SO2

POSITION PAPER

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Note

This document reflects the opinions the majority of the members of the working group.

It should not be considered as an official statement of the position of the European Commission.

Not all experts necessarily share all the views expressed here after.

The remarks or comments expressed after the last meeting of the group and not agreed during it have not been considered in this final version.
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0.- Background

The present Council Directive of July 15, 1980 on Air Quality Limit Values and Guide Values for Sulphur Dioxide and Suspended Particulates and its amendment were adopted to protect human health and the environment against adverse effects from SO2 and Suspended Particulates. For this purpose, the Directive lays down limit values for SO2 and Suspended Particulates which are mandatory over all the territory of the Member States (these limit values are interrelated); in addition, long term guide values are also fixed. Member States are requested to:
- establish measuring stations at sites where pollution is thought to be greatest and where the measured concentrations are representative of local conditions;
- measure according to specified procedures using reference or equivalent methods;
- reduce pollution emissions so that concentrations comply with the limit value and in the long run, achieve the guide values;
- inform the Commission about breaches of the limit value(s) and to take abatement measures;
The Commission is monitoring the implementation of the Directive with the view of ensuring harmonized practices.

The last report of the Commission on the implementation of the Directive gives an overview of the information collected since the adoption of the Directive. The Council Directive on the Assessment and Management of Ambient Air Quality requires a review of the present Directive on SO2 and Suspended Particulates according to the principles which are laid down in this new Directive ("The framework Directive"). The Framework Directive governs the scope of this present paper (and of the subsequent Directive) which addresses only the potential for sulphur dioxide to cause harmful effects on human health and the environment. Other pollutants sometimes associated with sulphur dioxide, such as suspended particles or pollutants for which sulphur dioxide is a precursor, e.g. acid aerosol and sulfates, will be addressed elsewhere. In particular, the problem of acidification and critical loads will be handled by specific initiative. Likewise, the terms of the Framework Directive preclude consideration of the potential global climatic cooling effects linked to SO2.

---

1. 80/779/EEC, O.J. 1229, 30.08.1980, pp. 30-48
2. 89/427/EEC, O.J. L
5. Acidification - Working paper from the staff of the Commission
1.1 Sulphur compounds in the air

At ambient temperature and pressure, sulphur dioxide is a colorless gas consisting of one atom of sulphur and two atoms of oxygen.

In the past (late nineteenth century and first half of the present century) sulphur dioxide in combination with sooty particles was responsible for smog episodes in industrial cities.

1.2 Sources, sinks and chemistry of SO2

Man made sulphur dioxide results from the combustion of sulphur-containing fossil fuels (principally coal and heavy oils) and the smelting of sulphur containing ores

Over the past 25 years, there has been a tendency towards declining emissions in most Member States, due to changes in the types or amounts of fuels consumed and emission control measures; in addition and more importantly, the pattern of the sources has changed away from small multiple sources (domestic, commercial, industrial) towards large single sources emitting SO2 from tall stacks.

Volcanoes and oceans are the major natural sources of sulphur dioxide. In 1993, these sources were estimated to contribute only around 2 % of the total emissions of sulphur dioxide in the EMEP area6.

After being released in the atmosphere, sulphur dioxide is further oxidized to sulfate and sulfuric acid forming an aerosol often associated with other pollutants in droplets or solid particles extending over a wide range of sizes.

SO2 and its oxidation products are removed from the atmosphere by wet and dry deposition.

In spite of these processes of transformation and removal, sulphur dioxide can be transported over large distances, causing transboundary pollution.

Nowadays, it is also recognized that sulfate (SO42-) aerosols play an important cooling role in the radiative climate of the earth through the phenomena of sunlight scattering in cloud free air and as cloud condensation nuclei.

---

6 EMEP-MSC-W Report 1/95
### 1.3 Trends in SO2 emissions

Tables hereafter give the inventories of SO2 emissions in the Member States and other countries for different years.

#### Table 1.3.1 - 1985 - Europe 12 (CORINAIR 85)\(^7\) (Emissions in 1000 t)

<table>
<thead>
<tr>
<th></th>
<th>Combustion</th>
<th>Refineries</th>
<th>Industrial Combustion</th>
<th>Industrial Processes</th>
<th>Road transport</th>
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<td>35</td>
<td>99</td>
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<td><strong>DK</strong></td>
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<td>4</td>
<td>61</td>
<td>16</td>
<td>11</td>
<td>334</td>
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<tr>
<td><strong>F</strong></td>
<td>610</td>
<td>224</td>
<td>444</td>
<td>105</td>
<td>99</td>
<td>1482</td>
</tr>
<tr>
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<td>145</td>
<td>416</td>
<td>149</td>
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<td>2316</td>
</tr>
<tr>
<td><strong>GR</strong></td>
<td>373</td>
<td>28</td>
<td>81</td>
<td>18</td>
<td>0</td>
<td>500</td>
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<td>55</td>
<td>2</td>
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<tr>
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<td>148</td>
<td>550</td>
<td>131</td>
<td>76</td>
<td>2090</td>
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<tr>
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<td>3767</td>
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<td>9032</td>
<td>895</td>
<td>2617</td>
<td>685</td>
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<td>13625</td>
</tr>
<tr>
<td><strong>%</strong></td>
<td>66%</td>
<td>7%</td>
<td>19%</td>
<td>5%</td>
<td>3%</td>
<td></td>
</tr>
</tbody>
</table>

#### Table 1.3.2 - 1990 - Europe 15 (CORINAIR 90)\(^8\) (Emissions in 1000 t)

<table>
<thead>
<tr>
<th></th>
<th>Public power</th>
<th>Combustion</th>
<th>Comb.</th>
<th>Product</th>
<th>Extr./dist.</th>
<th>Solvent</th>
<th>Road transport</th>
<th>Other mob.s./m.</th>
<th>Waste treat./disp.</th>
<th>Agric.</th>
<th>Nature</th>
<th>TOTAL</th>
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</tr>
<tr>
<td><strong>B</strong></td>
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<td>0.3</td>
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<td></td>
<td></td>
<td></td>
<td></td>
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<td>145.3</td>
<td>24.6</td>
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<tr>
<td><strong>G(e)</strong></td>
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<td>133.5</td>
<td>444.9</td>
<td>53.9</td>
<td>48.2</td>
<td>12</td>
<td>911.8</td>
<td></td>
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<td></td>
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<td><strong>I</strong></td>
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<td>0.6</td>
<td>177.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td><strong>L</strong></td>
<td>767.2</td>
<td>62</td>
<td>573.8</td>
<td>104.9</td>
<td>103</td>
<td>48.2</td>
<td>2253</td>
<td></td>
<td></td>
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<tr>
<td><strong>NL</strong></td>
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<td>4.1</td>
<td>43.3</td>
<td>73.6</td>
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<td>201.2</td>
<td></td>
<td></td>
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<tr>
<td><strong>P</strong></td>
<td>174.6</td>
<td>4.3</td>
<td>75.9</td>
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<td>13.8</td>
<td>3</td>
<td>282.7</td>
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<tr>
<td><strong>Sp</strong></td>
<td>1452.1</td>
<td>97.9</td>
<td>478.5</td>
<td>38</td>
<td>69.4</td>
<td>17</td>
<td>2205.7</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td><strong>Sw</strong></td>
<td>14.9</td>
<td>15.7</td>
<td>37.7</td>
<td>16.9</td>
<td>7</td>
<td>10.9</td>
<td>104.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>UK</strong></td>
<td>2729.1</td>
<td>208</td>
<td>702.5</td>
<td>18.5</td>
<td>63.1</td>
<td>65.5</td>
<td>3786.7</td>
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<tr>
<td><strong>EUR12</strong></td>
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<td>760.7</td>
<td>3061.4</td>
<td>599.8</td>
<td>44</td>
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<td></td>
<td></td>
<td>573</td>
<td>17058.3</td>
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</table>

\(^7\) CORINAIR - Inventory of the emissions of SO2, NOx, VOC in the E C in 1985 - EUR 13232

\(^8\) CORINAIR 90 : summary report nr 1 - EEA
Figure 1.3.1 : CORINAIR - Map of SO2 emissions

Table 1.3.3 - 1990 Emissions of Eastern European countries
<table>
<thead>
<tr>
<th>Country</th>
<th>TOTAL (1100 t)</th>
</tr>
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<tbody>
<tr>
<td>Bulgaria</td>
<td>2008,2</td>
</tr>
<tr>
<td>Czech republic</td>
<td>1862,7</td>
</tr>
<tr>
<td>Estonia</td>
<td>275,1</td>
</tr>
<tr>
<td>Hungary</td>
<td>905,3</td>
</tr>
<tr>
<td>Latvia</td>
<td>114,6</td>
</tr>
<tr>
<td>Lithuania</td>
<td>222,5</td>
</tr>
<tr>
<td>Poland</td>
<td>3273,1</td>
</tr>
<tr>
<td>Romania</td>
<td>1311,5</td>
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<tr>
<td>Slovak Republic</td>
<td>542,1</td>
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<tr>
<td>Slovenia</td>
<td>196,0</td>
</tr>
<tr>
<td>Tot East</td>
<td>10711,1</td>
</tr>
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</table>

Figure 1.3.2: Eastern European countries emissions by sector

Figure 1.3.3: Contribution from different parts of Europe to SO2 emissions
Table 1.3.4  - 1993 - Europe 15 / National Data (emissions in 1000 t)

<table>
<thead>
<tr>
<th></th>
<th>Public power</th>
<th>Combustion com., res. combust.</th>
<th>Industrial product. processes</th>
<th>Extr./distr. fossil fuels</th>
<th>Solvent use</th>
<th>Road transport</th>
<th>Other s./m.</th>
<th>Waste treat. disp.</th>
<th>Agric. Nature</th>
<th>TOTAL</th>
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</thead>
<tbody>
<tr>
<td>A</td>
<td>17.7</td>
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<td>76.6</td>
<td>34.6</td>
<td>15.9</td>
<td>1.6</td>
<td>2.1</td>
<td>3.4</td>
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<td>0</td>
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<td>15</td>
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<td>0.4</td>
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<tr>
<td>UK</td>
<td>2089</td>
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<td>59</td>
<td>55</td>
<td>4</td>
<td>10</td>
<td>3188</td>
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</table>

1.3.2 Trends and perspectives

These results confirm the trends in the decrease of SO2 emissions which have been reduced by an average of 20% between 1980 and 1990 in the Member States.

The emissions from the Eastern Europe are of the same order as those from EUR-15. They influence ambient sulphur dioxide levels in some Member States as a result of transboundary transport, and are likely to continue to do so in the future.

According to the results presented in the report on acidification, the foreseeable SO2 emissions will be reduced between 1990 and 2010 by 60% up to 91% in the European Community (according to the scenario retained) and by 29% up to 86 % in the other European countries.

1.4 Concentrations of SO2 in ambient air

1.4.0 Background level

Inland SO2 concentrations in remote areas away from any source (anthropogenic or biogenic) range up to 10 µg/m3 when measured on a 24 hour mean basis, but are less than 2 -μg/m3 when averaged over a year.

1.4.1 EU Data

From the report on the implementation of the Directive 80/779/EEC, it appears that the number of exceedances of the limit value (see 2.5 for details of limit values) has substantially decreased over the last ten years:

- 42 exceedances of the EU limit values have been reported for the reference period 1983/1984 (10 M-S)

- 5 exceedances were reported in 1990/1991 (12 M-S without the East Germany Länder); the inclusion of the new German Länder increase this figure to 26
This trend is confirmed by the data collected in the frame of the exchange of information on air quality (Decision 82/479/EEC): the mean of the annual mean concentrations has decreased from 55 g/m³ to 20 µg/m³ between 1978 and 1993.

With regard to the 98th percentile of the daily concentrations, the values have decreased from 200 µg/m³ to 60 µg/m³.

See figures 1.4.1 and 1.4.2.
On the basis of the data for the most recent 5 year available period (1988-1992), the current figures for the SO2 levels in the Member States (12 countries, excluding East German Länder) are in the following ranges:

10 - 45 g/m³ for the annual mean
40-140 µg/m³ for the 98th percentile of the daily means

No data for short term averaging time period are available from the European legislation

1.4.2 National data

The following table gives an overview of the highest concentrations recorded in the Member States in recent years in the most polluted areas, as well as typical 'representative annual means recorded in rural areas

Table 1.4.1 - Highest concentrations recorded in the most polluted areas (Concentrations in g/m³)

<table>
<thead>
<tr>
<th>Country</th>
<th>Urban / Industrial Stations</th>
<th>Rural Stations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Annual mean</td>
<td>98 Percentile (24 h)</td>
</tr>
<tr>
<td></td>
<td>93</td>
<td>94</td>
</tr>
<tr>
<td>A</td>
<td>10-25</td>
<td>9-30</td>
</tr>
<tr>
<td>B</td>
<td>20-64</td>
<td>20-53</td>
</tr>
<tr>
<td>DK</td>
<td>8.6-12</td>
<td>4.6-8.7</td>
</tr>
<tr>
<td>FI</td>
<td>3-10</td>
<td>3-10</td>
</tr>
<tr>
<td>FR</td>
<td>5-115</td>
<td>3-70</td>
</tr>
<tr>
<td>GR</td>
<td>3-10</td>
<td>3-10</td>
</tr>
<tr>
<td>IRL</td>
<td>5-115</td>
<td>3-70</td>
</tr>
<tr>
<td>P</td>
<td>6-11</td>
<td>8-11</td>
</tr>
<tr>
<td>Sp</td>
<td>30-54</td>
<td>29-53</td>
</tr>
<tr>
<td>Sw</td>
<td>9-7</td>
<td>9-7</td>
</tr>
</tbody>
</table>

* Half-hourly values

These results shows the range of variation of the SO2 concentrations. From one year to the other, these variations are mainly due to climatic conditions: temperature (influencing the emissions) and wind speed and direction, mixing height (strongly influencing the dispersion of pollutants).

High SO2 concentrations may be observed at various background stations: these levels are caused by different factors like transboundary transport or the direct influence of local point sources (power plant, industrial installation,...)
1.4.3 Data from other sources

The following figure gives the SO2 concentrations recorded in selected EMEP stations.

Figure 1.4.3 - SO2 concentrations in EMEP Stations
(mean over 4 winter months of maximum daily concentrations)
2. Risk assessment

2.1 Human health effects and risks

Sulphur dioxide is an irritant when inhaled and high concentrations may cause breathing difficulties in people exposed to it. People suffering from asthma and chronic lung disease may be especially susceptible to the adverse effects of sulphur dioxide and, within the range of concentrations that occur during the more extreme pollution episodes, it may provoke attacks of asthma.

The effects on health of sulphur dioxide concentrations to which we may be exposed in the ambient air have been studied in a number of different ways.

2.1.1 Short term exposure

On the basis of the present knowledge, it appears that responses to an exposure to SO2 occur very rapidly (within the first few minutes from commencement of inhalation); continuing the exposure further does not increase the effects.

The observed effects of exposure to SO2 include a number of symptoms such as lung function impairment (decrease in FEV1). A wide range of sensitivity has been demonstrated, both among healthy people and among those with asthma, who form the most sensitive group.

The re-evaluation made by WHO (Europe)\(^9\) on this to human health of SO2, concludes among other that:

- it is difficult to draw a consistent picture of exposure-response relationship;
- the minimum concentration evoking changes in lung function, in exercising asthmatics, is of the order of 400 ppb (1144 \(\mu\)g/m\(^3\)) but there is one example of small changes in airways resistance in two sensitive subjects at 100 ppb (286 \(\mu\)g/m\(^3\)).

WHO, on basis of the findings from experimental studies, has recommended a guideline for short term exposure (of 500 \(\mu\)g/m\(^3\) over 10 minutes)\(^{10}\); however, because relationships between 10 minutes and hourly mean vary according to the nature of local sources, no specific hourly means has been proposed.

---

\(^9\) WHO - European Center for Environment and Health - Update and revision of the WHO Air Quality Guidelines for Europe - Volume 6 Classical - 1996

\(^{10}\) This recommendation includes an uncertainty factor of 2 applied to the lowest observed adverse effect level
<table>
<thead>
<tr>
<th>Value (µg/m³)</th>
<th>Population Affected</th>
<th>Associated effects</th>
<th>Type of study</th>
</tr>
</thead>
<tbody>
<tr>
<td>572</td>
<td>Heavy exercising Asthmatics</td>
<td>Some symptoms</td>
<td>Chamber</td>
</tr>
<tr>
<td></td>
<td></td>
<td>No change in lung function</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Heavy exercising Normal subjects</td>
<td>No change / No response</td>
<td></td>
</tr>
<tr>
<td>1430</td>
<td>Resting Asthmatics</td>
<td>No change / No response</td>
<td></td>
</tr>
<tr>
<td>1148</td>
<td>Heavy exercising Asthmatics</td>
<td>Small change in lung function</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>No change / No response</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Heavy exercising Normal subjects</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2860</td>
<td>Normal subjects</td>
<td>Small increase in sRaw</td>
<td></td>
</tr>
</tbody>
</table>

### 2.1.2 Exposure over 24-hours periods

In 1987, on the basis of epidemiological studies where day-to-day changes in mortality, morbidity or lung function related to 24-h average concentrations of SO2 in the presence of particulate matter and other associated pollutants were analyzed, WHO (Europe) has recommended a guideline on a 24-h averaging period; the proposed figure of 125 µg/m³ includes an uncertainty factor of 2 applied to the lowest-observed-adverse-effect-level.

In the revision of its guidelines, WHO (Europe) has considered the results of more recent studies which have consistently demonstrated effects on mortality and hospital emergency admissions for total respiratory causes at lower levels of exposure. A specific guideline is not recommended by WHO (Europe) at the present stage, but it is believe that the levels will have to be set at a value lower than the 1987 guideline when more results accumulate.

One recent study\(^{11}\) points to rising cases of asthma with declining SO2 ambient concentrations; time series analysis presented in this study consistently show no association with the timing of asthma attacks, even when peak (hourly maximum) exposures are considered. However a second new study\(^{12}\) indicates that an increase in excess mortality could be linked to changes in SO2 concentrations; it also suggests that effects could appear for daily concentrations below 125 µg/m³.

With regard to the latter, only the results from individual studies are currently available; the 'meta-analysis' which is needed to confirm and precise these results is not ready. It has therefore not been possible to take these results into account in this paper.

---

\(^{11}\) Committee on the Medical Effects of Air Pollutants “Asthma and outdoor Air Pollution” London HMSO - 1995, p. 146

\(^{12}\) APHEA Project
2.1.3 Long-term exposure

In 1987 WHO (Europe) has also set a guideline value as an annual average of 50 µg/m³ to protect against long-term effects of SO₂ on health (this value derived along the same methods as for the 24-h exposure) also include an uncertainty factor of 2. On the same basis as for the 24 h, WHO (Europe) is of the opinion that no specific guideline can be recommended at this stage, but it is believed that the guideline level has to be set at a value lower than 50 µg/m³ for annual exposure.

2.2 Environment effects and risks

2.2.1 Effects

SO₂ directly affects vegetation by uptake through parts of the plants that are above the ground; the direct effects on leaves are mainly determined by air concentrations. Depending on the amount of SO₂ taken up per unit of time, various kind of biochemical and physiological effects take place in the plant tissue; these include the degradation of chlorophyll, reduced photosynthesis, raised respiration rates, and changes in protein metabolism. The lower plants such as lichens and mosses, due to their structure have a particular sensitivity to SO₂.

The decisive factors in the action of SO₂ on plants are existing stresses on the plant, the concentration of SO₂, the duration of exposure, and the frequency and sequence of impact; within certain ranges of concentration and for a given dose (concentration times exposure duration), the extend of foliar injury increases with increasing concentration.

The significance of very low concentrations of SO₂ on growth and yield, and on changing plant sensitivity to other environmental stresses is now also recognized.

Some plants can also recover in pollution-free periods if the duration of exposure to injuring concentrations is not too long and if the pollution-free period is sufficiently long.

Individual species and varieties, and individuals within a population, react with different degrees of sensitivity to stress resulting from air pollution.

Sulfur is also an essential plant nutrient. In certain areas, where soils are deficient in sulfur (mainly calcareous-based on chalk and limestone), atmospheric sulfur may be taken up by leaves of some species and help contributing to the plant vitality But uptake is low and therefore not relevant to the setting of limit values.

Due to falling emissions of SO₂ in many areas in Europe and to the recognition of O₃ and nitrogen compounds as being of much greater significance with regard to plant injury, the relative importance of SO₂ as a phytotoxic pollutant has diminished to a certain extent. Nevertheless SO₂ can locally play a role in vegetation damage, especially in combination with other pollutants.

The results of field observations and fumigation experiments have been used to determine quantitative dose-response relationship between SO₂ concentrations and
the effects on both annual and perennial plants and to derive guidelines accordingly. These guidelines are generally defined as annual and/or winter means.

WHO no longer advocates using a 24 h guide value in their update of the Air Quality Guidelines in view of evidence confirming that peak concentrations are not significant compared with accumulated dose.

<table>
<thead>
<tr>
<th>Annual and winter mean value (µg/m³)</th>
<th>Target Affected</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>Crops</td>
</tr>
<tr>
<td>20</td>
<td>Forests / Nat. Veget.</td>
</tr>
<tr>
<td>15</td>
<td>Sensitive forests / Nat. Veget.</td>
</tr>
<tr>
<td>10</td>
<td>Lichens</td>
</tr>
</tbody>
</table>

### 2.2.2 Exposure - National data

#### Austria

<table>
<thead>
<tr>
<th>Value (µg/m³)</th>
<th>Time period</th>
<th>Surface of ecosystems exposed to concentr. above the values</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>Year</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>0</td>
</tr>
<tr>
<td>30</td>
<td>Year</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>4 %</td>
</tr>
<tr>
<td>20</td>
<td>Year</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>15 %</td>
</tr>
<tr>
<td>15</td>
<td>Year</td>
<td>1%</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>35 %</td>
</tr>
<tr>
<td>10</td>
<td>Year</td>
<td>15 %</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>45 %</td>
</tr>
</tbody>
</table>

#### Netherlands

<table>
<thead>
<tr>
<th>Value (µg/m³)</th>
<th>Time period</th>
<th>Surface of ecosystems (km²) exposed to concentr. above the values (90; 91;92;93;94;95;2000)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>Year</td>
<td>0; 0; 0; 0; 0; 0; 0; 0; 0; 0</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>0; 0; 0; 0; 0; 0; 0; 0; 0; 0</td>
</tr>
<tr>
<td>30</td>
<td>Year</td>
<td>0; 0; 0; 0; 0; 0; 0; 0; 0; 0</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>50; 0; 0; 0; 0; 0; 0; 0; 0; 0</td>
</tr>
<tr>
<td>20</td>
<td>Year</td>
<td>930; 1100; 230; 75; 0; 0; -</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>4600; 3200; 2100; 130; 0; -; -</td>
</tr>
<tr>
<td>15</td>
<td>Year</td>
<td>4900; 6100; 3600; 2700; 0; 0; -</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>11500; 8700; 6300; 2700; 0; -; -</td>
</tr>
<tr>
<td>10</td>
<td>Year</td>
<td>15600; 19600; 12000; 8700; 6600; 4200; 1200</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>33000; 18600; 17600; 14000; 6800; -; -</td>
</tr>
</tbody>
</table>

### 2.3 Effects on materials and cultural heritage

Deterioration of materials and objects of cultural heritage is a process which occurs at a rate which is determined by meteorological parameters such as...
relative humidity, temperature and precipitation, and by air pollutants. Since the time of wetness and temperature exhibit only small variations in the temperate climatic zone, the concentration of atmospheric pollutants is often the dominant variable affecting the rate of corrosion. Among the anthropogenic air pollutants, SO2 can be considered as the most important in deterioration of several materials. Many materials are affected; among them, e.g. stones used in historic and cultural monuments, which have resisted atmospheric attacks for hundreds or even thousands of years. But during recent decades, an accelerated degradation of their surface has been observed in many parts of Europe.

There are several ways how SO2 emissions can contribute to corrosion of materials: it deposits readily on surfaces and is then subsequently converted to sulfates; in ambient air, SO2 is also partly converted to sulfate particulates which may be deposited on surfaces and can also cause corrosion. Both SO2 and sulfate particulates may also dissolve in rain droplets and increase the acidity of precipitations thus enhancing the phenomena of corrosion.

The decisive effect of SO2 on corrosion of several materials like metals, calcareous stones, or stained medieval glass windows has been shown in several laboratory and field exposures. In the last years, however, a synergistic corrosive effect of SO2 and NO2 and later of SO2 and O3 has been discovered first in laboratory exposure; this has been confirmed later on by field exposure studies. They enhance the corrosive effect of SO2 by promoting its oxidation to sulphate. This underlines the necessity to treat the deterioration of materials taking into account the interrelated role of SO2, NO2 and O3 in a multi-pollutant situation.

During the last decades, several field exposure programs have greatly contributed to enhancement of the present state of knowledge on the effects of acidifying air pollutants on materials.

Field studies have shown that the dry deposition has, in most cases, the dominating effect and that SO2 exerts the strongest corrosive effect both in unsheltered end sheltered exposure. The effect of wet deposition is demonstrated only for unsheltered exposure.

In practical and economic terms, the corrosion due to SO2 is closely tied to densely populated areas. Here, three conditions coincide: a high content of atmospheric pollutants, a high population density and a large use of materials. The corrosion rate decreases in general rapidly with increasing distance from the source of emission. In many regions, atmospheric corrosion is therefore a local effect. In certain densely populated regions such as Western or Central Europe, an important part of corrosion damage can also be caused by the transport of pollutants over national borders. From a trend analysis undertaken in the frame the UN-ECE ICP, it appears that at numerous sites where the SO2 levels have decreased between 1987 and 1992, a pronounced decrease has been found in the corrosion rates.

Based on the findings of various studies, experts are recommending the following guidelines:
<table>
<thead>
<tr>
<th>Annual mean (µg/m³)</th>
<th>Type of material</th>
</tr>
</thead>
<tbody>
<tr>
<td>15</td>
<td>Zinc, weathering steel</td>
</tr>
<tr>
<td>10</td>
<td>Bronze, limestone, sandstone</td>
</tr>
</tbody>
</table>

### 2.4 WHO guidelines for maximum concentrations of SO₂ in ambient air

#### 2.4.1 Health

- **Long term exposure:** < 50 µg/m³ Annual mean
- **Daily exposure:** < 125 µg/m³ 24 h
- **Short period exposure:** 500 µg/m³ 10 minutes

#### 2.4.2 Ecosystems

- **Crops:** 30 µg/m³ Annual mean + Winter mean
- **Forests / Nat. veget.** 20 µg/m³ Annual mean + Winter mean
- **Forests / Nat. veget.** 15 µg/m³ Annual mean + Winter mean
  
  (for areas where the accumulated temperature sum above + 5°C is less than 1000° days per year)
- **Lichens** 10 -g/m³ Annual mean

### 2.5 WHO guidelines versus SO₂ concentrations

#### 2.5.1 Long-term exposure

*European scale*

On the request of DG XI, the WHO/ECEH in collaboration with the EEA TC on Air Quality has prepared an assessment of population exposure to sulphur dioxide and its health impacts in the EU countries.

In line with the exposure assessments carried out for other reports, only the exposure of the urban population has been estimated; an urban area being defined as a settlement (administrative area) with more than 50,000 inhabitants. The analysis is base on data available for the most recent year from 1989 onwards.

**Annual concentrations**

The cumulative distribution of the population by annual mean concentrations of SO₂ is given in figure 2.5.1. Approximately a quarter of the urban population of the EU living in 10% of the towns are exposed to values exceeding the WHO guideline for long term exposure to SO₂ (50 -g/m³)
In almost one third of the towns (20) for which daily concentrations are available, SO2 levels above 125 µg/m3 have been measured at least once during the relevant year. This would imply that almost half (46%) of population living in the towns covered by the Exchange of Information would be exposed to high levels of SO2 for at least one day in a year.

When considering a threshold value of 250 µg/m3, one fifth of the population living in 10% of the towns would be exposed to high levels of SO2 at least once a year.

In figure 2.5.2, the distribution of the population exposed to daily values is expressed as a percentage of person-days.

A summary of the exposure data is presented in table 2.5.1.
On the basis of the information available in the APIS data base, the following information is available: (the figures are for the 8 countries which have reported on SO2 concentrations in the context of the exchange of information)
A significant number of exceedances of 125 \text{\textmu}g/m^3 are observed in different categories of areas; most of the exceedances occur during the winter period. The average number of exceedances for the period considered is in the range of 9 to 13, with a maximum of 48.

**National data**

Percentages of population exposed to levels above daily thresholds

**Austria**

<table>
<thead>
<tr>
<th>Year</th>
<th>Conc &gt; 250 \text{\textmu}g/m^3</th>
<th>Conc. &gt; 125 \text{\textmu}g/m^3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1995</td>
<td>0</td>
<td>100 000 peop.</td>
</tr>
</tbody>
</table>

**Netherlands**

<table>
<thead>
<tr>
<th>Year</th>
<th>Conc &gt; 250 \text{\textmu}g/m^3</th>
<th>Conc. &gt; 125 \text{\textmu}g/m3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1990</td>
<td>0</td>
<td>0.6%</td>
</tr>
<tr>
<td>1991</td>
<td>0</td>
<td>23%</td>
</tr>
<tr>
<td>1992</td>
<td>0</td>
<td>3.5%</td>
</tr>
<tr>
<td>1993</td>
<td>0</td>
<td>1.5%</td>
</tr>
<tr>
<td>1994</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1995</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
2.5.2 Short-term exposure

Due to the lack of data, an assessment of the exceedances of the 10 minutes guideline is not possible at a European level (EUR15)

National data

Population (percentage of the population or number of people) exposed to levels above short time thresholds

Austria

<table>
<thead>
<tr>
<th>Year</th>
<th>Conc &gt; 1000 µg/m3</th>
<th>Conc. &gt; 500µg/m3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1995</td>
<td>0</td>
<td>A few hundred peop.</td>
</tr>
</tbody>
</table>

Netherlands

<table>
<thead>
<tr>
<th>Year</th>
<th>Conc &gt; 1000 µg/m3</th>
<th>Conc. &gt; 500µg/m3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1990</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1991</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1992</td>
<td>0</td>
<td>0.7</td>
</tr>
<tr>
<td>1993</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1994</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1995</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

UK

<table>
<thead>
<tr>
<th>Year</th>
<th>Conc &gt; 1000 µg/m3</th>
<th>Conc. &gt; 500µg/m3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1994</td>
<td>7 \times 10^6 peop.</td>
<td>Max : 25 \times 10^6 peop.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>99.9 percent. : 0.3 \times 10^6 peop.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>99.8 percent. : 0.1 \times 10^6 peop.</td>
</tr>
<tr>
<td>2005</td>
<td></td>
<td>Max : 7 \times 10^6 peop.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>99.9 percent. : 0.02 \times 10^6 peop.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>99.8 percent. : 0</td>
</tr>
</tbody>
</table>

2.6 Existing EU standards

The present limit values fixed in the Directive 80/779/EEC as amended by Directive 89/427/EEC are the following:
### 2.7 Existing standards in Member States and other countries

#### 2.7.1- Austria

**Limit values (\(^\text{g/m}^3\))**

<table>
<thead>
<tr>
<th>Zone</th>
<th>Year</th>
<th>Summer</th>
<th>Winter</th>
</tr>
</thead>
<tbody>
<tr>
<td>I (NÖ, T)</td>
<td>50</td>
<td>100</td>
<td>NÖ : Niederösterreich (Lower Austria)</td>
</tr>
<tr>
<td>II (NÖ, T)</td>
<td>200</td>
<td></td>
<td>OÖ : Oberösterreich (Upper Austria)</td>
</tr>
<tr>
<td>III</td>
<td>300</td>
<td></td>
<td>St : Steiermark (Styria)</td>
</tr>
<tr>
<td>OÖ, St</td>
<td>50</td>
<td>100</td>
<td>T : Tirol (Tyrol)</td>
</tr>
</tbody>
</table>

**24HM (daily mean value)**

- Zone I (NÖ, T): Year 70, Summer 150
- Zone II (NÖ, T): Year 200, Winter 300 (3/day up to 500)
- Zone II (St): Year 200, Winter 300 (3/day up to 400 in winter)
- Zone III: Year 300, Winter 300 (3/day up to 500)
- OÖ: Year 140, Winter 300

**HHM (half hourly mean value)**

- Zone I (NÖ, St, T): Year 70, Winter 150
- Zone II (NÖ, T): Year 200 (3/day up to 500)
- Zone II (St): Year 200 (3/day up to 400 in winter)
- Zone III: Year 300 (3/day up to 500)
- OÖ: Year 140, Winter 300

- 70 | 150 (97.5 percentile)
- 30 | 60 (monthly mean)
Alert thresholds

<table>
<thead>
<tr>
<th></th>
<th>3HM</th>
<th>Associates PM10</th>
</tr>
</thead>
<tbody>
<tr>
<td>(three hour mean)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pre warning</td>
<td>400</td>
<td>&lt; 200</td>
</tr>
<tr>
<td></td>
<td>600</td>
<td>&gt;200 (sum of SO2 and particles)</td>
</tr>
<tr>
<td>Warning level 1</td>
<td>600</td>
<td>&lt; 200</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>&gt;200 (sum of SO2 and particles)</td>
</tr>
<tr>
<td>Warning level 2</td>
<td>800</td>
<td>&lt; 200</td>
</tr>
<tr>
<td></td>
<td>1000</td>
<td>&gt;200 (sum of SO2 and particles)</td>
</tr>
</tbody>
</table>

Forests protection limit values

<table>
<thead>
<tr>
<th></th>
<th>Summer</th>
<th>Winter</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(7 months IV-X)</td>
<td>(5 months XI-III)</td>
</tr>
<tr>
<td>24HM</td>
<td>50</td>
<td>100</td>
</tr>
<tr>
<td>HHM</td>
<td>70</td>
<td>150 (97,5 percentile for each month)</td>
</tr>
</tbody>
</table>

No HHM value above 140 300

For deciduous forest:

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>24HM</td>
<td>100 (summer)</td>
</tr>
<tr>
<td>HHM</td>
<td>150 (per month, 97,5 percentile)</td>
</tr>
</tbody>
</table>

Guide values

Health protection:
- HHM: 200 µg/m3: up to 3 HHM per day between 200 and 500 µg/m3 allowed
- 24HM: 120 µg/m3

Protection of forests:
- HHM: 50 µg/m3
- 24HM: 30 µg/m3
- Growing season: 15 µg/m3

2.7.2- Finland

Limit values

Health protection:
- Annual median of 24-hour means: 80 µg/m3
- Annual 98-percentile of 24-hour means: 250 µg/m3

Guide values

Health protection:
- Monthly 99-percentile of 1-hour means: 250 µg/m3
- 2nd highest 24-hour value of the month: 80 µg/m3

Vegetation protection:
- Annual mean: 20 µg/m3
2.7.3- Germany

**Limit values**

- **Annual median:** 120 µg/m³
- **Winter median:** 180 µg/m³
- **98 percentile:** 250 µg/m³

**Alert thresholds**

- **Pre-alarm:** 600 µg/m³
- **Level 1:** 1200 µg/m³
- **Level 2:** 1800 µg/m³

2.7.4- Netherlands

The existing standards are the following:

**Limit values (µg/m³)**

- **1-h mean:** 830
- **24-h mean:** 500
- **50-percentile (24h mean):** 75
- **95-percentile (24h mean):** 200
- **98-percentile (24h mean):** 250

**Target values (µg/m³)**

- **50-percentile (24h mean):** 30
- **95-percentile (24h mean):** 80
- **98-percentile (24h mean):** 100

In order to achieve the environmental quality objectives for acidification (2010), and on the basis of existing knowledge, the Netherlands consider that the average annual concentration for SO2 should not exceed:

10 "g/m³ (with a margin of exceedance of 5 "g/m³)

for both rural and urban areas.
2.7.5- UK

EPAQS, an independent group of expert reporting to the Secretary of State for the Environment is providing recommendations for Air Quality Standards on a range of pollutants. In their report on SO2, published in 1995, they recommended a standard of

100 ppb measured over a 15 minutes averaging period

The UK government will respond to this recommendation shortly

The UK Department of the Environment reports sulphur dioxide levels in the media on basis of 1 hourly measurements described in four bands:

<table>
<thead>
<tr>
<th>Level</th>
<th>Concentration</th>
<th>Conversion Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Very good</td>
<td>&lt; 60 ppb</td>
<td>160 µg/m3</td>
</tr>
<tr>
<td>Good</td>
<td>60-124 ppb</td>
<td>161-335 µg/m3</td>
</tr>
<tr>
<td>Poor</td>
<td>125-399 ppb</td>
<td>336-1061 µg/m3</td>
</tr>
<tr>
<td>Very poor</td>
<td>&gt; 400 ppb</td>
<td>&gt; 1062 µg/m3</td>
</tr>
</tbody>
</table>

These break points are under review in the light of the revised WHO guidelines and the recent Expert Panel on Air Quality Standards (EPAQS) report

2.7.6- USA

- Primary standards

<table>
<thead>
<tr>
<th>Type</th>
<th>Concentration</th>
<th>Conversion Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Annual mean</td>
<td>30 ppb</td>
<td>86 g/m3</td>
</tr>
<tr>
<td>24 h average</td>
<td>140 ppb</td>
<td>400 g/m3</td>
</tr>
<tr>
<td>(non-overlapping)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1 h</td>
<td>400 ppb</td>
<td>1144 g/m3</td>
</tr>
<tr>
<td>5 minutes</td>
<td>600 ppb</td>
<td>1716 g/m3</td>
</tr>
</tbody>
</table>

- Secondary standards

<table>
<thead>
<tr>
<th>Type</th>
<th>Concentration</th>
<th>Conversion Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 h average</td>
<td>500 ppb</td>
<td>1430 g/m3</td>
</tr>
</tbody>
</table>

2.8 Values to be considered as starting points with the view of EU standards

2.8.0- Introductory remark

The values proposed here after are based solely on health / environment considerations. For the cost-benefits analysis which is developed in chapter 4, it is recommended to consider as well values twice higher than the values proposed here, and at a range of compliance levels (99.9%, the recommended level and 98.0%). [This paragraph will be revised after completion of chapter 4.]
2.8.1 Limit values

2.8.1.1 Health protection

Daily value

The relevance of APHEA (i.e. preliminary results only are presently available) was discussed in section 2.1.2. Based upon the WHO (Europe) guidelines and taking account of the preliminary results of APHEA, it is recommended to have the following limit value:

\[ 125 \text{ g/m}^3 \]

not to be exceeded more than 3 times a year.

This value will be reconsidered after 5 years on the basis of the new scientific findings.

It is proposed not to fix a margin of tolerance.

The proposal to allow excursion above the limit value for maximum 3 days per year is based on an objective assessment of a realistic maximum number of measurements which may be non-representative because of either:

(i) instrument malfunction
(ii) transient local sources making the sampling unrepresentative of anything but a very localized area close to the sample inlet

Short period value

The levels based on a short time averaging period are highly variable in space and time, for these reasons the assessment of exceedance will be very difficult in practice:

- on the monitoring side, a great number of stations will be needed in order to identify the exceedances; in addition, a large amount of data will be collected with inevitable logistical problems in handling, use and presentation
- for modelling, neither meteorological nor emission data are available for this averaging time.

In order to overcome this practical difficulty, it is proposed to fix a threshold for a longer time period (1 hour) derived from the 10 minutes guideline defined by WHO.
The conversion factor between the WHO 10-minute guideline and an equivalent hourly value varies from place to place and it is, therefore, not currently possible to objectively define a universal hourly limit value based on the WHO guideline (See discussion in annex I). Although there was not full consensus within the working group, it was decided to set the limit value for the hourly average, not to be exceeded more than 24 times a year, at 350 µg/m³. According to UK data, this is roughly equivalent to a 99.9% compliance with the WHO guideline.

Because of the uncertainties, it will be required to measure in parallel on a 10-minute and on a 1 hour basis at some representative stations close to sources, where people live, in order to be able to revise this ‘equivalence’ factor at the time of revision of the limit values (in 5 years). Member States should be asked to report exceedances of the 500 µg/m³ level averaged over 10 minutes as well as all exceedances of 350 µg/m³ (hourly average).

A margin of tolerance of 150 µg/m³ is proposed; the limit value should be reached within 5 years after the entry into force of the Directive.

2.8.1.2 Ecosystems

Because of the wide variation of ecosystems and their sensitivities within the EU, it is appropriate to set a basic limit value that is protective for all ecosystems and which would be needed in regions without very sensitive ecosystems. It was not attempted to set region-dependent limit values based on the local sensitivities. Consequently, the limit values given in the Directive can not be expected to give the necessary protection in every region within the EU. It is, instead, a ‘safety-net’ value designed to give protection to the majority of ecosystems within the EU Member States will therefore be encouraged to designate, where appropriate, zones or areas where valuable ecosystems need to be protected and where more stringent limit values, established by the Member State, will apply.

The following limit value is proposed:

20 µg/m³ (annual and winter mean)

not to be exceeded over the year or the winter season

A margin of tolerance of 10 µg/m³ is proposed;

the limit value should be reached within [5] years

2.8.1.3 Materials

The values proposed in this document as starting points for fixing Limit / Alert threshold Values are intended to protect health and ecosystems. To avoid damage to cultural heritage, these values may not be sufficient. Therefore the Member States will be encouraged to designate, where appropriate, zones or
areas where monuments need to be protected and where more stringent limit values, established by the Member State, will apply.

Based on the present knowledge, the following guidelines could be used for the fixation of limit values:

- 15 “g/m³ (annual mean) for zinc and weathering steel
- 10 “g/m³ (annual mean) for bronze, limestone, sandstone

It is remarked that the proposals given in the following chapters do not apply to possible additional, stricter limit values that Member States may set for the protection of special ecosystems or cultural heritage.

2.8.2 Alert threshold

The alert thresholds are aiming at:

- informing the population in cases of exceedances of levels harmful for health in order to:
  - take precautionary measures
  - be aware of the possible cause of special health problem(s)
- taking short term measures aiming at reducing the concentrations

The following elements have been taken into consideration for a recommendation for an alert threshold:

- in cases of episodes characterized by bad dispersion conditions, larger areas are concerned; the duration of the exceedance is several hours. In these cases, information of the public and short time actions may be appropriate;

- exceedances caused by point sources are localized areas and are characterized by high spatial inhomogeneity and variability in time. These exceedances are difficult to identify and the adequacy / relevance of alert measures is questionable. Because of this, the alert threshold should only pertain to cases when a significant number of the population is exposed; depending on the monitoring density, this could imply that exceedance should be measured at more than one station.

- although the Framework Directive does not oblige Member States to take measures when the alert value is exceeded, the term alert (and even more so the German term “Alarm”) suggests that action should be taken. The terms “information” or “warning” seem to be more appropriate to use in communicating to the public, but the term “alert” is fixed in the Framework Directive. Member States should be free in choosing the appropriate wording when informing the public.
On these basis, the following alert threshold is recommended

350 µg/m³ exceeded during 3 consecutive hours
3. Assessment of concentrations

3.0 Introduction

The concentrations have to be assessed over the whole area of the Member States. Prior to the entry into force of the Directive, a preliminary analysis should be made to determine the concentration distributions over the Member States to enable them to define, at the entry into force of the Directive, appropriate monitoring networks and other assessment techniques.

For the implementation of the directive, the use of several assessment techniques will be possible under minimum requirements regarding the number of measuring points, the type of measuring techniques and the mathematical techniques; these requirements depend on the ratio between the concentration and the limit value, and also on the limit value itself.

3.1 Assessment under the existing Directive 80/779/EEC

The Directive only requires measurement for determining the ambient concentrations.

The Directive requires that "Member States shall establish measuring stations ..., in particular in zones where the limit values ... are likely to be approached or exceeded ...; the stations must be located at sites where pollution is thought to be greatest and where the measured concentrations are representative of local conditions"

3.1.1- Monitoring stations

The information available to the Commission (GIRAFE Data Base) shows that:

+ there is a large variability between Member States both in numbers of monitoring stations and their siting;
+ there is no clear pattern in the siting of the stations.

The following table give some information about the siting of stations (1990).
Table 3.1.1

<table>
<thead>
<tr>
<th>Member State</th>
<th>Total Nb. of stat.</th>
<th>Number of stations reporting for Directive</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total</td>
<td>Mixed</td>
</tr>
<tr>
<td>Austria</td>
<td>200</td>
<td>195</td>
</tr>
<tr>
<td>Belgium</td>
<td>66</td>
<td>66</td>
</tr>
<tr>
<td>Denmark</td>
<td>29</td>
<td>18</td>
</tr>
<tr>
<td>Finland</td>
<td></td>
<td></td>
</tr>
<tr>
<td>France</td>
<td>232</td>
<td>229</td>
</tr>
<tr>
<td>Germany</td>
<td>556</td>
<td>424</td>
</tr>
<tr>
<td>Greece</td>
<td>30</td>
<td>29</td>
</tr>
<tr>
<td>Ireland</td>
<td>52</td>
<td></td>
</tr>
<tr>
<td>Italy</td>
<td>382</td>
<td>253</td>
</tr>
<tr>
<td>Luxembourg</td>
<td>5</td>
<td>1</td>
</tr>
<tr>
<td>Netherlands</td>
<td>123</td>
<td>104</td>
</tr>
<tr>
<td>Portugal</td>
<td>51</td>
<td>47</td>
</tr>
<tr>
<td>Spain</td>
<td>556</td>
<td>501</td>
</tr>
<tr>
<td>Sweden</td>
<td></td>
<td></td>
</tr>
<tr>
<td>UK</td>
<td>299</td>
<td>299</td>
</tr>
</tbody>
</table>

3.1.2 Measuring method

The Directive specifies a reference method to be used for its implementation; Member States could use this method or others which produced results which were either demonstrated to correlate satisfactorily or to show a reasonably stable relationship when measurements were made in parallel with those obtained using the reference method.

For the sampling, the reference method is the ISO-4219 method.

For the analysis, the reference method is the ISO-6767 (TCM /pararosanilene).

3.2 Basic principles resulting from the Directive on Ambient Air Quality Assessment and Management

3.2.1 Purposes of assessment

The assessment is aiming at

a) checking whether the limit values / alert thresholds are exceeded anywhere over the territory of Member States;

b) supporting the management of air quality where limit values/alert thresholds are exceeded;

c) making adequate information available to the public.
3.2.2- Targets addressed

Two targets are identified:

- the human health
- the ecosystems

3.2.3- Assessment methods

Article 6 of the Framework Directive gives prescriptions regarding the assessment methods to be applied. It stipulates that in "agglomerations" (which have a special status in the Framework Directive) measurements are always mandatory, and further it links assessment regimes to two levels below the limit value which serve as criteria to distinguish between these regimes. These two levels will be described hereafter as x% and y% of the limit value (see Figure 3.2.1)

![Figure 3.2.1 - Principle of the limit value - x & y percentages - Margin of tolerance](image)

It is important to note that exceedance of the limit value determines whether the air quality within a zone is in compliance or not, and does not differentiate between the assessment regimes prescribed. Conversely, exceedance of x% or y% determines which assessment regime is prescribed, while it has no implications for air quality management. Figures 3.2.2a and 3.2.2b illustrate this.
Figure 3.2.2a - Implication of exceedance of the limit value, x% and y% for compliance judgment and assessment requirements, respectively.

Figure 3.2.2b - Exceedance areas (relate to limit value) and high concentration areas (relate to x% and y% of the limit value).
Although zones can be regarded as the basic areas for air quality management in the Framework Directive, it would not be efficient to prescribe a uniform assessment regime, including the associated network density requirements, for an entire zone when x or y% of the limit value is exceeded in only a small area of the zone. It is more efficient to link the assessment regime to the areas where the exceedance of x and y% of the limit value takes place instead of zones. In the following these areas will be referred to as high concentration areas. Relating this to the requirements of the Framework Directive, one arrives at four types of areas, each with its own assessment regime (two types of high concentration areas and two types of areas where the levels are below y% of the limit value):
1. Areas where levels exceed x% of the limit values (regime 1 in Figure 3.2.2);
2. Areas where levels exceed y% of the limit values, but not x% of the limit values (regime 2);
3a. Areas in agglomerations where levels are below y% of the limit values (regime 3a);
3b. Areas in non-agglomeration zones where levels are below y% of the limit values (regime 3b).

Type 3a takes into account the requirement that measurement is mandatory in agglomerations. It allows to make a distinction between agglomeration where levels are near or higher than the limit values (included in type 1 and 2) and agglomerations where the concentrations are far below the limit values (type 3a). It is remarked that the requirement to measure in agglomerations where levels are far below the limit and alert values is questionable. It should be further considered to restrict the measurement regime 3a to agglomerations with levels < y% where the alert value is exceeded.

The Framework Directive gives several prescriptions regarding these four types. Table 3.2.1 indicates the assessment regimes associated with these types of areas.

<table>
<thead>
<tr>
<th>Area</th>
<th>Assessment regime, from the strictest (top) to the mildest (bottom) requirements</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Where levels &gt; x%</td>
<td>Based on continuous measurements (at least one site per zone), may be supplemented by modelling</td>
</tr>
<tr>
<td>2. Where levels &gt; y%</td>
<td>Combination of continuous measurement (at least one site per zone) and modelling allowed</td>
</tr>
<tr>
<td>3a. Where levels &lt; y%, in agglomerations</td>
<td>At least one continuous measuring site per agglomeration, combined with modelling, objective estimation, indicative measurements</td>
</tr>
<tr>
<td>3b. Where levels &lt; y%, in non-agglomeration zones</td>
<td>Modelling, objective estimation, indicative measurements</td>
</tr>
</tbody>
</table>
3.2.4 Assessment in space and time

3.2.4.1 General

This section first provides a general background on the temporal and spatial framework. After the general introduction, the temporal and spatial aspects of the assessment will be discussed in detail and specifications will be proposed. Because the definition of limit values in time and space is essential for the assessment strategy, it will be revisited and elaborated here.

**Difference between time and space: representativeness**

Concentrations vary in time and in space. The most important goal of the assessment is to provide a description of the *concentration distribution in time and space*, as complete and accurate as possible. Although time and space have in principle various aspects in common (see Table 3.2.2 below), a monitoring network deals very differently with time and space: stations usually measure at all times, but at very few places. In particular, the problem of *representativeness* of concentrations measured at a certain point is predominant for space, but hardly for time.

**A common framework for measuring and modelling**

In spite of their limited possibilities to measure the concentration distribution in space, monitoring networks are commonly used to characterize the pollution over the entire territory. The network results can provide a general picture of the pollution levels when a good measuring strategy is applied: by choosing representative sites the measuring results for a few spots can be used for the rest of the territory. *Macro-scale siting criteria* attempt to distribute stations over locations that are representative for large areas and *micro-scale siting criteria* attempt to ensure that extremely small-scale variations are avoided. Although these criteria have a clear spatial implication, they can not always be translated in the form of mapping information that is given by mathematical models. In the following a general framework is proposed that can serve as a common operational concept for measuring and modelling. It will be used to define the assessment in time and space. It is remarked that this framework is not expected to significantly affect the current practice of measuring or modelling.

**Two types of spatial coverage**

Before specifying the framework for assessment in time and space, it should be noted that the Framework Directive gives several reasons to assess the concentration distribution. In view of the purposes of the assessment (section 3.2.1), two types of spatial coverage should be distinguished, which are to be followed in parallel:

a) focusing on the areas within the zones where the highest concentrations occur for compliance analysis: are limit values or alert values exceeded?

b) addressing the levels in the other areas within the zones for other air quality management purposes (e.g. for assessing the total exposure of the general population, or for trend analysis).

Siting criteria for these two types of stations are difficult to reconcile: the first type of stations should be sited at *hot spots*, the second type are typically sited to monitor the
urban and rural background\textsuperscript{13} levels. Therefore the two types should be distinguished in the development of an assessment strategy.

*Specification in time and space of air quality parameters to be assessed*

It is not sufficient to generate a picture of the concentrations in time and space that is complete, detailed and accurate as possible, one also needs to define the parameters of the concentrations (limit values, alert values or other parameters for air quality management purposes) that are to be assessed. Each of these must have its own temporal and spatial characteristics, which can be expressed in analogous terms:

1. The area/period over which compliance will be judged;
2. The area/period over which the limit value/alert value should be applied;
3. The time/space over which one should average before comparing the concentration to the limit value/alert value;
4. The statistical parameter of the concentration distribution that is used for comparison with the limit value/alert value.

Table 3.2.2 elaborates this for time and space. In a background document this will be discussed in more detail.

\textsuperscript{13} The term background level refers to the level in a relatively large area, excluding local peaks. Levels in city parks are typical urban background levels, levels that are not more than usually affected by sources within many kilometres are rural background levels.
Table 3.2.2 Summary of the characteristics of the concentration distribution in time and space that have to be assessed

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Characteristic applied to time</th>
<th>Characteristic applied to space</th>
</tr>
</thead>
<tbody>
<tr>
<td>Period/area over which compliance will be judged</td>
<td>Reference period. For LVs this period usually is a (calendar) year. For AVs it is of the order of hours.</td>
<td>Reference area. In the Framework Directive this is the zone.</td>
</tr>
<tr>
<td>Period/area over which LV should be applied</td>
<td>Application period. Depends on when the targets are sensitive. For human health normally the entire year; for ecosystems it can be the period when plants are sensitive, e.g. the winter period.</td>
<td>Application area. Depends on where the targets are. E.g. an ecoLV may apply only in rural areas.</td>
</tr>
<tr>
<td>Time/space over which one should average before comparing the concentration to the LV/AV</td>
<td>Averaging time. Depends on the time in which the adverse effect builds up (also on operational convenience). For health often an hour, a day or the year is taken.</td>
<td>Averaging area. The averaging area is often not explicitly addressed in the definition of a LV. Instead, it is implicitly dealt with by prescribing siting criteria for monitoring stations, on the basis of a (often vague) notion of the area that the station should cover. A monitoring station is usually not located exactly at the square meter where the highest concentrations are expected, but at a location that is thought to be representative of a (somewhat) larger area.</td>
</tr>
<tr>
<td>Statistical parameter of the concentration distribution</td>
<td>Ideally the maximum value in the reference period is taken, but for practical reasons a number of exceedances (percentile) is often allowed. For an averaging time of one year there is only one value.</td>
<td>Usually the maximum value in the reference area is taken. (In UNECE protocols, however, the 95-percentile of each 150 km area is taken.)</td>
</tr>
</tbody>
</table>
In the following these general notions will be elaborated on the basis of separate discussions of the aspects time and space.

3.2.4.2 Time

Four different temporal aspects will be discussed:
A. The reference period
B. The application period
C. The averaging times and statistical parameters
D. The development over time of the assessment procedure

A. The reference period
The period for judging compliance period is part of the limit value: one calendar year. However, the winter period for the Eco limit value will be taken as a contiguous period, starting in October of the preceding year.

B. The application period
The Health limit values apply during the entire year. The Eco limit value applies (1) to the entire year and (2) to the winter half year.

C. The averaging times and statistical parameters
The definitions of the limit and alert values explicitly state the statistical parameters and averaging times of the concentrations to be assessed for judging compliance (see Table 3.2.3). In addition to these, other averaging times and statistics can be necessary for the purpose of air quality management (AQM): for reasons of continuity (trend analysis; current practice in assessments) the maximum of hourly averages has been added. Table 3.2.3 gives a summary.

<table>
<thead>
<tr>
<th>Averaging time</th>
<th>Statistics</th>
<th>Purpose</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 hour</td>
<td>24 exceedances per year allowed</td>
<td>Health limit value, AQM</td>
</tr>
<tr>
<td>24 hour</td>
<td>3 exceedances per year allowed</td>
<td>Health limit value, AQM</td>
</tr>
<tr>
<td>Winter half year</td>
<td></td>
<td>Eco limit value, AQM</td>
</tr>
<tr>
<td>Year</td>
<td></td>
<td>Eco limit value, AQM</td>
</tr>
<tr>
<td>1 hour</td>
<td>3 consecutive hours</td>
<td>Alert value, AQM</td>
</tr>
<tr>
<td>1 hour</td>
<td>Maximum</td>
<td>AQM</td>
</tr>
<tr>
<td>10 minutes***</td>
<td>Relation to hourly averages</td>
<td>AQM</td>
</tr>
</tbody>
</table>

* Equivalent to the 99.7-percentile
** Equivalent to the 99-percentile
*** At some stations only

D. The development over time of the assessment procedure

Four aspects of changes in time of the assessment procedure are distinguished:

(1) the preliminary assessment, to be executed before definitely establishing the assessment methodology in an area,
(2) the revisions of the assessment regime,
(3) the period when the temporary margin of tolerance for the limit value applies and
(4) the continuity aspect for trend analysis.

1. Preliminary assessment
Before the assessment system to be used in an area can be definitively established, a preliminary assessment of the air quality situation in the Member States is required. This assessment should identify the areas where the concentrations are above x% and y% of the limit value and should also give information for air quality management purposes. If historic data are available, this assessment should be based on the situation in the last five years. A description of the initial assessment will be given in a guidance document that will be written by the EEA/TCAP, JRC and the European Commission.

2. Revisions of the assessment regime
When the assessment regime needed in a certain area has been determined on the basis of the preliminary assessment, the assessment system will be set up. However, the assessment regime, which depends on whether the limit values are in danger of being exceeded, may change due to long-term trends in the concentrations. A period of one year would be too short to judge this; even statistics for long time periods like a year fluctuate due to annual meteorological variations. Consequently, in areas where the levels are normally somewhat below the limit value, the levels may fluctuate to values above it in an unfavorable year. The introduction of the factor x (see below) attempts to avoid that in situations where the limit values are in danger to be exceeded, less stringent assessment requirements would enter into force after a year when no exceedances happened to occur. If the assessment regime would yearly be fixed by exceedances of x% of the limit value in the previous year, it would also fluctuate from year to year. The same applies to assessment regimes based on exceedance of y% of the limit value. To avoid the assessment requirements to change on a yearly basis, a period of five year for revision the assessment regime is proposed. The assessment regime will be based on the median value of the five annual exceedance rates: if three or more years were in exceedance the assessment regime will be based on exceedance, if only less than three years were in exceedance the assessment regime will be based on no exceedance. The numerical values for x and y will be discussed in section 3.2.5.

In case the levels undergo a rapid and structural change, e.g. due to the introduction of important sources, an additional half-term assessment is needed to determine whether the assessment system should be adapted to the new assessment needs.

3. Temporary margin of tolerance
When the Directive enters into force, a margin of tolerance will be introduced. During this period the values x and y will be taken as percentages of the limit value excluding the temporary margin (see also Figure 3.2.1). Since the assessment regimes are not linked to the temporary margin, the temporary margin will not affect the assessment procedures.

4. Continuity
For trend analysis purposes it is important that stations remain in operation for a long period. This should be an important consideration in revising and optimizing a network.

3.2.4.3 Space

The following spatial aspects will be discussed:

A. Zones
B. Areas
   B.1 High concentration areas
   B.2 Averaging areas
   B.3 Areas where to apply the limit value/alert value

A. ZONES
Each Member State must divide its territory into zones and specify the borders of each zone. Zones serve to judge compliance; two types of zones exist:
- Zones in which no areas exist where a limit value is exceeded; these zones are in compliance with the Directive.
- Zones in which areas exist where one or more of the limit values are exceeded; these zones are not in compliance with the Directive, and the MS are obliged to take specified air quality management actions (analysis, reporting, abatements).

The Framework Directive also attaches to zones a function in the prescription of the assessment method. For that purpose, two types of zone are distinguished:
- "Agglomerations": zones with more than 250 000 inhabitants or zones with less than 250 000 inhabitants but where the population density justifies, for the Member State, the need for air quality assessment and management;
- Other zones.

For practical reasons, non-contiguous built-up areas that are smaller than agglomerations may be gathered to constitute together one larger zone.

B. AREAS

B.1 High concentration areas
The exceedance areas, i.e. areas where the concentrations exceed the limit value, and high concentration areas, i.e. areas where x% of it or y% of it, are of special interest, since they have a status different from other parts of the zone. Areas where a limit value is exceeded will be the focus of air quality management. High concentration areas need to be assessed more intensively than the other parts of the zones. A high concentration area may be much smaller than the zone to which it belongs, but may also cover the entire zone (and extend beyond it). Section 3.3 discusses how these areas relate to the various assessment regimes.

B.2 Averaging areas
It is not always reasonable to demand that the concentration on every square meter is below the limit value. Not only would this give practical assessment problems, but, even more importantly, there are fundamental reasons to judge compliance not on an extremely small spatial scale. The situation is different for limit values for health, limit values for ecosystems, the guidelines for materials and the alert value.
. Health limit values
For health, there are no clear rules concerning the minimum area size that is relevant for human exposure. Walking people can move over a considerable distance in the time that is needed for effects to build up: in case of the short term SO₂ effect time of 10 minutes the walking distance is a kilometer. On the other hand, people who live at a location of high concentration may spend most of the year within an area of a few square meters. Although they will typically spend less time outdoors than indoors (where SO₂ levels are lowered by deposition onto walls), there may be prolonged periods, e.g. warm episodes, during which they remain in their garden or have doors and windows wide open.

. Eco limit values
For ecosystems the exposure situation is quite different. A plant is fixed to its own square meter and is always outdoors. The general purpose of limit values for ecosystems is however, in contrast to the situation for human beings, not to protect individual plants, but to protect ecosystems as a whole, which means that not every square meter could be fully protected. In the critical load approach taken in the framework of the Convention on Long Range Transboundary Air Pollution the emission reduction protocols are now based on a protection of 95% of the surface in each 150x150 km² grid cell. Such a statistical criterion can not be applied in a straightforward way to the assessment of results of a monitoring network, since a very large number of stations would be required to derive a statistical quantity of that type.

. Materials guideline
Like vegetation, materials are usually fixed to a location. In contrast to the limit value for vegetation, a guideline for materials would aim at the protection of individual targets (monuments and other objects of cultural heritage). So, small-scale areas where the long-term average concentration is high are relevant.

. Alert value
For alert values one can argue that only exceedances over a considerable area justify alerting the general population. In the reduction strategy during ozone episodes in Germany short-term actions are only taken when the threshold value is exceeded over an extended area covered by at least three stations. This does not mean that in case of small-scale exceedance of the concentration of the alert value no local short-time actions would be needed.

To take these considerations into account, averaging areas for the various limit and alert values will be defined. It is remarked that this would allow a part of the averaging area to have concentrations above those specified in the limit values/alert value.

This approach would lead to a set of averaging areas which depend on the limit value/alert value. Such a set could be quite practical when judging compliance on the basis of a detailed map of the concentration distribution. Model calculations or combinations of monitoring data and other assessment methods have the potential to provide such maps. However, for the choice of the location of a monitoring station
for compliance analysis, the approach of defining averaging areas per individual limit value would give problems, since a single station cannot represent the average over all the different surrounding averaging areas belonging to the various limit/alert values. Again, a practical choice has to be made. It is proposed to set two different averaging areas: one for judging compliance with the health limit value and, if appropriate, the limit value for materials and one for the Eco limit value. For siting monitoring stations this is a practical option, since when stations are set up for compliance analysis it will often be known whether the Health limit value (or materials limit value) or the Eco limit value is in danger of being exceeded.

. Averaging area for Health limit value (and Materials if appropriate)
In order to protect the population it is proposed to set the averaging area for the Health limit values not too large. An averaging area of 10 000 m² is proposed for compliance analysis. In terms of monitoring siting, this means that the station should not be sited to measure local peaks with a spatial extent smaller than 100 m. This spatial extent seems also reasonable for the protection of cultural heritage.

. Averaging area for Eco limit value
In rural areas where the levels are below the Health limit value, the Eco limit value can be approached or exceeded. Since this limit value should be not be applied to small areas, it is proposed to use an averaging area of 1000 km². This means that the station should not be sited to measure local peaks with a spatial extent smaller than 30 km.

. Averaging area for alert values
Also for the alert value an averaging area could be proposed. However, it is preferred to relate the spatial extent of the alert value to the exposure of the population than to a specified spatial extent. In Section 2.8.2 it was stated that it should only pertain to cases when a significant number of the population is exposed. So, in densely populated areas a smaller area would be sufficient to inform the population than in a sparsely populated area. The spatial extent is not further specified here.

Table 3.2.4 summarizes the averaging areas.
Table 3.2.4 Averaging areas per type of limit value and guideline

<table>
<thead>
<tr>
<th>Threshold value</th>
<th>Averaging area</th>
</tr>
</thead>
<tbody>
<tr>
<td>Health limit value, Materials guideline (if appropriate)</td>
<td>10 000 m²</td>
</tr>
<tr>
<td>Ecosystems limit value</td>
<td>1000 km²</td>
</tr>
</tbody>
</table>

† This does not protect smaller valuable ecosystems. It is the responsibility of the Member State to protect those ecosystems (see also section 2.7).

**Averaging areas in measuring and modelling**

The approach described above will be elaborated in section 3.3 and 3.4.1 on measuring and other assessment methods respectively. It is useful to relate the concept of averaging area here to the practice of modelling and measuring. For modelling, the averaging area should (in the ideal case) be equal to the model resolution used for compliance analysis: variations within the averaging area should not be resolved, while peaks of the size of the averaging area should not be smoothed or averaged over larger grid areas. For measurements, the averaging area defines the micro-siting of stations: stations should not measure micro-scale peaks within the averaging area, but one should attempt to site stations so that they, as far as possible, measure the average concentration over an area (approximately) as large as the averaging area.

In practice, the concept of averaging area should be applied in a flexible way. For measuring the concept can be regarded as a quantitative way of expressing that a station should not be too close to a source. The exact siting of stations is usually subject to many practical limitations, and the micro-scale concentration distribution is often not known well.

**Averaging area versus representativeness**

In measuring strategy the averaging area is not clearly distinguished from areas for which a station is representative. The averaging area, being the minimum size that one should consider in compliance analysis, is, however, usually much smaller than the area of representativeness of a station. For example, the averaging area around a street station is a limited area around the station, while this station (more precisely: this averaging area) can be representative for many other streets.

**B.3 Application areas**

The application area depends on the various limit and alert values.

- **Health and alert values**
  People can be present at virtually all types of locations within the territory of Member States. Consequently, the limit values for health protection and alert values should apply to the whole territory of Member States.

- **Limit values for the environment/ecosystem**
  Of the possible limit values for ecosystems, the most generally applicable value, pertaining to ecosystems that are widely present in the Community, was chosen in chapter 2. It should apply in every region in the EU outside built-up areas. Some
Member States have ecosystems within their borders that are not protected by this general limit value; it will be the task of the Member States to maintain the air quality at levels that are sufficiently low to protect these ecosystems (see Chapter 2).

. Transition area
Since the concentrations will not drop steeply beyond the border of a built-up area there will usually be an area around (continuously) built-up areas where the concentrations gradually decrease from urban to rural levels. The limit value for ecosystems is effectively more stringent than the limit values for health. When the Eco limit value would be rigorously applied directly beyond the border of every built-up area, exceedance would be difficult to prevent in the areas around it when the urban air quality would be just below the health limit values. Because of this, it is proposed to allow Member States to define around agglomerations and other built-up areas transition areas to which the Eco limit value will not apply. The maximum size of the transition area is defined as follows. All locations within a given distance from the border of a built-up area can be part of the transition area. This given distance is equal to 3 x the distance between the center of the built-up area and the border (more precisely: between the center of the built-up area and the point of the border that is closest to the location considered).

In summary, it is proposed that the limit value for environment protection should apply everywhere in the EU, except in the agglomerations and other built-up areas and their transition areas.

. Limit value for cultural heritage
The target of a limit level for materials is the cultural heritage, such as historical buildings and monuments; it is not necessary to extend the area of application beyond the locations where these objects actually are present, and since the objects are fixed to their place, it is proposed to restrict the area of application to the area covered by sensitive objects of cultural heritage. It will be the task of the MS to designate the objects that need protection, either as categories or individually.

3.2.5 Factors x and y

3.2.5.1 Factor x
Due to meteorological variations the concentrations parameters that are used for the assessment fluctuate from year to year. In situations where the concentrations are near the limit value, the concentrations may randomly vary above and below the limit value. With the goal of achieving a high level of protection, the Framework Directive requires the same level of assessment effort in areas in danger of exceeding the limit value as it does for those areas which are in exceedance. These areas in danger of exceeding the limit value are defined as being above x% of the limit value, where x is less than 100. If this criterion would be applied on an annual basis, the assessment requirements, including those for monitoring, could change from year to year. To stabilize this criterion, a period of five years was proposed in section 3.2.4; to judge whether the concentrations are above x or y%, the median value of the exceedance rates of five years would be taken. Exceedance of x% should be judged similar to
exceedance of the limit value: the number of exceedances allowed in the limit value also applies to the x% threshold.

The factor x will be chosen on the basis of the inter-annual variation of the concentrations. If (in three out of five years) the concentrations are above x% of the limit value, the most stringent assessment regime applies. If these concentrations are below x% of the limit value, the Framework Directive relaxes the obligations regarding the assessment system somewhat. The accuracy of this less stringent assessment methodology should be sufficient to make it reasonably certain that the concentrations found near x% of the limit value will in reality not be above the limit value.

The inter-annual variation depends on the averaging time and the statistical robustness of the concentration parameter concerned. Shorter averaging tends to increase the variability; higher percentiles (particularly the highest one, i.e. the maximum) fluctuate more than lower percentiles. The same applies to the accuracy of the assessment methods: concentrations based on shorter averaging time and higher percentiles tend to be more sensitive to (measuring or modelling) errors. Consequently, the optimal value for x depends on the concentration parameters used in the various limit values.

The Framework Directive does not explicitly state whether the assessment method in a certain area can be different for the various limit values, depending on their exceedances, or should be the same for all limit values. Since it would be impractical to have many different assessment regimes per limit value in a single area, it is proposed to have two assessment regimes per area, one for the Health limit values (and Materials limit value if appropriate) and one for the Eco limit value. This differentiation between requirements for the two types of limit value avoids that a strict assessment regime would be demanded in rural areas when only the Eco limit value would be in danger of being exceeded.

A second question is how these two assessment regimes should relate to the various limit values. Two choices could be made. The "above x%" assessment regime could be made obligatory when x% of any of the limit values is exceeded, or one could link the assessment regime to a single one of the limit values that it is associated with. An obvious advantage of the first choice is that it gives more certainty that the most accurate assessment regime applies if any limit value is in danger of exceedance. On the other hand, one should realize that the short term Health limit value has a large inter-annual variation and so the x value would have to be relatively low, requiring a large monitoring effort. The gain of such increased monitoring work would in terms of accuracy be rather low for the short term levels. For local peaks the accuracy of measured short term exceedances around local sources is typically low because of the representativeness problem. For exceedances of the short term limit value due to long-range transport one does not need an elaborate assessment methodology. Therefore it is proposed to use only one limit value for the determination of the assessment regime for the Health limit value. The 24-hour mean value is regarded as the most suitable one, because it lies between the annual average and the short term value. For the Eco limit value, which would apply to relatively large averaging areas, where small-scale peaks are not very important, the
winter mean is usually the strictest value. So, the winter mean limit value is proposed for the determination of the associated assessment regime.

In the above approach it is accepted that a relatively mild assessment regime (corresponding to <x%) applies to an area where the levels are between x% and 100% of the 1-hour mean limit value. It is, however, not desirable to allow situations to occur where the 24-hour mean limit value is below x% of the limit value, leading to the relatively mild assessment regime, while the other Health limit value (1-hour mean) is found to be exceeded. Therefore the requirement is added that in case a health limit value is exceeded the strictest assessment regime should apply. (The same applies in principle to exceedance of the annual mean Eco limit value while x% of the winter-mean is not exceeded - this seems a highly improbable situation, though.)

The numerical value of x is derived from empirical data on the interannual variability of the concentrations. From German data during the period 1986-1995 the standard variation, after correction for the long-term trend, was determined for a variety of SO₂ air quality parameters. Table 3.2.5 summarizes the results for 24-hour average concentrations.

<table>
<thead>
<tr>
<th></th>
<th>Rural sites</th>
<th>Source related sites</th>
<th>Urban background sites</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean</td>
<td>25%</td>
<td>18%</td>
<td>21%</td>
</tr>
<tr>
<td>98-percentile</td>
<td>44%</td>
<td>43%</td>
<td>45%</td>
</tr>
<tr>
<td>99-percentile</td>
<td>43%</td>
<td>45%</td>
<td>56%</td>
</tr>
<tr>
<td>Maximum</td>
<td>28%</td>
<td>47%</td>
<td>43%</td>
</tr>
</tbody>
</table>

A similar analysis of APIS data by the Commission is depicted in Figure 3.2.2: for a selection of stations the standard deviation of the 98-percentile of 24-hour mean values was (after correcting for the trend) somewhat smaller: 20-30% typically, with a highest value of 60%.

Based on the considerations given, a value of 60% is proposed for x%. 

Table 3.2.5 Year-to-year variation of 24-hour averaged SO₂ concentration parameters in Germany
3.2.5.2 Factor y

The Framework Directive allows to use the mildest assessment regime when the concentrations are sufficiently far below the limit values. The accuracy of that methodology should be sufficient to conclude when concentrations are found to be below \( y\% \) of the limit values, that limit values are not exceeded in reality. Because of this, the factor \( y \) may be chosen on the basis of the accuracy of the methods allowed in the mildest assessment regime in the range of the limit values.

Since the accuracy of assessment methods generally depends on the concentration parameter (averaging time, percentile), the optimal value for \( y \) in principle depends on the concentration parameter.

Similarly to the choice of \( x \), it is proposed to determine the assessment regime for the Health limit values on the basis of the value of the 24-hour mean concentrations and the regime for the Eco limit values on the winter mean. A value of 40\% is proposed for \( y \).

3.3 Measurement strategy

3.3.1 General

*Theory versus practice*
Before specifying the measuring strategy for SO2, it is remarked that the design of a monitoring network is in practice always a compromise of theoretical considerations and practical restrictions. The assessment criteria given here should be approached as much as is reasonably possible. This holds especially true for the concepts of high concentration areas and averaging areas. Although these can be operationally implemented in assessments by models, strict and formal application in measuring strategy is often not possible because the concentrations are only approximately known. One should realise, however, that similar, though less explicit notions are commonly used in measuring strategies. Further, it is very important to note that the assessment of the various pollutants covered by Daughter Directives should be harmonised, e.g. regarding the siting of multicomponent stations in urban areas. The prescriptions given here have been drawn up without detailed knowledge of the prescriptions for the pollutants of the other Daughter Directives. For reasons of efficiency the prescriptions should be harmonized where possible, and possibilities to measure several pollutants at one station should be promoted.

**Measurements alone are insufficient for assessment and air quality management**
The Framework Directive gives certain prescriptions concerning the measurement strategy (see section 3.2). Even a dense measuring network can not give a complete picture of the concentrations in a zone, since it does not measure everywhere. At least there should be, in addition to the measurements, an interpretation of the measurement results. So, a meaningful measurement strategy can not be defined without considering how the measurement results will be complemented with some sort of additional assessment (see also section 3.4.1).

**Practical possibilities of "other assessment methods"**
The Framework Directive stipulates that the air quality in Member States should be assessed on the basis of common methods and criteria. For the Community as a whole it would be desirable to implement a sophisticated combination of measuring and other assessment methods in all Member States. However, the methodology of combining measurements and other assessment methods is still in development and far from completion. The practice and the experience in the various Member States are very different. Therefore, it would not be effective to prescribe a sophisticated assessment technique. On the other hand, it would not be useful to adjust the assessment methodology to the least developed systems in the EU. Because of this, two assessment methods of different sophistication are proposed to be allowed.

**Two equivalent methods allowed**
The most important goal of the harmonizing the assessment methodology is that the quality of the results for the Member States should be equivalent (the term equivalent will be discussed below). It is proposed to allow two types of assessment and to require these to be equivalent:
1. an assessment essentially based on measurements alone,
2. an assessment based on measurements and supplementary assessment.

The first method is the purely measurement-based approach that has been employed in many networks, but which provides no objective basis to estimate concentrations at locations where no station is present. Consequently, a relatively large number of stations is required to give a satisfactory picture of the concentration distribution in a zone. The second method uses existing scientific knowledge in addition to monitoring results and
requires less stations to give a satisfactory description of the concentration distribution in a zone.

Ideally, the criterion of equivalence of both assessment types should be expressed as the required accuracy of the results (exceedances and other information for air quality management purposes) of the two methods. However, the accuracy of the concentrations in the first method is in general undefined at locations where no station is present, and so it is not possible to quantify the accuracy in general terms. In addition, the accuracy of models and other mathematical methods is usually known for a limited number of situations, and it is difficult to specify the accuracy in general. Consequently, a more subjective approach is inevitable.

### 3.3.2 Network density in the case of supplementary assessment

**Guidance document**

The second type of assessment, based on measurements and supplementary assessment, will not be described here. It will be described in a separate guidance document, which applies to all four Daughter Directives that are currently in development. The guidance document is intended to be an annex to the four Directives.

**Network density depends on the supplementary assessment method**

The added value of the supplementary assessment should at least compensate the reduction in the number of stations compared to the case of no supplementary assessment. As long as this assessment method has not been described, it is difficult to express its added value in terms of the numbers of stations that can be omitted. Assumptions will be made here regarding the results of the supplementary assessment in order to arrive at a specification of the minimum density of stations. It is assumed that the supplementary assessment will result in an annual report on the spatial distribution of the concentrations in each zone, including territory covering information on the exceedances, and that this report will be forwarded to the Commission together with the measurement data from the measuring stations. For the rural levels this distribution is in the form of maps. For the urban scale and local scale it is in the form of spatial statistics; in particular for each limit value the total area (in km$^2$) where exceedance occurred is quantified. Maps for these smaller spatial scales could be optional. See also Chapter 5 on reporting. The maps should be of sufficient accuracy, but it is very difficult to quantify this accuracy. Is would be meaningless to require that the quality of the information in the maps should be equivalent to that of a network that would exist in the case of no supplementary assessment, since the concentration in such a network is specified only where a station is present. The minimum number of stations would at least be the minimum that the Framework Directive prescribes: continuous measurements should be done in each agglomeration and in each zone where the levels are above x%. So, in those zones the minimum number of station should at least be one. Additional minimum numbers are not given here. It is proposed to maintain the current size of the existing networks in the Member States, and to require that any planned reductions should be justified to the European Commission. In addition, it should be required that the number of stations must be sufficient to generate the maps and/or statistics on the spatial distribution.

### 3.3.3 Network density in the case of no supplementary assessment
In Section 3.2.4.1 it was argued that the measuring strategy is in principle different for stations for compliance analysis (stations in most polluted areas) and stations for air quality management alone (background stations). This does not necessarily imply that both type of stations should be distinguished in the definition of minimum station densities. Where levels are not below $y\%$ of the limit value these stations may often coincide and serve both purposes. For the case that levels are below $y\%$ of the limit value, one can argue that compliance stations are not necessary. In order to avoid too complicated prescriptions, the minimum densities will not be prescribed for compliance and background stations separately, but for both types together.

Density prescriptions not for zone but for high concentration area
In section 3.2.3 the assessment regime was linked to the high concentration areas. Consequently, for the designation of the network density it is necessary that the high concentration areas are approximately known. In the following it is assumed that the preliminary assessment will yield an indication of the high concentration areas both for $x\%$ of the limit value and for $y\%$ of the limit value. In the case that a preliminary assessment would fail to identify high concentration areas within a zone, it will, by default, be assumed that the high concentration area (of $x\%$ or $y\%$, depending on the highest concentrations found) coincides with the entire zone.

Station types
For $SO_2$ three station types are relevant:
- rural stations
- urban (background) stations
- local stations

The rural stations of all Member States together should give the general pattern of concentrations over the European territory. The EMEP network has a similar function (and, if fulfilling the Directives requirements, might be combined with the network for the Directive). The urban stations should represent the urban background levels as well as possible. For large urban areas several stations need to be used, which together represent the spatial pattern of the concentrations.

Rural and urban stations are similar to station types for other pollutants, but the siting of local stations depends on the pollutant. For $NO_2$, lead and $PM_{10}$, the concentration gradients near traffic justify the designation of road side stations. For $SO_2$, industrial sources are probably the main causes for small scale concentration peaks, but near other types of sources, e.g. where sulphur containing fuel is used for heating of buildings, local concentration increases may also occur. The strategy of specifying an $a\ priori$ number of local stations, as is done for road side stations in parallel position papers, would not be suitable for industrial stations. The patterns of road side pollution are much more homogeneous throughout the EU than of industrial $SO_2$ pollution, and consequently the number of stations should not be linked to city size or to the presence of industry, but should be directly related to the expected size of polluted areas (high concentration areas).

Minimum station densities
For the determination of the network density the following additional elements are proposed:
The station density will be defined as the number of stations per inhabitant for the Health limit value and as the number of stations per km$^2$ for the Eco limit value.

In case of exceedance of x% or y% of a Health limit value, the density will be higher in built-up areas than in other, rural areas.

High concentration areas within built-up areas that are too small to constitute an agglomeration will be subject to the same station density requirements as agglomerations (except the requirement that there should be at least one station in each agglomeration).

The minimum station densities proposed are given in Table 3.3.1.

Table 3.3.1. Minimum density of stations in the case that no supplementary assessment is made

<table>
<thead>
<tr>
<th>Type of area</th>
<th>Total area to be covered</th>
<th>Minimum station density</th>
</tr>
</thead>
<tbody>
<tr>
<td>Agglomerations and other built-up areas</td>
<td>Area &gt; x% Health LV $^*$</td>
<td>10 per 10$^6$ inh.</td>
</tr>
<tr>
<td>Agglomerations and other built-up areas</td>
<td>Area &gt; y% Health LV $^*$</td>
<td>3 per 10$^6$ inh.</td>
</tr>
<tr>
<td>Agglomerations and other built-up areas</td>
<td>Area &lt;y% Health LV $^*$</td>
<td>1 per 10$^6$ inh.</td>
</tr>
<tr>
<td>Rural areas</td>
<td>Area &gt; x% Eco LV $^*$</td>
<td>10 per 10000 km$^2$</td>
</tr>
<tr>
<td>Rural areas</td>
<td>Area &gt; y% Eco LV $^*$</td>
<td>1 per 10000 km$^2$</td>
</tr>
<tr>
<td>Agglomeration</td>
<td>&lt; y% all LV</td>
<td>1 per agglomeration</td>
</tr>
<tr>
<td>Non-agglomeration</td>
<td>&lt; y% all LV</td>
<td>1 per 10000 km$^2$</td>
</tr>
</tbody>
</table>

$^*$ For 24 hours.

For winter half year; only where the Eco limit value is applicable.

It is remarked that the number of stations can be set much more cost-effectively when the causes of exceedances are taken into account. Two very different situations can be given as examples: on the one hand a zone in which several industrial sources of different types each have an exceedance area around it, amounting to e.g. a total exceedance area of 10% of the zone, and on the other hand a zone where 100% of the area is in exceedance due to long-range transboundary transport of air pollution. For the first zone Table 3.3.1 prescribes 10 times less stations than for the second zone, while the number of stations that would actually be needed to quantify the exceedances in the first zone would be larger than in the second zone, where a single station might suffice. In an assessment without supplementary assessment this distinction can not be made, since one can not assume that the representativeness of the stations would be known. However, it is proposed that if the Member State demonstrates that the exceedance is mainly due to long-range transport (i.e. that more than 2/3 of the concentrations during exceedance is due to sources at distances larger than 100 km), the number of stations can be reduced by a factor of 10, with a minimum of 1 station per zone if the levels are above y% of the limit value. Without a detailed analysis the consequences of this reduction factor are difficult to assess. The chosen factor of 10 may be too large. The Commission should reconsider the value before implementing it in the Directive.
C. Resulting network density

At first sight the station densities required seem to be very high. In practice, however, three principles have been introduced which reduce the station densities very considerably:

- The station densities given for levels above y\% or x\% of a limit value do not refer to the entire zone, but only to the areas where the levels are actually above y\% or x\%.
- For rural stations the SO2 levels are often mainly due to long-range transport, which allows, in areas above y and x\% of the limit value, to reduce the number of stations by a factor of 10.
- In case of supplementary assessment the station densities can be reduced.

3.3.4 Siting criteria

The strategy for the siting of monitoring stations can be separated into two main elements: criteria for the macro-siting (or network design), which describe how the stations of a network should be distributed within the entire concentration field that is to be assessed, and criteria for the micro-siting, which describe how the station should be exactly positioned within the area that was chosen on the basis of macro-siting, in particular with respect to very small-scale concentration gradients. It will be argued below that macro-siting should be related to the concepts of representativeness, and that micro-siting should be related to the concept of averaging area, introduced earlier in this paper.

Macro-siting

Macro-siting should optimize the information on the concentration distribution within the territory to be assessed. A second aim of macro-siting is to optimize the generation of air quality management information, i.e. data for the analysis of source contributions to the levels and of trends, but this will not be discussed here.

Before elaborating macro-siting criteria, the concept of representativeness will be discussed in more detail. Also the concentration data that the assessment should produce should first be addressed.

The concept of representativeness is particularly important for the assessment of numerous similar small-scale situations, like streets or small industries, which can not be individually assessed (by monitoring or modelling). To solve this, one often assumes that the results of an assessment of one location can be used (are representative for) for other, similar locations. Some examples may clarify this. Concentrations monitored in one or a few streets are assumed to be representative for the other relevant streets. The background levels in a city are assumed to be characterized by one or two stations. A set of model calculations of the concentration distribution around a few small industrial sources is assumed to be representative for similar sources elsewhere. The essence of using the concept of representativeness is that data for a small set of locations can be translated/extrapolated to data for a much larger area (though with limited accuracy). This is also the essence of macro-siting strategy.

Section 3.4 below discusses "other" assessment methods, including mathematical methods, to extrapolate measurement data to other locations. It is advantageous to take the potential of these methods into account in the macro-siting strategy. However, since a generally accepted methodology does not yet exist, it is not possible to have a particular
method in mind when developing a macro-siting strategy. The strategy should therefore be
general and flexible enough to link up to the existing way of working, and on the other hand
it should incorporate the potential of combining measurements with mathematical methods.

In Chapter 5 it is discussed how the concentration distribution should be reported. It is
stated there that maps of the concentration distribution are to be preferred, but that
statistics of the spatial distribution are an important minimum option, and should always be
given. Particularly for small-scale peaks near small industrial emitters or near traffic a
statistical description in terms of the area in exceedance is important and even more useful
at the EU level than maps. This implies that the reports should not be restricted to merely
the air quality at the stations, but should also give information on locations without a
station. A practical way to do this and to link this to the measuring network is to divide the
entire territory in areas of types that correspond to station type (rural, urban, industrial,
street). The spatial concentration distribution over each type of area can be implied from
the concentration data of the station(s) of the corresponding type. (Further subdivisions in
area types could be made if the available data allow this, e.g. regionally differentiation in
large Member States.)

Departing from the goals of the assessment, the macro-siting strategy can now be
described. It will be expressed only in general terms here and its further elaboration will be
left to the expert group attached to the Directive. The basic principle was stated already
above: \textit{macro-siting of stations should optimize the information on the spatial concentration
distribution within the zones.}

For \textit{compliance stations}, the network designer should answer the question how the
spatial distribution of exceedances can best be described. (Since the measurements are
continuous in time, the temporal distribution will be directly available.) The designer should
first estimate \textit{where} exceedances may be expected (in the first stage of implementation of
the Directive this will be the preliminary assessment, later it will be the revision of the
assessment). Then the designer should distinguish \textit{at which types of locations} the
exceedances are expected. The following types are expected to be relevant for SO\textsubscript{2} (but
further subdivisions may be distinguished if useful):
- Rural background locations\textsuperscript{13}
- Urban background locations\textsuperscript{13}
- Industrial locations

The designer should then investigate how a limited number of stations should be
distributed to give the best description of the exceedances in the territory.
Each relevant location type should be covered by one or more stations of the
corresponding type. Out of the very large number of locations of a certain type that are to
be assessed, the designer should select one or several locations that are, as good as
possible, representative of all other locations of this type. The designer should consider the
possibilities to generalize the measured concentrations, i.e. translate the results to the
other locations of the type considered. Depending on the type of locations, this could e.g.
be done by mathematical inter/extrapolation, by modelling or (as is currently often done) by
declaring stations to be representative for areas. Based on the possibilities to generalize
the results of measurements at individual locations, the designer should then optimize the
number of stations per station type and determine the measurement locations. The
designer should report the estimated or calculated representativeness of each station for
the entire set of location types that it represents (e.g. by reporting whether a street station represents the worst case (maximum) in the area or a typical (median) busy street - this should be elaborated in more detail). In the case of no supplementary assessment, the set of compliance stations by itself should be as much as possible representative of the exceedance situations that occur in the zone. In the case of supplementary assessment, this would also be important, but in addition the station locations should be chosen so optimize the possibilities for generalization.

For **background stations** (as far as these are not compliance stations) the designer should follow the same procedure as above, in this case, however, not aiming at the locations where exceedances may occur, but at all other locations.

The above procedure hypothetically assumes that the existing network can be completely redesigned. In practice, the possibilities for restructuring the network are more limited. Also for reasons of continuity (e.g. for trend analysis) one should change the locations of existing stations only as a last resort. The existing network should, however, be analyzed according to the above procedure, and for existing stations that are not changed, the information on the representativeness should be reported.

**Micro-siting**

The purpose of micro-siting is to position the inlet of the station so that the measured concentration approaches as closely as possible the local concentration which prevails in freely mixed air. Apart from practical criteria such as accessibility, safety, availability of electrical power, which will not be elaborated here, the major decision is to choose the exact position within the area that was chosen on the basis of the macro-siting strategy.

Vertically, the height of the inlet should be between 1.5 m (the breathing zone) and 4.5 meter above the ground. It should be at least at 1 m from any wall or structure and at least 20 m from a tree or any other major sink of SO2. There must not be any immediate local source of SO2.

The precise site should be chosen on the basis of the averaging area: the measurement should capture as good as is practically possible the concentration averaged over the averaging area. This implies that too small-scale peaks (or dips) in the concentration, i.e. peaks or dips that have a smaller scale than the averaging area, should be avoided. A difficulty is that the averaging area depends on the limit value: for the Health limit value the averaging area is smaller than for the Eco limit value. This problem, which is due to the point-wise character of the measurement, is inherently associated with fixed stations. In practice, the network designer should attempt to find for each station an acceptable approximation of what would be ideally desired. For a compliance station, the designer should consider which type of limit value exceedance is aimed at. If, e.g. near a local source, the Health limit value is the main reason to place the station, the corresponding averaging area of 10 000 m² (see Table 3.2.4) should be the primary aim. If, e.g. in a rural area, exceedance of the Eco limit value is the main reason to place the station, one should aim at an averaging area of 1000 km².
3.4 Other assessment methods

3.4.1 Mathematical methods

General

Modelling is always useful
The Framework Directive explicitly mentions the possibility to use models (or, more
generally, mathematical methods) in cases that the concentrations are higher than x% or
y% of the limit value, and allows the sole use of modelling where y% of the limit value is not
exceeded. In general, any methods that are able to expand the measuring results where
the limit values are approached or exceeded can be of great value, both for analyzing the
extent of exceedances and for air quality management. Using mathematical methods
obeys the general principle that in assessments one should attempt to use all existing
knowledge (here on emissions and dispersion).

Modelling source contributions and concentration distributions
Two important applications of modelling should be distinguished: (a) the analysis of the
causes of air pollution, i.e. the contributions from the various sources of air pollution, and
(b) the description of the concentration distribution in time and space.
(a) Since dispersion models explicitly relate emissions to concentrations, they are well
suited to analyses how the pollution is caused. By changing the emissions input the
effect of abatement measures can be also simulated. Since the control of emissions
is important item in the Framework Directive, particularly when limit values are
exceeded, this type of model application is very important. Other methods can also
be used to analyses source contributions, e.g. comparison of concentrations at hot
spots with those at background sites or source recognition methods. This type of
application of mathematical methods, though important, will not be discussed here
further (the analysis of the problem, the identification of the causes and the
measures to take are under the initiative of the Member States).
(b) Modelling for the description of the concentration distribution in time and space will
be discussed in more detail in the following paragraphs.

Mathematical methods to describe the concentration distribution in time and space

Various combinations of measurements and modelling
It is important to note that the distinction between measuring and other assessment
methods (interpretation, interpolation of measurements, modelling) is not as absolute as is
often suggested. Figure 3.4.1 illustrates that there is an almost continuous spectrum of
mixtures of measurements and other assessment methods. Neither of the two extremes is
useful for investigating the state of compliance of a zone: 100% measuring (i.e. doing
measurements that are not generalized at all) gives incomplete information, while, at the
other extreme, 100% modelling (i.e. applying models that have not in any sense been
validated) gives unreliable information.
Interpretation versus modelling
There is no fundamental difference between assessment by common sense and assessment by dispersion models. Models may be described as mathematical formulations of one's understanding. In the following the term model will be used for any formalized (algorithmically) method to calculate concentrations. So, an official statement that the concentration measured at a specified location is representative for the concentration at other specified locations can be regarded as a model. Interpolation methods are also models in this sense.

The relation between models and measurement data
The distinction between validated and unvalidated models is not always as relevant as is often believed. On the one hand, even unvalidated models have (indirectly) been based on measurements. On the other hand, even validated models can have erroneous results. For example, verification of point source models tends to focus on validation of the dispersion part of it. When a validated model is applied to an intermittent source which emits at unknown intervals, the calculated peak concentrations are unreliable.

Combinations of models and measurements
In this section some important examples of the application of mathematical models and the relation with measurements are discussed.

a. Using models without local measurements
In situations where no local measurement data are available and where direct inter/extrapolation of the results of the nearest stations can not be applied (e.g. near a small point sources) models can be used to estimate the local concentrations. The credibility of the results depend on the quality of the emissions and dispersion input parameters, and on the results of (earlier) model validation studies.
b. Integrating modelling and measuring results

In general, the quality and credibility of modelling results will improve when calculated concentrations are directly compared with concentrations that are measured within the time period and the area that the calculations pertain to. A very important question is how differences between calculated and measured concentrations should be dealt with. Often, inaccuracies of the input (emissions, meteorology) are large enough to explain the differences. In such cases, it is justified to improve the modeled concentration field by adjusting the input (within the uncertainty range) to improve the agreement. This procedure can be regarded as intelligent interpolation, rather than modelling. It has the advantage that it adds information on emissions and dispersion to the information given by the monitoring stations, without degrading the monitoring results. Especially when the model has been specially designed for this procedure, it can be a powerful assessment tool. It should be noted that this procedure is not (yet) generally applied. An example of an operational procedure is the CAR model as used in The Netherlands. This model contains a few adjustable parameters, which are annually fitted to the results of ten street stations and is subsequently used to calculate concentrations in complete networks of streets.

c. Interpolation of measuring results

More common than the intelligent interpolation described above is the direct interpolation which does not take information on emissions or dispersion into account. This is useful for uniform areas, but one should be aware that small-scale variations can not be identified. This method is often used for large scale patterns (continental, rural) and sometimes also for urban background patterns.

Types of models

Many models for the dispersion of SO2 have been developed and applied. These models need input regarding emissions, meteorology and sometimes topography. In most areas many sources contribute to the concentrations, and so the calculation of the concentrations requires a very extensive emission data base, which even needs to include sources in other countries. Often, one only calculates the contributions of local sources with the model, while the contributions of other sources are taken into account by adding measured background concentrations. This is the most common combination of models and measurements. Numerous variations of the Gaussian plume model are in use for local dispersion of point source emissions. Models for the dispersion at regional and national scales are somewhat less numerous, and models for the long range transboundary transport of SO2 are in operation at several research institutes.

Not only the distinction according to the spatial scale should be made, also the differentiation between long-term models and short-term models (which almost always operate on an hourly basis) is important. Since the assessment of threshold exceedance requires that short-term levels should be expressed in the numbers of exceedances of thresholds, models should be able to calculate the concentration distribution in time. Although most short-term models are capable to calculate a complete time series over the year, the addition of the background levels can give operational difficulties, e.g. because background percentiles may not be simply added to the percentiles of the local contribution.

In some countries a standard model for the dispersion of air pollution exists, but this is not applicable for all spatial and temporal scales that are covered by the assessment requirements of the Dt. Currently European scientists are working on harmonization and standardization of models. Regarding interpolation, several methods exist that may be
regarded as standard methods. These methods are useful for situations where a straightforward spatial interpolation is justified. There is, however, no generally accepted methodology for “intelligent” interpolation, which uses more information that the monitoring results.

**Criteria for models**
Since there are no standard methods available that can be prescribed as the only methods allowed or as reference methods, the requirements of the models (and other mathematical method) will need to be described in other ways, preferentially in terms of the accuracy of the results. It should be noted that it would be unrealistic to require that the model results are more accurate than the results of a (dense) monitoring network, which also have several inherent shortcomings. A distinction should be made between the requirements for the various assessment regimes. The accuracy requirements for models is in section 3.5.

**Reporting the results of mathematical methods**
An important question is what the form of the results of these additional methods should be. Until now, the reports of results of air quality assessment in the framework of EU air quality directives have been limited to statistics of measurement results. In Chapter 5 this matter will be discussed.

**3.5 Data quality objectives**
Data quality objectives must be established in order to comply with the assessment objectives. They will be defined in terms of required precision and accuracy, minimum time coverage and minimum data capture. Below, these requirements are preliminary expressed as the expected capabilities of the assessment methods. For the time being, the possibilities to relate the requirements directly to the assessment regime not considered.

Required precision and accuracy (expressed as maximum uncertainty of the assessment method):

- Mandatory measurements: ±15% (for individual measurements, including sampling, calibration and instrumental errors)
- Indicative measurements\(^\text{14}\): 30% (individual measurements)
- Modelling: 50% and 30%, respectively for daily and yearly averages
- Objective estimation: 100%

The proposed values are based on the performances that can be achieved by implementing techniques corresponding to the current state of the art for the various methods, and taking into account the provisions of Article 3 of the Framework Directive (approval of the measuring devices ensuring accuracy of the measurements, quality assurance programmes organized by the Commission).

Minimum time coverage of the measurements:

\(^{14}\) The term “indicative” refers to measurements performed by means of mobile laboratories or temporary fixed stations, as well as to cost-effective manual methods, such as in particular diffusive sampling methods.
Continuous measurements: 100% (continuous or quasi-continuous measurements)
Indicative measurements: 20% (every fifth day, or at random, or 2.5 months per year)

Minimum data capture:
Continuous measurements: 90% (breakdown and interruptions for maintenance and calibration allowed for 36 days per year)

### 3.6 Measurement methods

The measurement of SO2 can be divided in three separate steps:
- the sampling method;
- the measurement or analysis method;
- the calibration method (when the analysis method is not absolute).

The following tables gives the most current used methods and their main advantages and disadvantages.

#### 3.6.1 Existing sampling methods

<table>
<thead>
<tr>
<th>Method</th>
<th>Description</th>
<th>Reference</th>
<th>Advantages / Disadvantages</th>
<th>Used by</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Laminar flow manifold</td>
<td>Flow 150 l/min, tube diameter 15 cm (see Fig. 1) Inert material: ‘conditioned’ glass, stainless steel, Teflon</td>
<td>EPA</td>
<td>+ isokinetic sampling, sample unaffected - SO2 losses if no heating</td>
<td></td>
</tr>
<tr>
<td>2. Turbulent flow manifold</td>
<td>Modular sugar cane design (see Fig. 2) Inert material: glass, stainless steel, Teflon</td>
<td></td>
<td>+ low cost, modular construction - loss of particulates, SO2 losses if no heating</td>
<td></td>
</tr>
<tr>
<td>3. Sampling without manifold</td>
<td>Direct connection of analyser inlet to station sampling head</td>
<td></td>
<td>+ low cost, efficient sampling</td>
<td></td>
</tr>
</tbody>
</table>
## 3.6.2 Existing measuring methods

<table>
<thead>
<tr>
<th>Method</th>
<th>Description</th>
<th>Reference</th>
<th>Advantages / Disadvantages</th>
<th>Used by</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>1. Manual methods</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.1. TCM method</td>
<td>Bubbling in absorbing solution (TCM) + colorimetry</td>
<td>ISO 6767</td>
<td>+ cost effective - discontinuous and time consuming measurements</td>
<td></td>
</tr>
<tr>
<td>1.2. Thorine method</td>
<td>Bubbling in absorbing solution (H2O2) + colorimetry</td>
<td>ISO 4221</td>
<td>- handling of dangerous substances (Hg)</td>
<td></td>
</tr>
<tr>
<td>1.3. Total acidity method</td>
<td>Bubbling in absorbing solution (H2O2) + back titration</td>
<td>ISO 4220</td>
<td>- possible interferences - handling of hazardous substances</td>
<td></td>
</tr>
<tr>
<td>1.4. Titrimetric method</td>
<td>Bubbling in absorbing solution (H2O2) + titration</td>
<td>----</td>
<td>- not SO2 specific</td>
<td></td>
</tr>
<tr>
<td>1.5. Diffusive sampling</td>
<td>Diffusive sampling onto absorbent (TEA, Na2CO3) + colorimetry or ion chromatography</td>
<td>----</td>
<td>+ cost effective, ideal for large scale monitoring</td>
<td></td>
</tr>
<tr>
<td><strong>2. Automated methods</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.1. Coulometric method</td>
<td>Redox reaction in electrolytic cell + electric current measurement</td>
<td>----</td>
<td>+ continuous, real time measurement - requires regular calibration and maintenance</td>
<td></td>
</tr>
<tr>
<td>2.2. Conductimetric method</td>
<td>Reaction with H2O2 + conductivity measurement</td>
<td>----</td>
<td>- possible interferences</td>
<td></td>
</tr>
<tr>
<td>2.3. Flame photometry (FPD)</td>
<td>Burning of sample in hydrogen rich flame + flame photometry</td>
<td>----</td>
<td>- possible interferences - poor linearity, not SO2 specific</td>
<td></td>
</tr>
<tr>
<td>2.4. U.V. fluorescence</td>
<td>Fluorescence after excitation to higher energy level + light emission measurement</td>
<td>ISO 10498</td>
<td>- interferences (NO)</td>
<td></td>
</tr>
<tr>
<td><strong>3. Long path optical methods</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3.1. DOAS spectrometry</td>
<td>Differential optical absorption along path length</td>
<td>----</td>
<td>+ simultaneous multi-component analysis - integrated concentration over path length</td>
<td></td>
</tr>
<tr>
<td>3.2. DIAL spectrometry</td>
<td>Differential optical absorption of backscattered laser beam</td>
<td>----</td>
<td>+ easy, maintenance free operation - measurement at roof level, expensive analyzer, measurement disturbed by fog</td>
<td></td>
</tr>
</tbody>
</table>
### 3.6.3 Existing calibration methods

<table>
<thead>
<tr>
<th>Method</th>
<th>Description</th>
<th>Reference</th>
<th>Advantages / Disadvantages</th>
<th>Used by</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. TCM method</td>
<td>Bubbling in absorbing solution (TCM) + colorimetry.</td>
<td>ISO 6767</td>
<td>+ cost effective&lt;br&gt;+ basic calibration method&lt;br&gt;+ precise and accurate&lt;br&gt;- handling of hazardous substances (Hg)&lt;br&gt;- control of reagents purity required&lt;br&gt;+ primary calibration method&lt;br&gt;+ diffusion rate may be determined by simple weighing&lt;br&gt;+ continuous production of calibration gas mixture&lt;br&gt;+ cost effective method&lt;br&gt;+ precise and accurate (uncertainty ± 5%)&lt;br&gt;- control of SO2 purity is required&lt;br&gt;+ primary calibration method&lt;br&gt;+ good precision and accuracy (uncertainty ± 3%)&lt;br&gt;+ cost effective method&lt;br&gt;- control of SO2 purity required&lt;br&gt;- also suited for other pollutants&lt;br&gt;- difficult handling&lt;br&gt;- unknown stability of low concentration mixtures with time&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision for high concentration mixtures&lt;br&gt;- poor stability of low concentration mixtures with time&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method</td>
<td></td>
</tr>
<tr>
<td>2. Permeation method</td>
<td>SO2 permeation through a membrane into a flow of carrier gas, at constant temperature + periodic or continuous determination SO2 losses by weighing.</td>
<td>ISO 6349</td>
<td>+ cost effective&lt;br&gt;+ precise and accurate (uncertainty ± 5%)&lt;br&gt;- control of SO2 purity is required&lt;br&gt;+ primary calibration method&lt;br&gt;+ diffusion rate may be determined by simple weighing&lt;br&gt;+ continuous production of calibration gas mixture&lt;br&gt;+ cost effective method&lt;br&gt;+ precise and accurate (uncertainty ± 5%)&lt;br&gt;- control of SO2 purity is required&lt;br&gt;+ primary calibration method&lt;br&gt;+ good precision and accuracy (uncertainty ± 3%)&lt;br&gt;+ cost effective method&lt;br&gt;- control of SO2 purity required&lt;br&gt;- also suited for other pollutants&lt;br&gt;- difficult handling&lt;br&gt;- unknown stability of low concentration mixtures with time&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision for high concentration mixtures&lt;br&gt;- poor stability of low concentration mixtures with time&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available</td>
<td></td>
</tr>
<tr>
<td>3. Static volumetric method</td>
<td>A known volume of SO2 is added to a known volume of supplementary gas, under controlled pressure and temperature conditions.</td>
<td>ISO 6144</td>
<td>+ precise and accurate (uncertainty ± 3%)&lt;br&gt;- control of SO2 purity required&lt;br&gt;- also suited for other pollutants&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision for high concentration mixtures&lt;br&gt;- poor stability of low concentration mixtures with time&lt;br~- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method</td>
<td></td>
</tr>
<tr>
<td>4. Gravimetric method (high or low concentration mixtures)</td>
<td>A chamber is weighed before and after introduction of a certain quantity of SO2, then filled up with air and pressurized.</td>
<td>ISO 6142</td>
<td>+ gas cylinders commercially available&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method</td>
<td></td>
</tr>
<tr>
<td>5. Dynamic volumetric method</td>
<td>Introduction of a given flow rate of a gas into a constant flow rate of a supplementary gas. The gas is usually a high concentration gas mixture obtained by the gravimetric method.</td>
<td>ISO 6145</td>
<td>+ gas cylinders commercially available&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method&lt;br&gt;- gas cylinders commercially available&lt;br&gt;- easy handling&lt;br&gt;- good precision&lt;br&gt;- unknown accuracy&lt;br&gt;- no primary calibration method</td>
<td></td>
</tr>
</tbody>
</table>

### 3.6.3 Reference measurement method

The reference measurement method of the current directive is the TCM method (ISO 6767). Because of the toxicity of this method, only a few member States are still implementing it nowadays.

On the basis of the experience that was collected in the Member States and also during the European Commission's QA/QC programmes, the following reference method is proposed:
- measuring method: UV fluorescence (ISO 10498)
The TCM method, as well as the static volumetric dilution method, have proven to be equivalent to the proposed reference calibration method (permeation). This was validated by parallel measurements and interlaboratory testing in different QA/QC programmes. It should be noted that the availability of equivalent but independant calibration methods is very valuable in order to check the reliability of the reference method (“cross check principle”).

3.6.4 Quality Assurance and Quality Control of measurements

Quality assurance is a system of procedures that ensures that:
- measurements are precise and accurate,
- results are comparable and traceable,
- data are representative of ambient conditions,
- optimal use is made of resources.

The major constituents of a quality assurance program concern:
- network design (see separate chapter): number of stations, siting criteria;
- measurement technique: sampling, analytical and calibration procedure;
- equipment evaluation and selection: validation of methods, test of instrument performances;
- routine site operation: calibration in field conditions, maintenance, management and training.

QA/QC procedures are described in the WHO UNEP GEMS/AIR Methodology Review Handbook Series, Volume 1, "Quality Assurance in Urban Air Quality Monitoring" and in the EC "Instruction Manual for Air Pollution Monitoring, Volume 1: Sulphur Dioxide Monitoring".

Currently QA/QC programs only exist in a few monitoring networks of the EU Member States and with a variable degree of efficiency. This latter was shown by the recent field intercomparison organized by ERLAP in 36 network stations (see attached results).

With the increase of the monitoring networks foreseen with the implementation of the framework directive, it is expected that a lot of new laboratories, with among them a great number of private companies, will be in charge of the monitoring task. This will require particular measures to assure the quality and comparability of the measurements and the capability of the laboratories:

-Accreditation of laboratories: different standardized QA/QC systems have been developed in recent years such as the Good Laboratory Practice (OECD), the ISO 9000 and the EN 45000 laboratory accreditation procedures. The EN 45001 procedure was developed by CEN in collaboration with the Commission and is best adapted for testing laboratories in the field of air pollution measurements. Laboratories asking for accreditation are audited by a national or international accreditation organization. This audit mainly concerns aspects such as laboratory installation and equipment, qualification and training of personnel, proper quality control, technical
audit and traceability of the measurements. The request for laboratory accreditation is the only enforceable way to ensure an effective QA/QC procedure.

- Validation of the measurement methods and standardization at CEN or ISO level.

- Certification of equipment: test of instrument performances (the development of a standardized CEN test procedure is therefore urgently needed).

- Organization of intercomparison at EU level: organization by the Commission of EU wide intercomparison exercises (round robin tests, inter-laboratory exercises, spot checks in the monitoring networks) to ensure comparability of the measurements at international level.

- Publication by the Commission of guidance documents, organization of trainings and workshops.
4. - Cost implications

This chapter will address the cost implications only in general; it will not be attempted to quantify the costs necessary to meet the proposed limit values. An analysis of cost associated to the proposed values will be presented in a separate document.\textsuperscript{15}

4.1 Introduction

In chapter 2, the proposed WHO guidelines for ambient $\text{SO}_2$ concentrations have been compared with available measured concentrations in the EU countries. It is estimated that about a quarter of the urban EU population is exposed to $\text{SO}_2$ average annual concentrations $>50\ \mu \text{g/m}^3$ (WHO human health maximum exposure guideline for long term exposure in ambient air). Also, it has been estimated that possibly half of the urban population is exposed to levels of $\text{SO}_2 >125\ \mu \text{g/m}^3$ daily concentration (the proposed WHO guideline for 24 hours) at least once a year. For the short term guideline of $500\ \mu \text{g/m}^3$ an exposure estimate could not be made. Similar evaluations have been made for ecosystems exposed to levels above the WHO guidelines.

The values proposed in section 2.8 for consideration as starting points for health limit values relate to the daily and short term averages, mentioned above, but are differently defined (exceedances are allowed). In addition, the cost implications should be considered for a range of limit values.

In chapter 1, the main sources for $\text{SO}_2$ emissions have been indicated. On a national and on EU scale the contribution from stationary combustion is predominant. In 1990 over 80% of total $\text{SO}_2$ emission came from stationary combustion, especially, from large combustion plants for public and industrial power generation. In general, emission from Large Combustion Plants (LCP) does not strongly affect the local ambient air quality, but the air quality at large distance from the combustion site. Residential/institutional combustion processes emit about 6-7% of total $\text{SO}_2$ emission and are closely connected to urban increases of $\text{SO}_2$ levels. Also, road transport, contributing about 2-3% of total emission is by enlarge confined to urban areas. Emissions related to industrial processes only represent some 3-5% of total $\text{SO}_2$ emission, but can have a very strong local impact on air quality in an industrial area.

In general terms, three types of areas can be considered regarding the relation between $\text{SO}_2$ emission and $\text{SO}_2$ air concentration, namely, rural, urban and industrial areas.

The air concentration in rural areas will be mainly determined by emissions from distant Large Combustion Plants, due to long-range transportation processes. Of course, the variation in air concentration between several rural sites can be quite large, as illustrated by concentration data in chapter 1, depending on the input from distant sources.

Urban area air quality will also be determined by emission from distant sources, but the levels will have, in addition to the rural background, contributions due to urban related activities, in particular residential combustion and traffic.

\textsuperscript{15} ‘Economic evaluation of air quality targets for sulphur dioxide, nitogen dioxide, fine and suspended particle matter and lead’
The air quality in industrial areas is determined by the rural background, a possible urban contribution and by locally important industrial activities in particular industrial processes such as ore melting processes.

The following table summarizes the three area types and the interaction between \( \text{SO}_2 \) air quality and source contribution. Of course, a natural source like volcano eruption will have a large impact on air quality, but is not considered here because of its incidental character.

<table>
<thead>
<tr>
<th>Area type</th>
<th>Sources determining air quality</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rural</td>
<td>Large Combustion Plants (LCP’s)</td>
</tr>
<tr>
<td>Urban</td>
<td>See rural + Residential combustion, Traffic</td>
</tr>
<tr>
<td>Industrial</td>
<td>See rural and urban + Industrial processes</td>
</tr>
</tbody>
</table>

The source groups not only differ in emission rate but also in temporal emission characteristics. Large stationary combustion, in general, is more or less a constant activity over the year, showing only some variation between winter and summer period. Clear exceptions are District Heating Plants and fossil fuel fired power plants for additional power generation in the winter period - additional to nuclear or hydro power generated plants.

Residential combustion for heating exhibits clear seasonal variation, but also over the week and the day, while traffic shows a distinct variation over the week and the day as well.

Industrial processes tend to be more or less constant over the year, except for organised holiday periods. However, emission peaks can occur at any moment, e.g in case of limited good-housekeeping practice.

In summary, when considering costs necessary to reach limit values for health and ecosystem protection as under consideration for EU one has to be careful not to limit the assessment to averages over time and/or space, but also to consider averages over shorter times and areas, among which are higher values than the averages. For long-term averages and large-area averages much work has been done already in the framework of the Second Sulphur Protocol. In space, residential combustion and industrial processes are probably the most important causes of relatively small-scale peaks in urban and industrial areas respectively. In time, emission peaks may lead to higher peaks than would be expected on the basis of unfavourable dispersion alone.

### 4.2 Reduction of ambient \( \text{SO}_2 \) air concentrations

As the \( \text{SO}_2 \) emission is for the largest part related to fuel combustion processes it is important to consider possible reductions in relation to energy demand, type of fuel consumed and improvement of abatement techniques.

Options for reductions of the emissions from large combustion plants are applying Best Available Technology - a clear distinction should be made between BAT for existing plants and BAT for new ones- and changing from hard and brown coal fired to natural gas fired installations. Also the change from fossil fuel combustion for power generation to other power sources will reduce the \( \text{SO}_2 \) emission. For reductions of rural exceedances large combustion plants are probably the most important source category.
For residential combustion, which can be associated with exceedances at the urban level (not due to rural background levels) changes in fuel type used, decreases of energy demand for heating by applying energy saving building techniques, and applying block heating could reduce SO2 emission considerably. The extent to which emission on a local/urban scale could be reduced varies strongly over the EU region.

For process industry, emission reduction could be achieved by applying Best Available Technology and by improvement of good housekeeping practice. Such reductions should be considered for exceedances in industrial areas (if not due to rural background levels).

It is clear that large scale reduction of emission from large combustion plants will decrease exceedances not only in rural but also in urban and industrial areas. On the other hand, peak concentrations in urban and industrial areas are of a local nature. Consequently an emission reduction of only the local sources may be more cost-effective.

4.3 Benefits arising from the reduction of ambient SO2 air concentrations

In general terms, the reduction of SO2 emissions have an influence not only on the reduction of the effects that are directly due to exposure to SO2, but also the effects due to sulphate and to sulphur deposition. The benefits include the following: decreased risks for health effects, less production loss from crops, reduced stress on forest and natural vegetation, reduced nutrient leaching in sensitive soils, recovery of some acidic lakes, prevention of further acidification of sensitive lakes, reduced deposition impacts on exposed materials, and improved visibility.

For certain effects, such as acute human health effects, changes in air quality will result in immediate changes in effects. For other effect categories, where damage accumulates over time, or interacts with other stress parameters, e.g. forest vitality, total damage will be reduced although the benefits may not be observed immediately.

4.4 Costs of measures

For the cost estimation of measures to reduce large combustion plant (LCP) SO2 emission both primary (process integrated) and secondary (end-of-pipe) measures are relevant. Within the primary measures fuel cleaning, reducing up to 15 - 20% of sulphur content of solid fossil fuels and the installation of Claus plants with 98% abatement efficiency are important options. For the secondary measures, the installation of the lime stone wet scrubbing process (LWS) is the most beneficial option. Although within Europe this process already is applied on a large scale, it will take substantial investments for building new installations or retrofitting already existing plants. Also, the application of this desulphurisation process will result in yearly additional costs for process operation, being more or less comparable with the installation costs.

Within the EU-15 region about 62% of the total amount of energy consumed for residential combustion is covered by natural gas, which has a very low SO2 emission. Liquids represent 26% of total residential energy consumption, while solids account for 11%, more or less equally distributed over hard and brown coal and over biomass - mainly wood. Reduction of SO2 emission from residential combustion of liquids and solids could be realised by changing
to the use of natural gas instead and/or the large scale introduction of district heating and the use of heat pumps. This would mean substantial investments for the construction of gas transportation and distribution networks. Also a more complete desulphurisation of the liquids used for residential combustion would be beneficial as a emission reduction measure.

With respect to the contribution to total industrial process emissions of \( \text{SO}_2 \) the copper production and to a lesser extent the zinc production are the most important processes. The most relevant primary reduction measure for copper production would be the installing of flash smelting and continuous smelting processes instead of the applied conventional processes. This, however, would implicate a very drastic reorganisation of the larger part of the copper production industry. Therefore, secondary measures based on desulphurisation of off-gases by catalytic conversion and adsorption processes seem to be more feasible.

4.5 Conclusions

To reduce rural exceedances, emission reduction measures of large combustion plants are most important. To reduce urban exceedances, large scale residential combustion of liquid and/or solid fuels in the winter period should probably be aimed at, and exceedances in industrial areas could be effectively be reduced by reductions of emissions due to industrial processes. Also, for decreasing the risk of exceeding hourly or daily \( \text{SO}_2 \) concentration limit values, a diversification of reduction measures, based on locally important sources is required.
5.-Reporting the results

Article 11 and Annex 4 of the Framework Directive lay down the information that Member States will have to report to the European Commission. Depending on the levels, the required information may include data on the concentration levels in the zones, the causes of the pollution and other air quality management information. This Chapter focuses on how data on the levels in the zones could be reported to the Commission.

In Chapter 3 it was remarked that the assessment strategy and the requirements for reporting the results of the assessment can not be developed independently. Even more so, the assessment strategy should be directly aimed at generating the results that should be reported. Since the form of the results of the new assessment tools introduced, in particular mathematical models, differs very much from the form of measurement results, the currently existing reporting procedure should be reconsidered.

Until now, the reports of results of air quality assessment in the framework of EU air quality directives have been limited to statistics of measurement results. This is basically a report of the temporal pattern of concentrations at a limited number of points in space (station sites). For reasons of harmonization the European Commission has spent much effort in defining standardized reporting formats.

In addition to the concentration statistics, also an extensive description of the stations is reported to the Commission, including information on the surroundings of the stations, such as the type (urban, suburban or rural), characterization (residential, commercial, industrial, agricultural, natural) and sources. Although this typification gives satisfactory information on the station itself, it does not include any information on how representative the station is for other locations of the same type. Since it is known that Member States apply different measuring strategies, particularly regarding the location of stations with respect to the highest values, it is not possible to extrapolate the reported data to territory-covering information.

In Section 3.3 on measuring strategy it was proposed to add to the information on stations at least additional information on how representative a station is for the type of locations that it belongs to (is it an "average" site, or the worst case).

The Framework Directive allows the use of modelling in zones where the levels are below y% of the limit value and requires reports on these zones every three years. It would be very useful to develop a common form for reporting such modelling results for the future Daughter Directives. This also applies to the results of modelling in areas where the concentrations are above y% of the limit value. When a combination of modelling and measuring is applied, it would be unsatisfactory when the reports to the Commission would be limited to the data of the monitoring stations. The Commission would receive less (though better defined) data in the case of supplementary assessment than in cases without it.

It is proposed to develop a reporting format for the concentrations that includes, besides the statistics of the temporal distribution of measured concentrations, information on the spatial concentration distribution in the zones. There are several options to standardize the reporting of calculated concentrations at other locations than at monitoring sites.
1. Taking the current format as the starting point, the simplest way of reporting might be to report concentrations calculated by mathematical methods for a selection of locations. This form of reporting for hypothetical stations could leave the conventions of reporting unchanged.

2. A more complete reporting option would be to extend the standard way of reporting to the incorporation of more than just point-wise spatial information. One example could be to develop several spatial concentration statistics (analogous to the temporal statistics that are now being reported by monitoring stations). Important examples of this would be the total area above the various limit values and the value of the various concentration parameters spatially averaged over the zone. Data such as the total number of inhabitants in the area above the limit values could also be added.

3. The most complete reporting would be to report the complete spatial concentration pattern in the form of maps, in addition to the statistical information mentioned under option 2.

The first option, reporting for hypothetical stations, would be a solution that hardly uses the added value of models. The third, most ideal option will require the standardization of maps. Although current Geographical Information Systems provide excellent possibilities to standardize the exchange of country-wide concentration maps (generated by interpolation or large scale models), the exchange of maps of small-scale peaks near low point sources (generated by small-scale models and perhaps by statistical methods) may give rise to complications. In any case, a drastic change of the formats for information exchange will be needed. The second option seems to be the most feasible one for the short term, although it would by far not cover all information generated by the mathematical methods. It is therefore proposed to define several area-oriented parameters that should be added to the point-wise parameters given by monitoring stations. It is further proposed to develop in the forthcoming years a format for the exchange of maps and to test it on a voluntary basis. It could be considered for implementation in the Directive at a later stage, possibly according to a time schedule prescribed in the Directive.

Relation with guidance document
The guidance document will define how a supplementary assessment could be used to reduce the number of stations. The procedure is based on the idea that complementing pure measurement with other information will improve the quality so much that less stations are needed to obtain equivalent results. It would be useful to extend the scope of the guidance document and address also the reporting procedure for the assessment after implementation.
A practical point discussed here under is the number of exceedances per year that should be allowed. Arguments favouring no or very few exceedances are the following. For the general public a limit value expressed as a level that is allowed to be exceeded several times is more difficult to understand than a maximum allowed value. Also, a maximum allowed value can be chosen as a direct equivalent of the WHO guideline, while a value that is allowed to be exceeded can only be approximately expressed as an equivalent value - this would be based on measured frequency distributions. The larger the number of allowed exceedances is chosen, the larger the variability in this empirical relation is.

On the other hand, there are strong arguments against expressing the limit value as the maximum. Of all statistical parameters, the maximum concentration is the most variable one. This would mean that a zone may, from year to year, fluctuate in and out compliance with the limit value. Since this variation is often mainly due to meteorological conditions, the compliance state would have a large variation that can not be influenced by air quality management. From the administrative point of view one should attempt to minimize such fluctuations. A second practical reason not to choose the maximum is that the maximum measured concentration can not be measured very reliably. This may be due to instrumental malfunction or to interruptions for maintenance and calibration; anomalous maxima may also occur as a result of unrepresentative sampling during a small period, e.g. because of a very incidental source such as the exhaust of an incorrectly placed truck during a short time.

If for these practical reasons exceedances would be allowed, the choice of the number of exceedances remains. The larger this number would be, the lower the fluctuation and the measuring difficulty would be. The numbers proposed in the Working Group ranged between zero and the number that corresponds to 2% of all hours in a year.

The Working Group did not arrive at a full consensus regarding the question whether any exceedances should be allowed. As a compromise, which could be supported by the majority of the Working Group, it is proposed to define the limit value not as the maximum value, but to allow 24 exceedances of the derived "equivalent" 1 hour limit value over one year.

Consequently, two conversion steps are needed: from 10-15 minutes averages to equivalent 1 hour averages, and from the maximum to an equivalent value that is allowed to be exceeded 24 times. Using empirical information the two steps could also be derived as a combined (total) conversion factor. For both conversion steps one has to take into account that the conversion factors vary from place to place and that the actual ratios fluctuate from year to year. Further the ratio between 10-15 minutes averages to 1 hour averages depends on the statistical parameter that is considered (the maxima have the largest conversion factor, while the averages do not differ).
The temporal frequency distribution of concentrations in an area under the strong influence of a single source tends to be steeper than the distribution of background levels, and so the conversion factor from the maximum to an equivalent value that is allowed to be exceeded tends to be larger for local peaks. The local peaks, being higher than background levels, are more likely to exceed a limit value, and so they are important to take into account, even though the area of such locally elevated levels may be relatively small.

Due to this variability, a conversion factor can at the same time be too strict for one place and too lenient for another place. In particular, if one would chose the equivalent hourly limit value on the basis of the average ratio derived from measuring stations, one would allow exceedances of the WHO guideline at all stations where the actual ratio is larger than the average ratio, so at about 50% of the stations.

If one would take the most conservative point of view, and attempt to set a 1 hour limit value allowed to be exceeded 24 times which would virtually exclude any exceedance of 500 g/m³ averaged over 10-15 minutes, one would have to set an extremely low limit value. Calculations by KEMA (1996) illustrate this for situations where the background concentrations can be neglected. In practical situations the background contribution is often of importance, but one should realize that large ratios can indeed occur. In Austria a ratio of 2.5 was measured between the 10 min. maximum and the 30 min. maximum at a monitoring site strongly influenced by a special industrial plant. This ratio approaches the arithmetically maximum possible ratio of 3. Ratios supplied by Germany and the UK are lower: the ratio between the maximum of 10-15 and 60 minutes average is typically around 1.2.