changes in concrete can be simulated in an empirical test by laying a tinted, transparent film over unweathered concrete. The colour observed is the colour the concrete would be after four to five years' weathering.

Bibliography

[1] DIN 53237 - Testing of pigments; pigments for colouring cement and lime-bound building materials (February 1977)

[2] P. Kresse, Betonwerk + Fertigteil-Technik, Issues 8 and 9, 1985

[3] G. von Szadkowski, Coloring Technologies in other Countries, Mobay Corporation, Pittsburgh, USA, 1990

Colour	Trivial name	Chemical designation	Chemical formula	Weathering te… grey cement	wh… ce…
red	iron oxide red	iron (III) oxide	α-Fe_2O_3	X	
yellow	iron oxide yellow	iron (III) oxide hydroxide	α-$FeOOH$	X	
	lightfast yellow	nickel titanium yellow	$(Ti,Ni,Sb)\ O_2$		[illegible]
	lightfast yellow	chrome titanium yellow	$(Ti,Cr,Sb)\ O_2$		[illegible]
green	chrome oxide green	chrome (III) oxide	α-Cr_2O_3	X	
	cobalt green	cobalt nickel aluminate	$(Co,Ni,Zn)_2(Ti,Al)O_4$		X
blue	cobalt blue	cobalt aluminate	$Co\ (Al,Cr)_2O_4$ $CoAl_2O_4$		X X
brown	iron oxide brown	blends of α-$FeOOH$ and/or α-Fe_2O_3 and Fe_3O_4		X	
black	iron oxide black	iron (II,III) oxide	Fe_3O_4	X	
white	titanium dioxide	titanium (IV) oxide	TiO_2	X	[illegible]

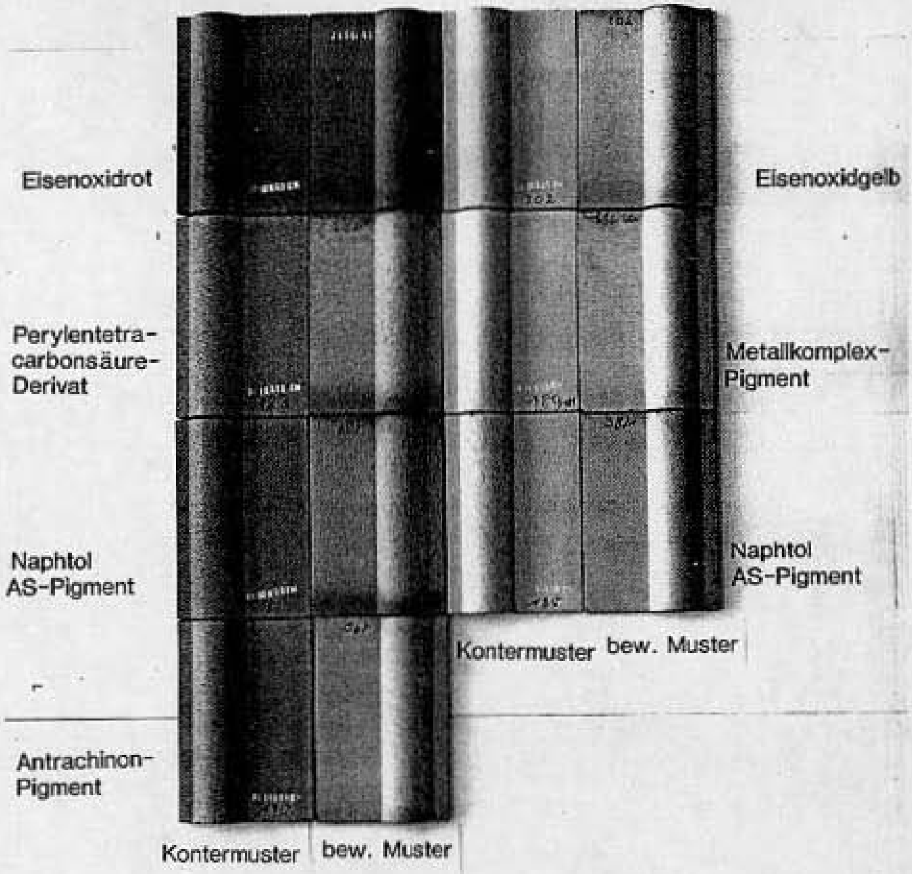
BAYER

The most important oxide pigments (weathered)

PK-…
2…

Table 1

Weathering of various pigments
12 months'
outdoor weathering at Krefeld



Kontermuster = control sample
bew. Muster = weathered sample

Fig. 1: Weather stability of iron oxide and organic pigments in concrete

Fig. 2: Weathering station at Krefeld (concrete roof tiles)

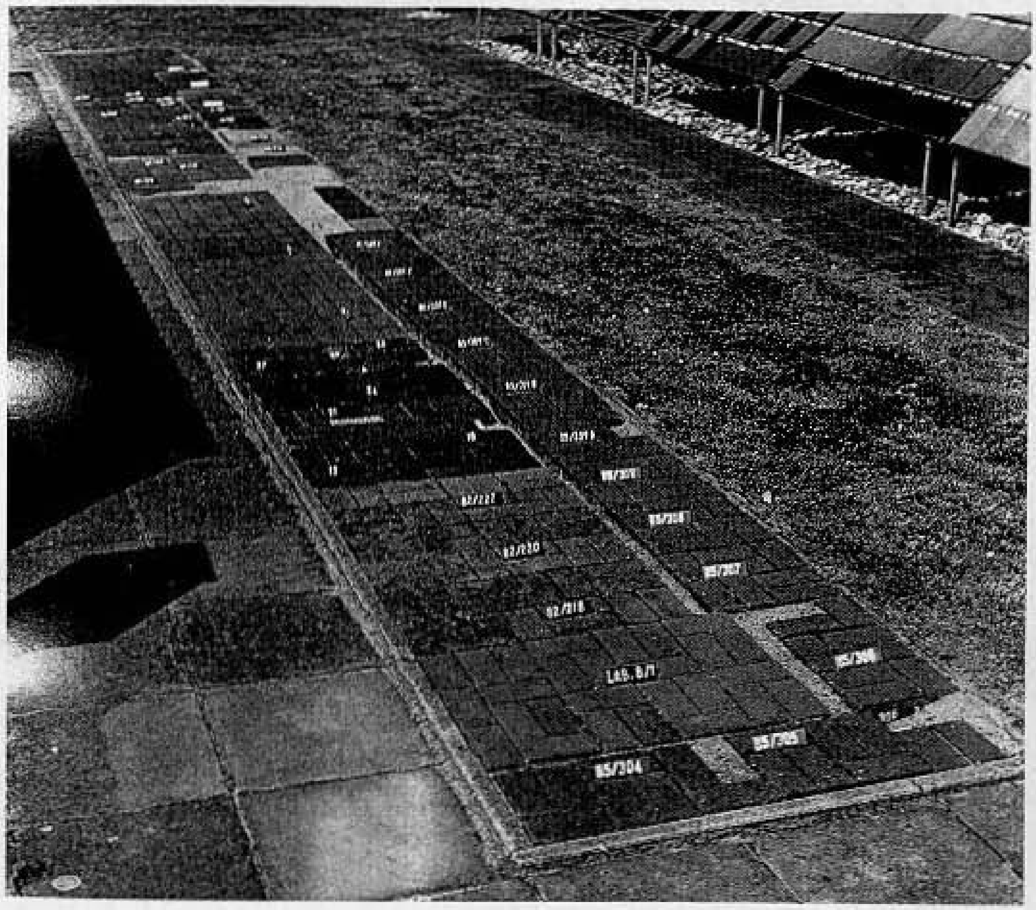


Fig. 3: Weathering station at Krefeld (concrete pavers)

3D colour system

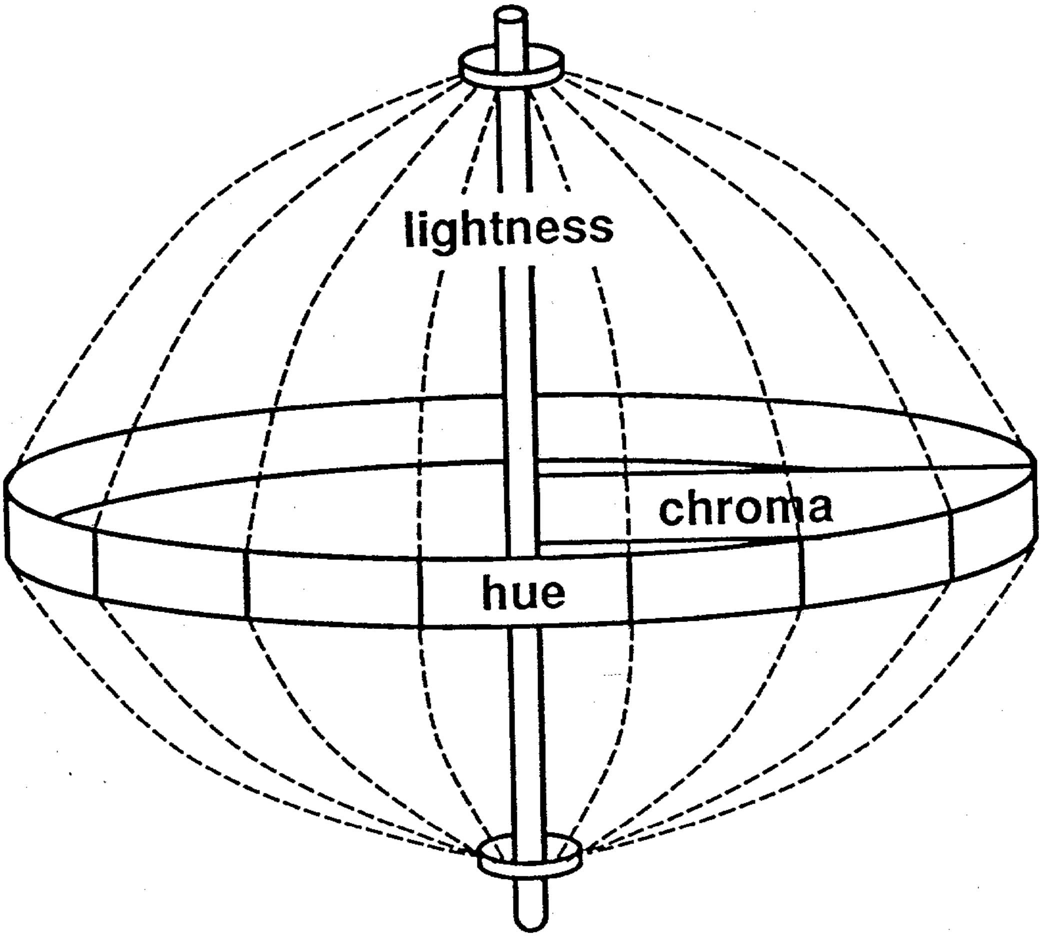


Fig. 4:

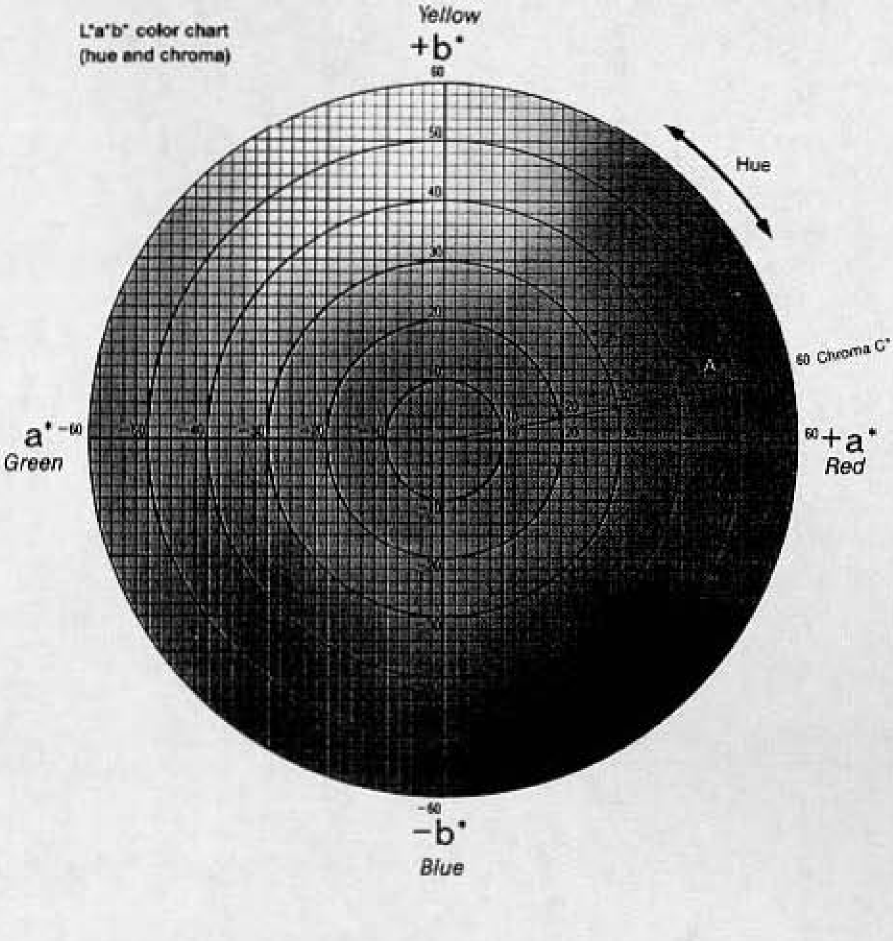


Fig. 5: Colour plane of uniform lightness L* in the CIELAB colour system

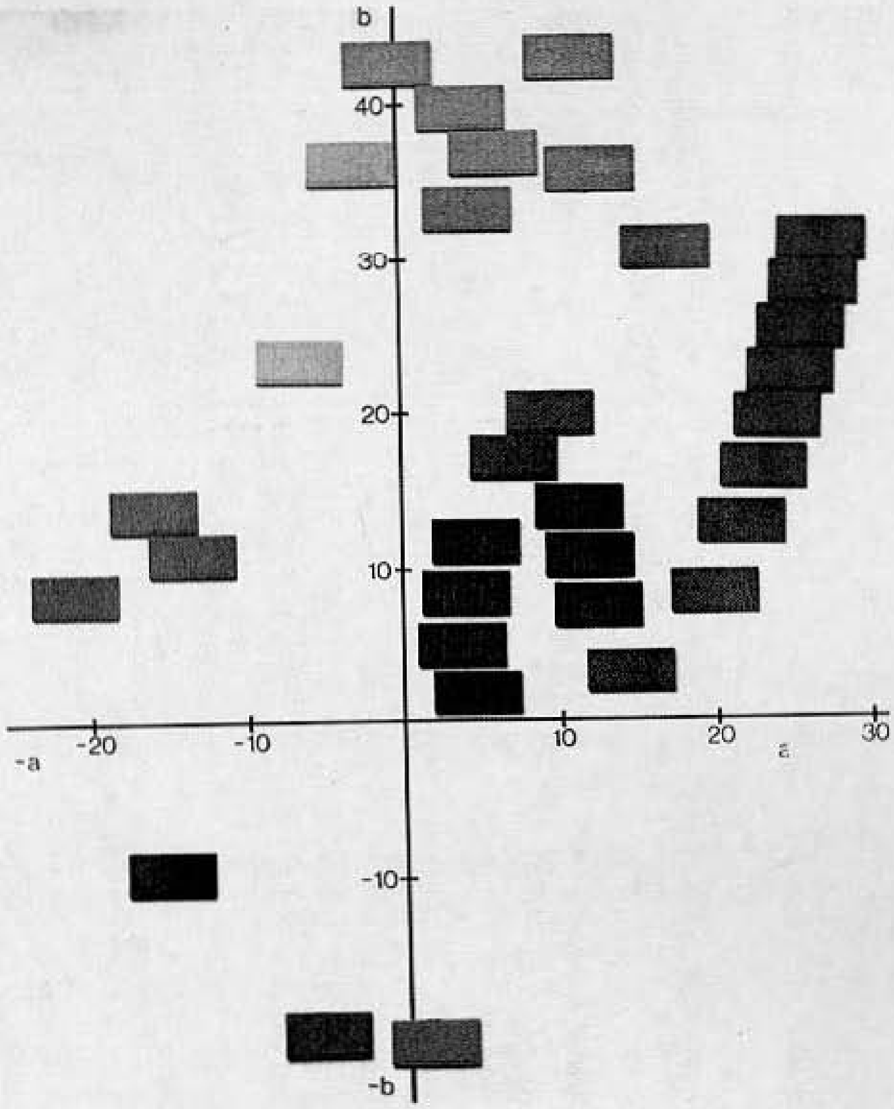
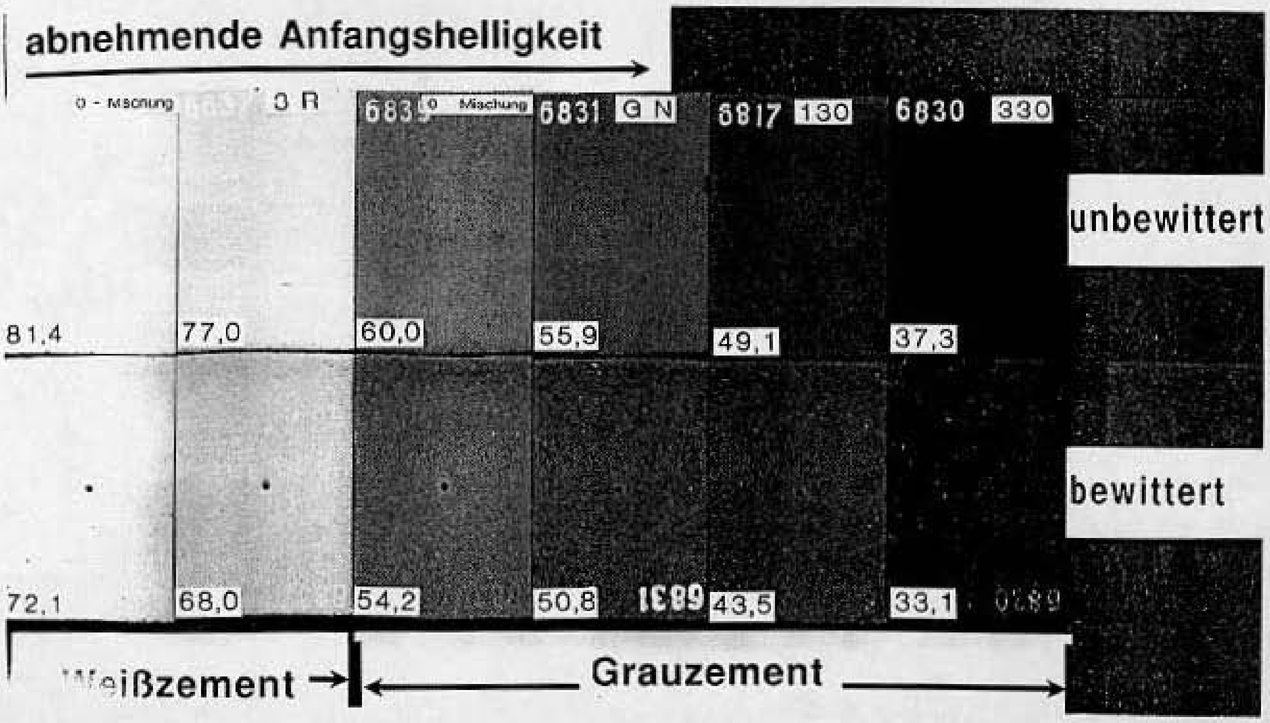
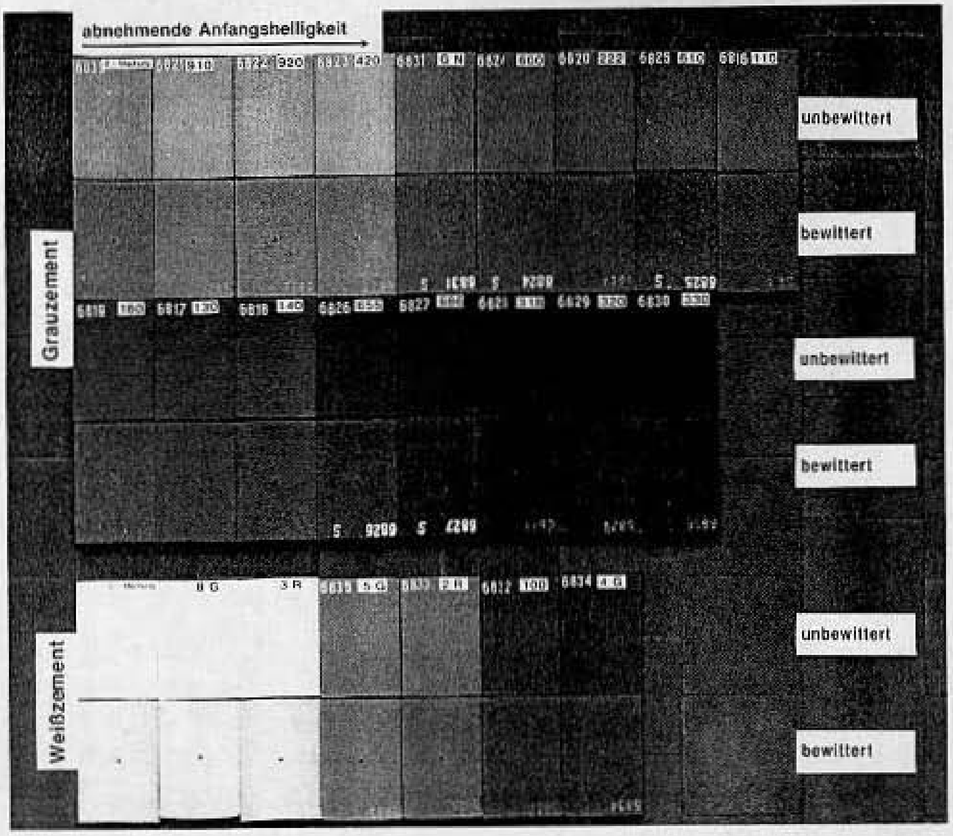


Fig. 6: Various pigmented concrete samples classified in the CIELAB colour system



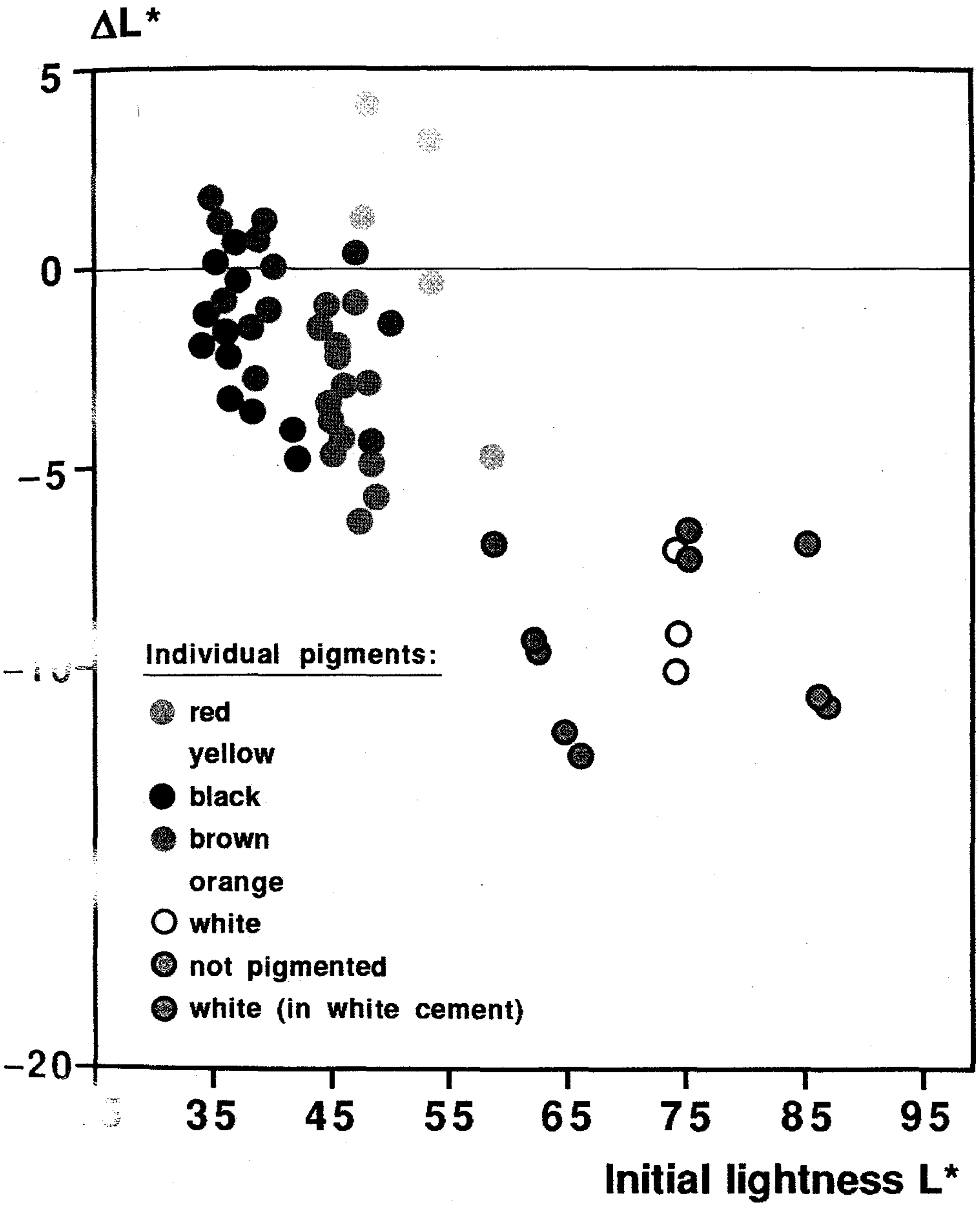
abnehmende Anfangshelligkeit = Decreasing initial lightness
unbewittert = unweathered
bewittert = weathered
Weißzement = White cement
Grauzement = Grey cement

Fig. 7: Change in lightness after 5 years' outdoor weathering in a marine climate



abnehmende Anfangshelligkeit = Decreasing initial lightness
unbewittert = unweathered
bewittert = weathered
Weißzement = White cement
Grauzement = Grey cement

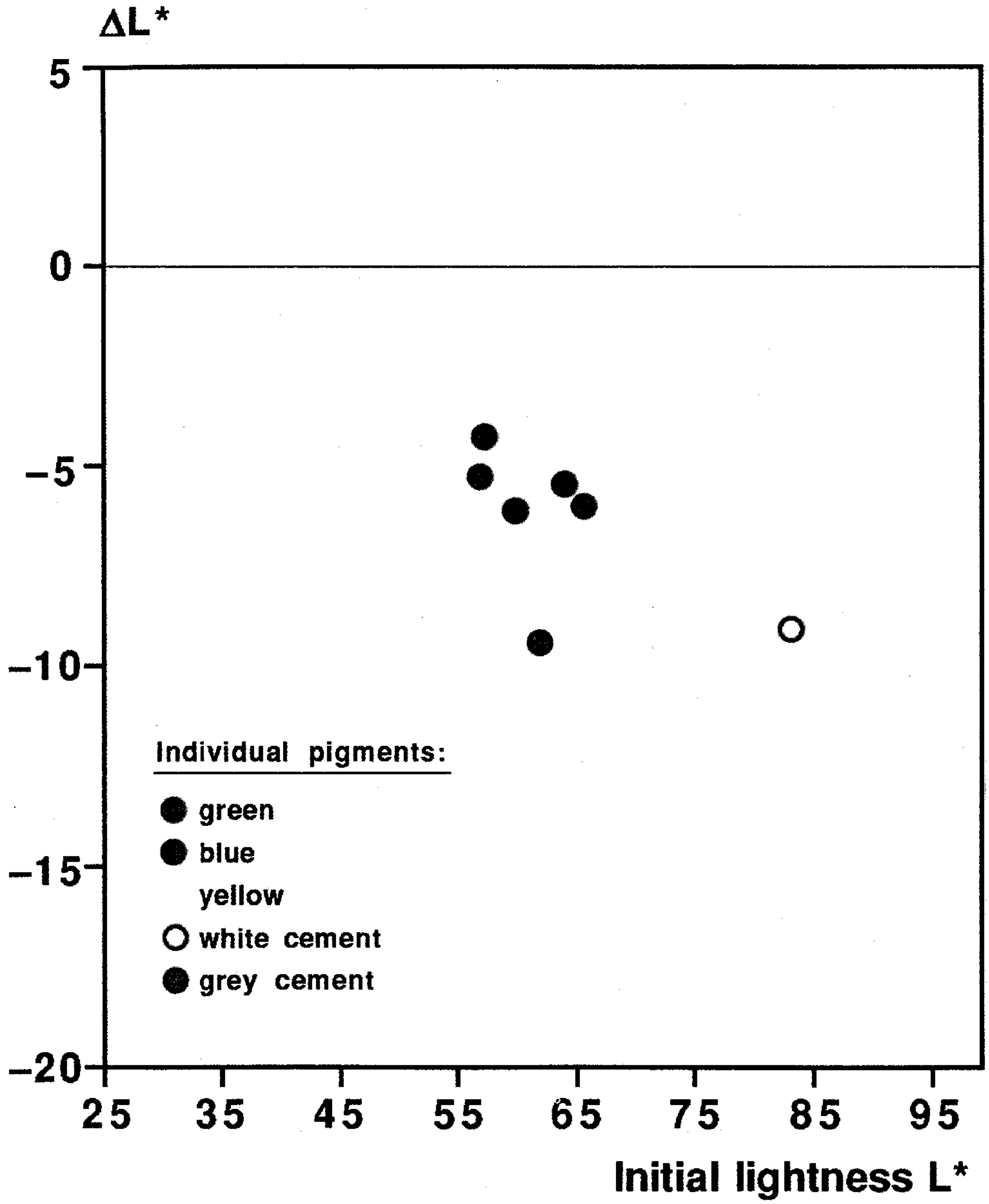
Fig. 8: Outdoor weathering (5 years) of inorganic colour pigments (marine climate)



Change of lightness during weathering as a function of initial lightness
(concrete roof tiles, grey cement)
(exposure: 3 years at Krefeld)

PK–A Fe
2/91

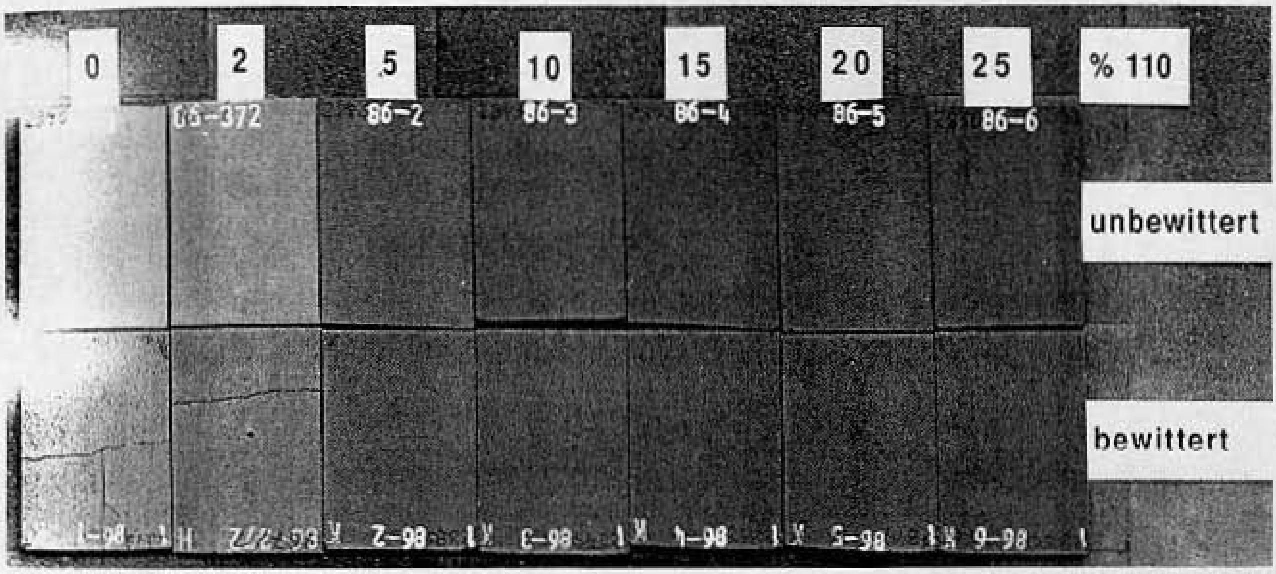
Fig. 9:



Change of lightness during weathering as a function of initial lightness
(concrete roof tiles, white cement)
(exposure: 3 years at Krefeld)

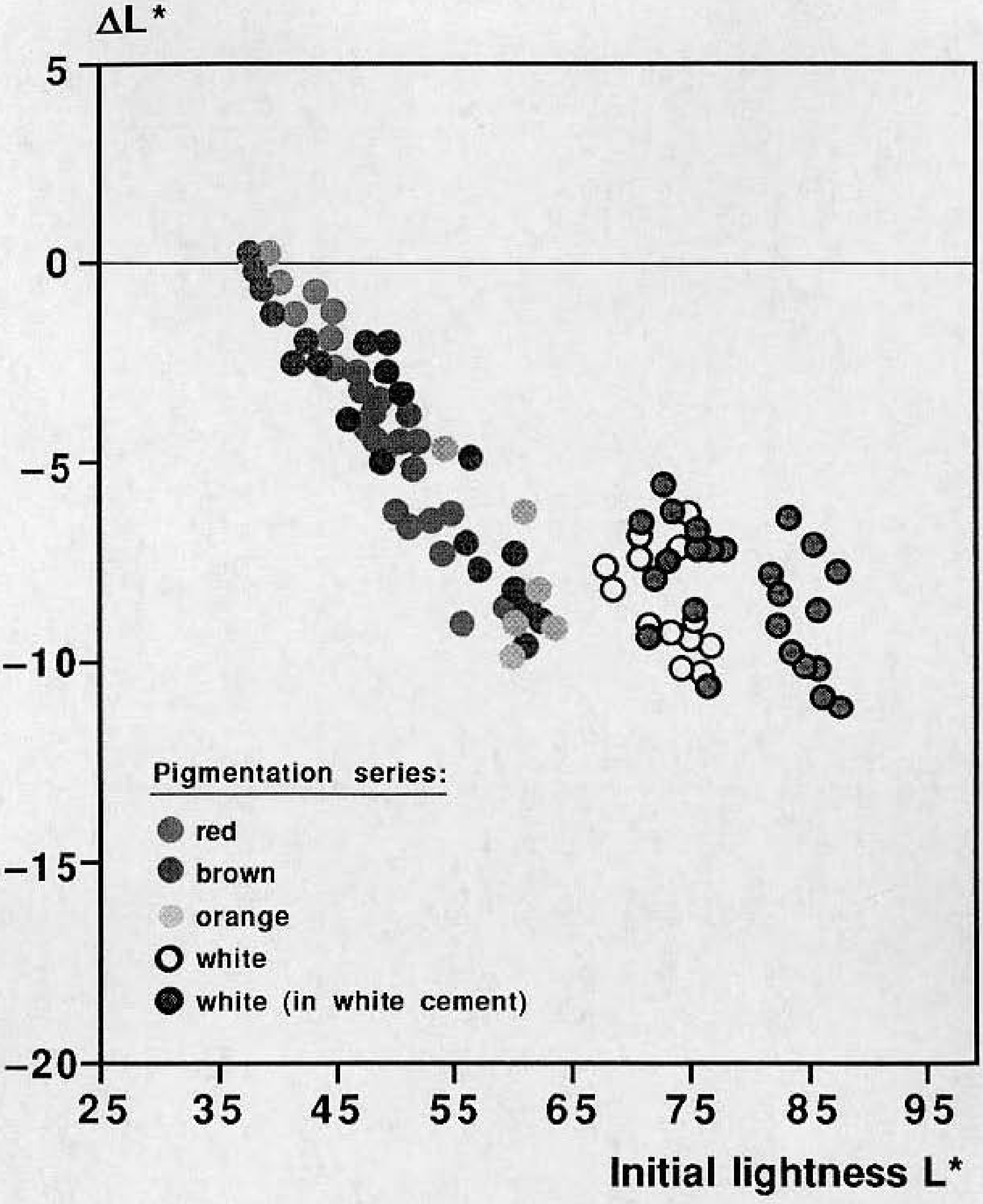
PK–A Fe
2/91

Fig. 10:



bewittert = unweathered
ewittert = weathered

Fig. 11: Pigment concentration and outdoor weathering (4 years' at Krefeld)



Change of lightness during weathering as a function of initial lightness

(concrete roof tiles, grey cement)
(exposure: 3 years at Krefeld)

PK–A Fe
2/91

Fig. 12:

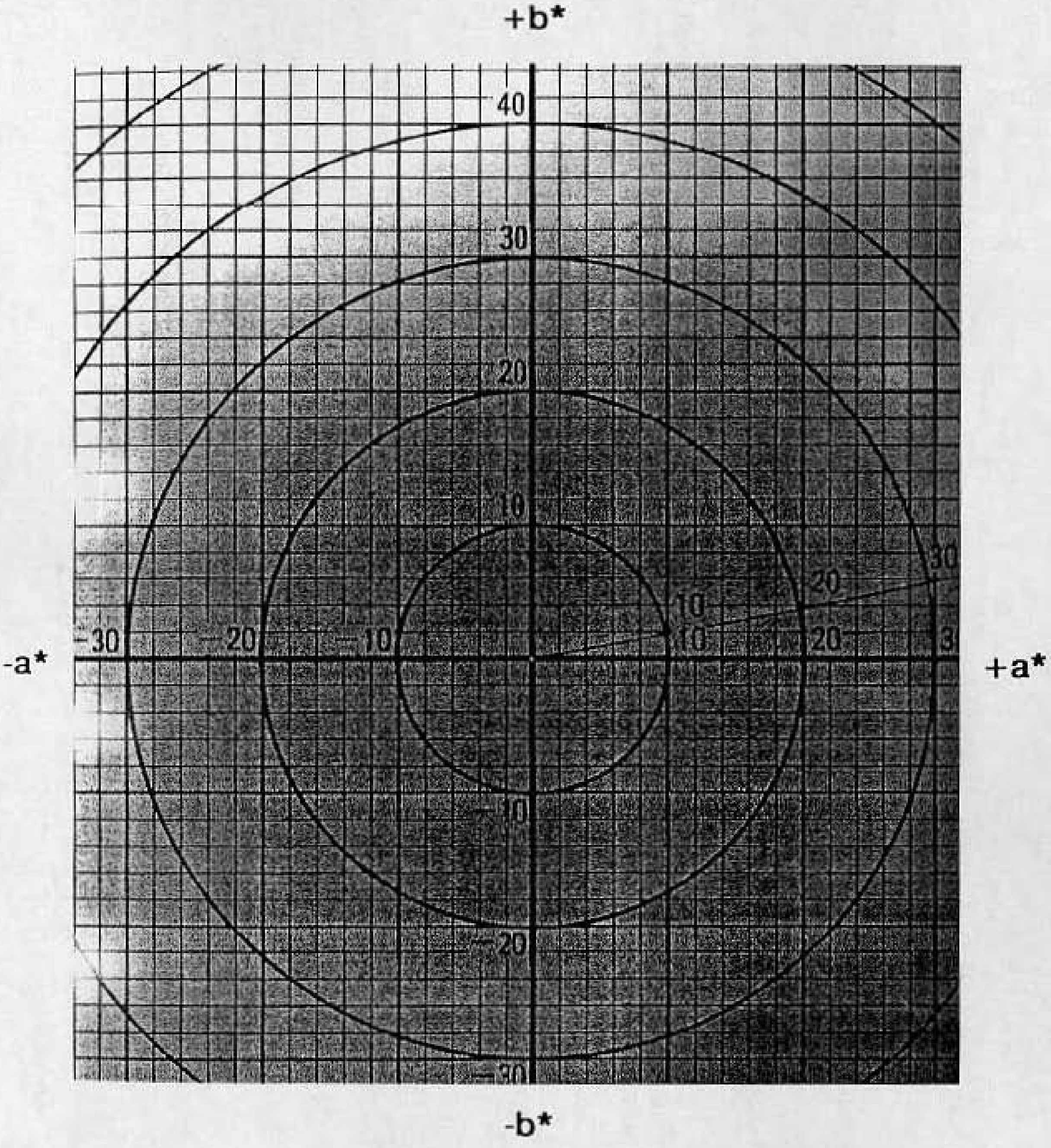


Fig. 13: Section of the colour plane shown in Fig. 5

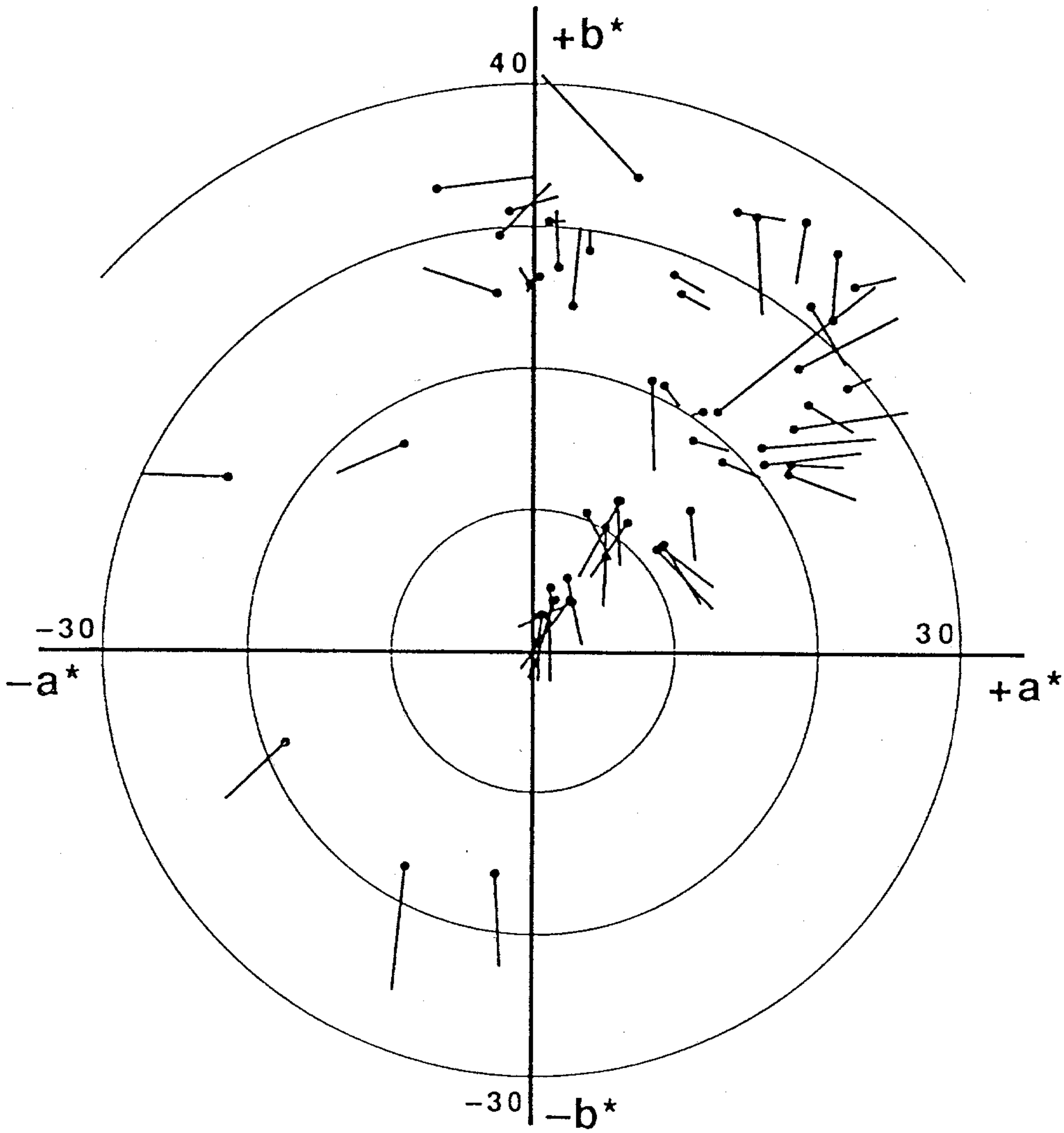


Fig. 14: Colour changes resulting from outdoor weathering (based on Fig. 13)

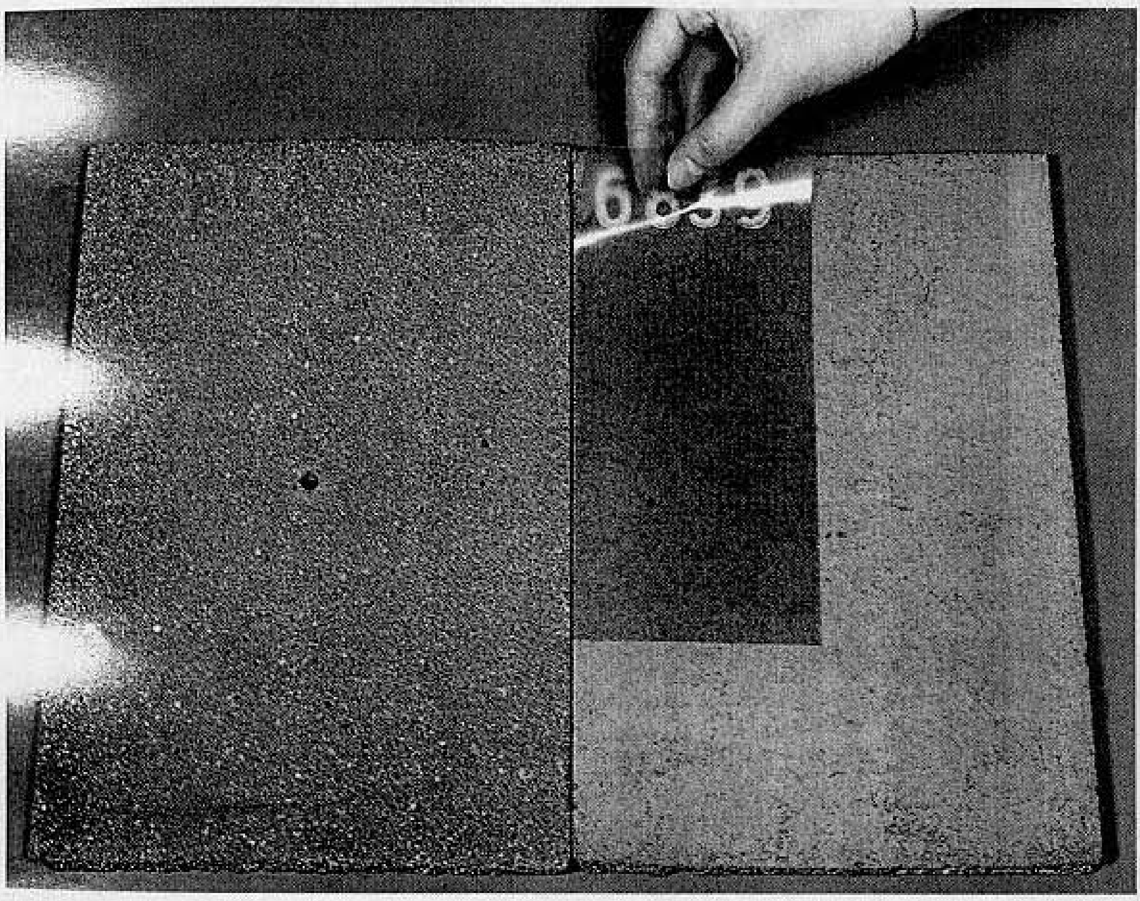


Fig. 15: Simulating the effect of outdoor weathering using a tinted, transparent film

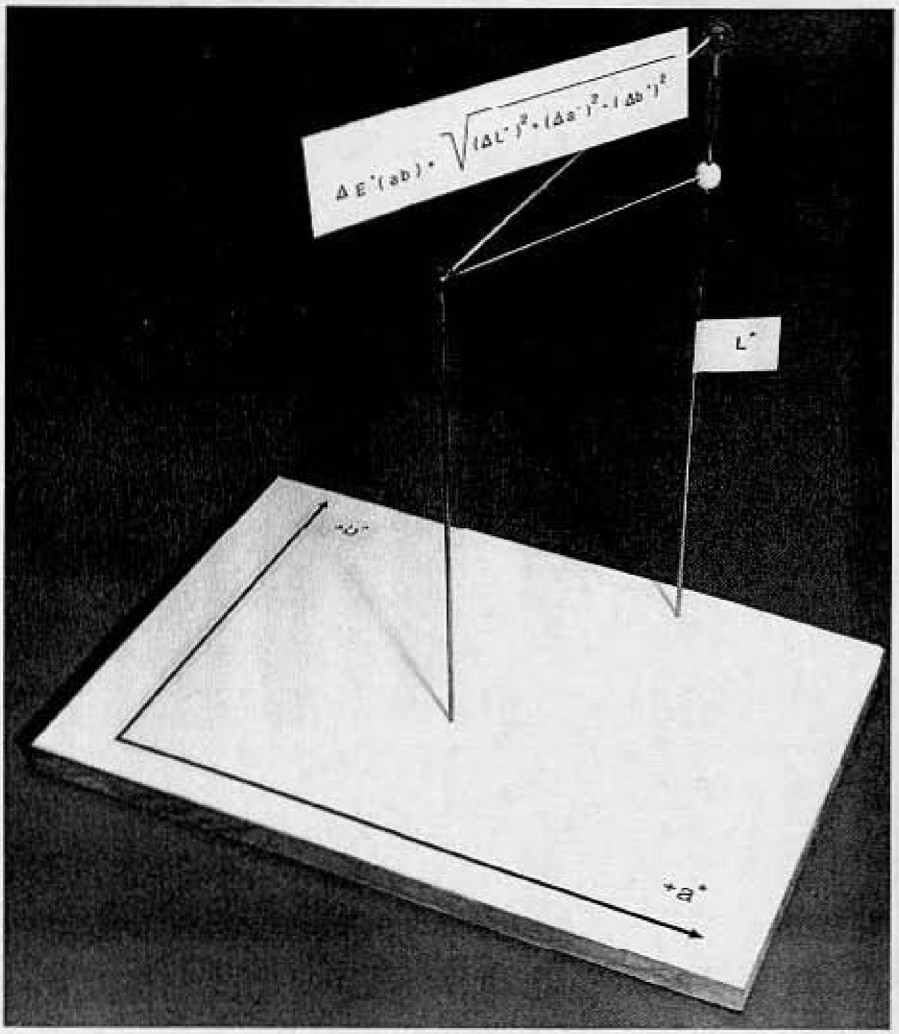


Fig. 16: Model showing the overall colour change $\Delta E^*(a,b)$

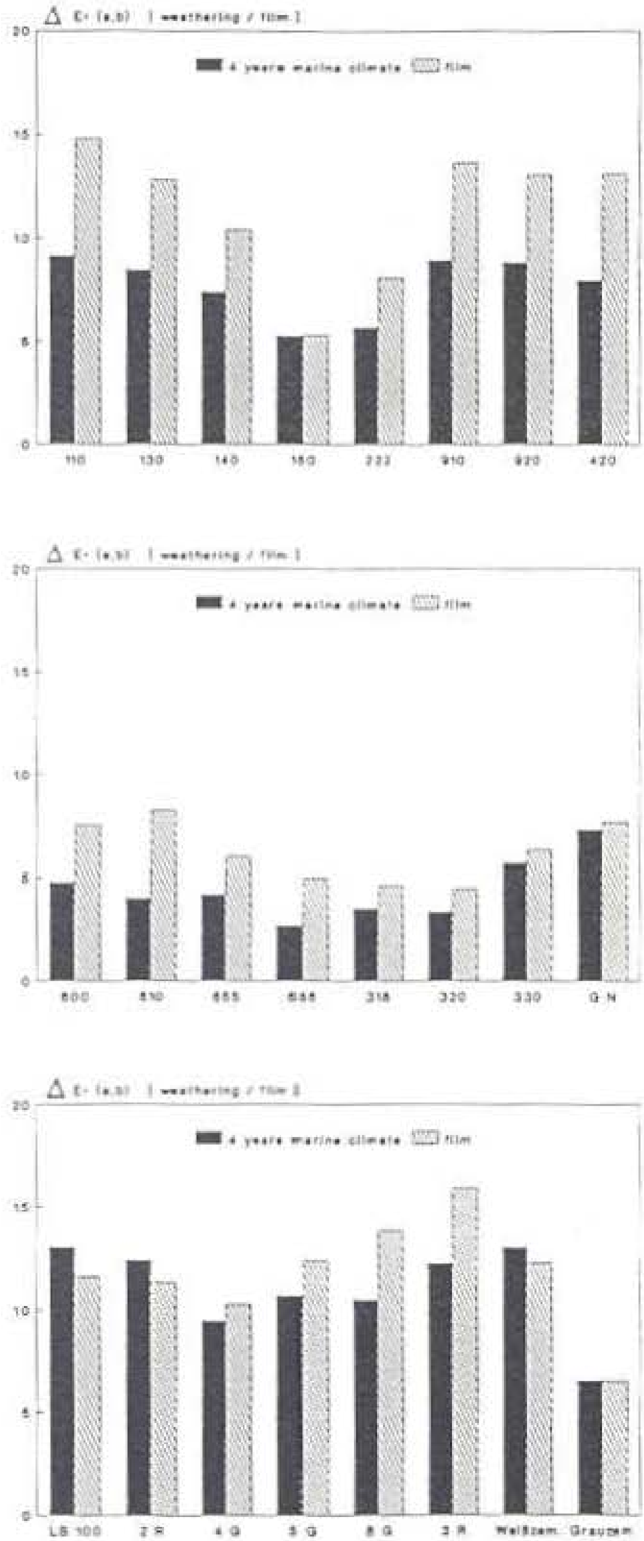


Fig. 17: Comparison of colour change $\Delta E^*(a,b)$ observed after outdoor weathering and measured through a tinted, transparent film