*Chapter 9*

# Particles

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## 9.1 Particles and their long-range transboundary nature. The situation around 1980

"Particles in air" is a parameter, which is not homogeneous, but consists of a variety of pollutants having quite different physical and chemical characteristics. Particles in air vary in size from ultra fine, just formed particles of a few nanometers aerodynamic diameter, to coarse particles of 10-50 micrometers. The very fine, as well as the coarse particles, have a short lifetime in the atmosphere and are not subject to long-range transport. Submicron particles from 0.1 to 1 μm form the main mass of the long-range transported particles. Particels are emitted to the atmosphere, but also formed via oxidation and via reactions between gases.

The dominant problem connected with particle pollution from earlier than 1980 and to the present days has been the risk for effects on human health, in particular in urban areas and areas near traffic. In former decades, particle pollution was considered as a local pollution problem. Consequently, measurements of particle mass have been included in EMEP only since the late ninties. When EMEP was started, particulate sulphate was included in the programme since it formed a large part of the transboundary flow of sulphur. Later when also nitrogen was included, the transboundary flow of particle-bound nitrate and ammonium was of interest to study. However, for technical measurement reasons the sums of gaseous and particle bound nitrogen compounds, separated into chemically oxidised and reduced nitrogen were chosen as parameters to be measured. Later also particle-bound heavy metals and persistant organic pollutants have been introduced into the EMEP programme.

## 9.2 Emissions of particles

Particles are emitted naturally and from anthropogenic sources. Particles are also formed in the atmosphere via oxidation of gases such as sulphur dioxide, nitrogen oxides, ammonia and VOCs. Natural sources include wind-blown dust from deserts, unpaved roads, seasalt, volcanic sources, and particles formed via biogenic emissions. Anthropogenic sources include combustion (both from mobile and stationary sources), industrial processes, traffic (including resuspension, road and tyre wear), storage and handling of materials.

Particles also contain toxic and persistant compounds (se further chapter 7 and 8) and base cations (see chapter 5). In addition particles contain carbon, both organic and elemental carbon, mineral dust and water. There are no official time series of emission data available for particles in the same way as for sulphur and nitrogen. Only data for 2000 and 2001 are so far reported to the EMEP emission database.

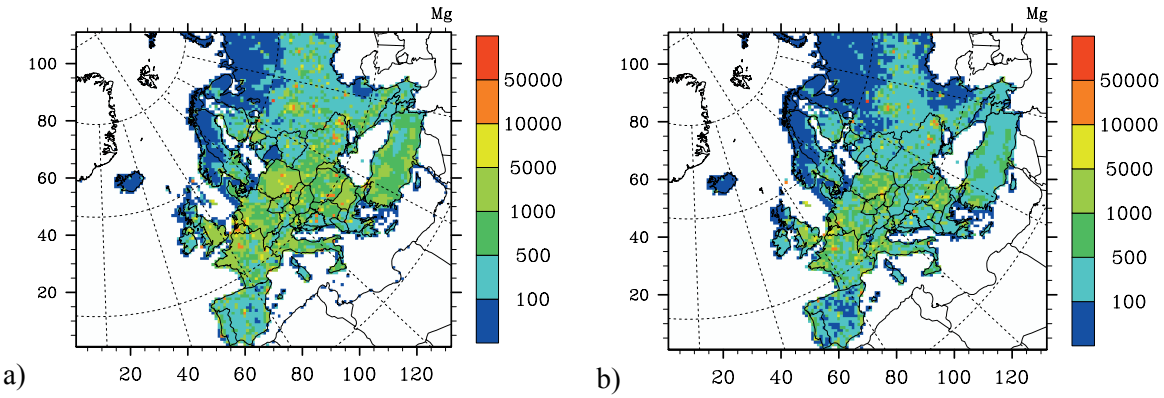



**Figure 9.1** Spatial distribution of particle emissions a) $PM_{10}$ and b) $PM_{2.5}$ in 2001 (Kahnert et al., 2003). Units: Tonnes/year/grid square.

Some countries have studied the long-term emissions of particles and included them in their national EMEP assessments; Germany and Slovak Republic, See figure 9.2. The emission data for particles show a decrease by a factor of approximately 5 between 1990 and 2000.

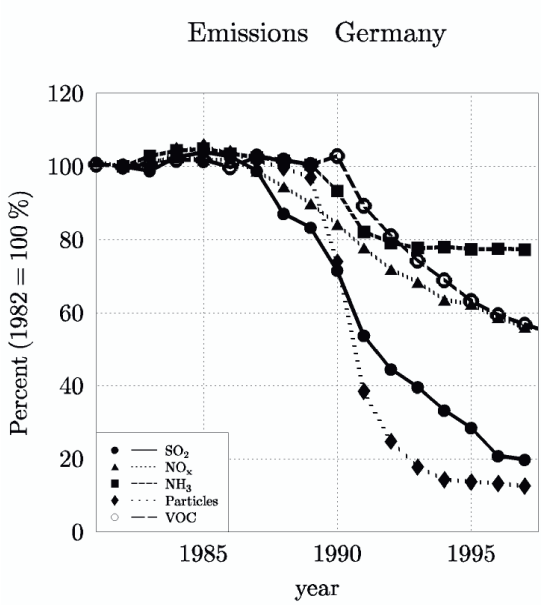



**Figure 9.2a** Emissions of particles in Germany, relative decrease from 1980 to 2000. Units: % in relation to 1980. (Wallasch 2004)

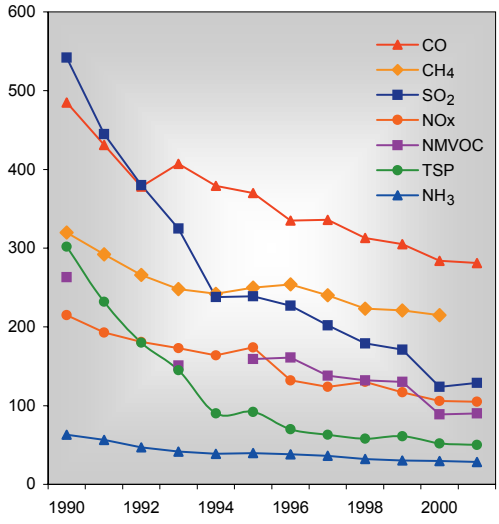



**Figure 9.2b** Pollutants emissions, including emissions of total suspended particles (TSP) in the Slovak Republic. Units: ktonnes/year. (Mitošinková et al., 2004)

A number of studies made in the 1980s showed a considerable decrease in particles emissions in many of the western European countries and the USA, as a consequence of introducing emission standards for several sources including power production and road traffic. Fuel shifts from coal to oil and gas contributed also to declining emissions. In addition, to improve urban air quality, the small-scale coal burning for residential heating was substituted by other energy sources, such as oil and gas and district heating.These changes were mainly achieved in the seventies and eighties.

## 9.3 Particles in European air

Besides the risk for human health effects, aerosols have an impact on climate including radiative effects and visibility. The levels and trends of particle concentration are consequently important to follow. Particle measurements as $PM_{10}$ were introduced in EMEP in 1998 and a number of countries have now started measurements. In 2000 26 stations in Germany, Spain and Switzerland reported particle data to EMEP. In 2002, measurements were initiated in another 11 countries. The number of sites measuring particles is expected to increase further in the nearest future, partly driven by EU legislation. In addition to $PM_{10}$, measurements of $PM_{2.5}$ are made in a number of countries. Currently $PM_1$ is not being measured routinely, but this parameter is of considerable relevance inter alia for modelling, because PM1 is less affected by local resuspension.

The data on $PM_{10}$ concentrations at rural sites are so far few, at least from what is reported to international databases. From 2000 and 2001 some data are reported, Figure 9.3. The measurements show that the annual average levels at many EMEP and other rural sites are around 20 µg/m$^3$, or even higher, which is equivalent to the indicative limit value of the first EC daughter directive on air quality (1999/30/EC). It should also be stressed that a recent assessment of the World Health Organisation has indicated that there is no threshold below which particles have no effect on human health.

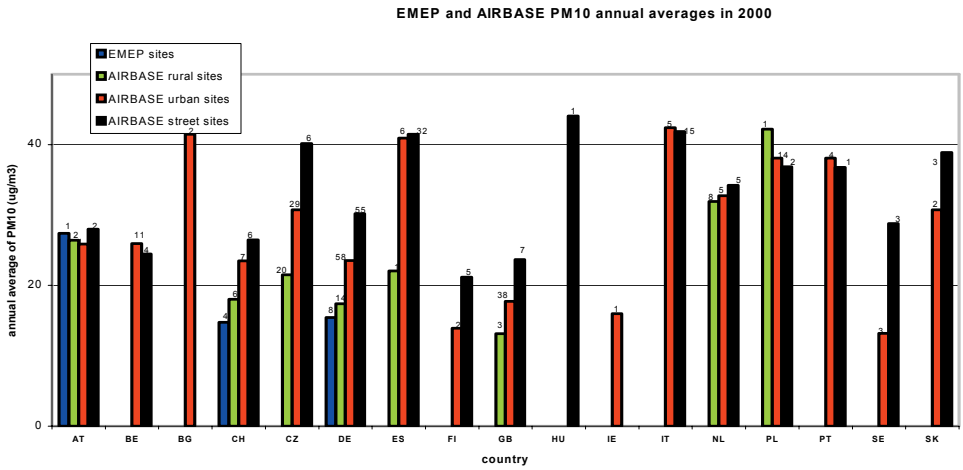



**Figure 9.3a** Annual country averages of $PM_{10}$ at EMEP stations, other rural sites, urban sites and AIRBASE street sites in 2000. Numbers over each bar indicate the number of observation stations contributing to each country average. Units: µg/m$^3$ (Tsyro et al., 2003)

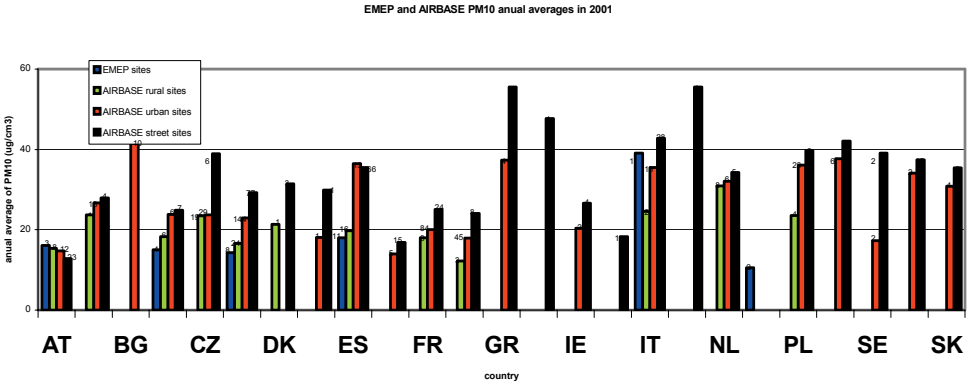
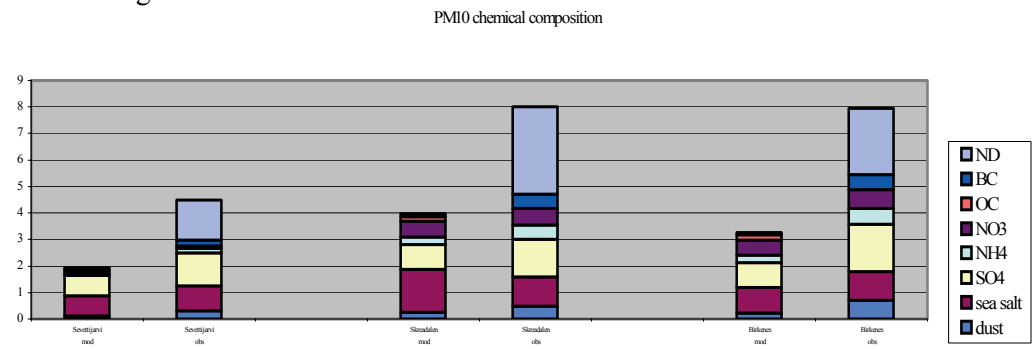



**Figure 9.3b** Annual country averages of $PM_{10}$ at EMEP stations, other rural sites, urban sites and AIRBASE street sites in 2001. Numbers over each bar indicate the number of observation stations

contributing to each country average. Units: µg/m$^3$ (Tsyro et al., 2003)

Particles are not only described by the particle mass concentrations of different size fractions. Data on the chemical composition provide essential information e.g., for source apportionment and model validation. About a third to half of the particle mass consists of inorganic compounds such as sulphate, nitrate and ammonium at European background sites. Dust can be found at high concentrations at sites in southern Europe and seasalt near the sea. Current regional air quality models tend to udnersatimate the total PM mass. This may be due to sources unknown or not included in the model-calculations (like dust events), understimations in the emission inventories and uncertainties and artefacts in the measurements. Further measurements are urgently needed to improve the knowledge on the particle composition.

Natural background




Rural air

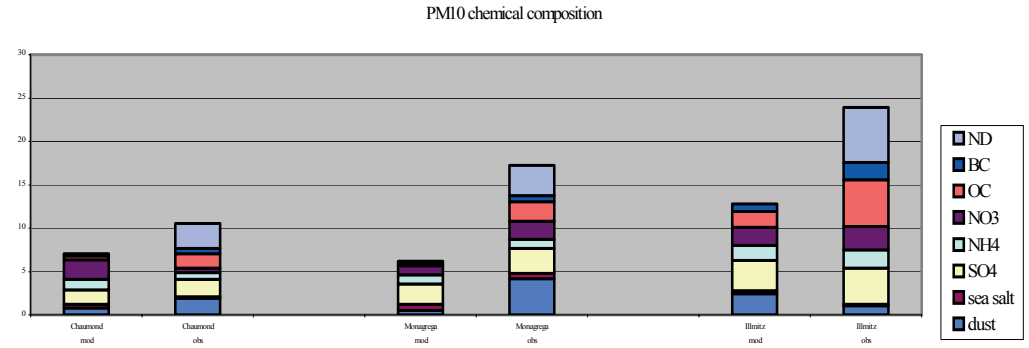



**Figure 9.4** Measured and model-calculated $PM_{10}$ concentrations in natural background and rural air and its composition: ND=not determined, BC=base cations, OC= organic carbon, NO3, NH4 and SO4, sea salt and dust (Tsyro et al., 2003) Units: µg/m$^3$

Since the reported data and time series are few, it is difficult to evaluate trends of PM. In the EMEP programme the inorganic components sulphate, nitrate and ammonium have been measured and the trends of these can say something about the development in particles. There is a good correlation between $PM_{10}$ mass concentration and secondary inorganic compounds. However, the inorganic fraction usually accounts for only 30 to 50% of the total particle mass (Figure 9.4), so the trends for these components cannot give the trends in particle mass.

In a number of countries, the concentrations of particles have been measured nationally at background and rural air quality monitoring stations. A large number of different parameters were measured using different methods, even if data are not reported to the EMEP database. Total suspended matter (TSP) has been measured in countries including Germany, Slovak Republic, Spain and Switzerland. In other countries, such as Sweden, soot has been measured as a particle indicator at the EMEP sites.

Measurements of TSP are available since the early eighties from a number of Swiss sites. $PM_{10}$ measurements started in 1997 and during 1997-98 parallel TSP and $PM_{10}$ sampling was made in order to evaluate the relation of TSP and $PM_{10}$. For rural sites a very stable $PM_{10}$/TSP of 0.88 (±0.02) was observed for annual means (Gehrig and Hofer, 2000). This allowed an estimation of $PM_{10}$ trends based on the TSP data before 1997. Over all, a decreasing trend is observed.

Sulphate, nitrate and ammonium form a considerable part of the transboundary particle pollution. Reduced emissions of these gases will decrease the levels of particles in the air. As mentioned in the earlier chapters on sulphur and nitrogen compounds, this decrease is not linear in relation to the emission decrease.

Co-located parallel measurements of $PM_{2.5}$ and $PM_{10}$ were made at 7 Swiss sites since January 1998 (Figure 9.5), (Gehrig and Buchmann, 2004). For sites within the Swiss plateau the $PM_{2.5}$ concentrations are highly correlated. The long-term averages of the $PM_{2.5}/PM_{10}$ ratios vary only from 0.75 to 0.76 (with the exception of the traffic-exposed site of Bern, 0.59). The correlation between the daily values of $PM_{2.5}$ and $PM_{10}$ is high at all sites. $PM_{10}$ as well as $PM_{2.5}$ exhibit characteristic seasonal trends (in line with $SO_2$ and $NO_2$) with elevated concentrations during the cold season. This is the case at all sites, with the exception of the elevated sites (Chaumont and Jungfraujoch). The main reasons are meteorological effects of frequent inversions during winter and good vertical mixing during summer. In contrast, the elevated sites show the lowest values in winter. They are, especially during wintertime, often situated above the inversion layer and thus protected from the emissions of the lowlands.

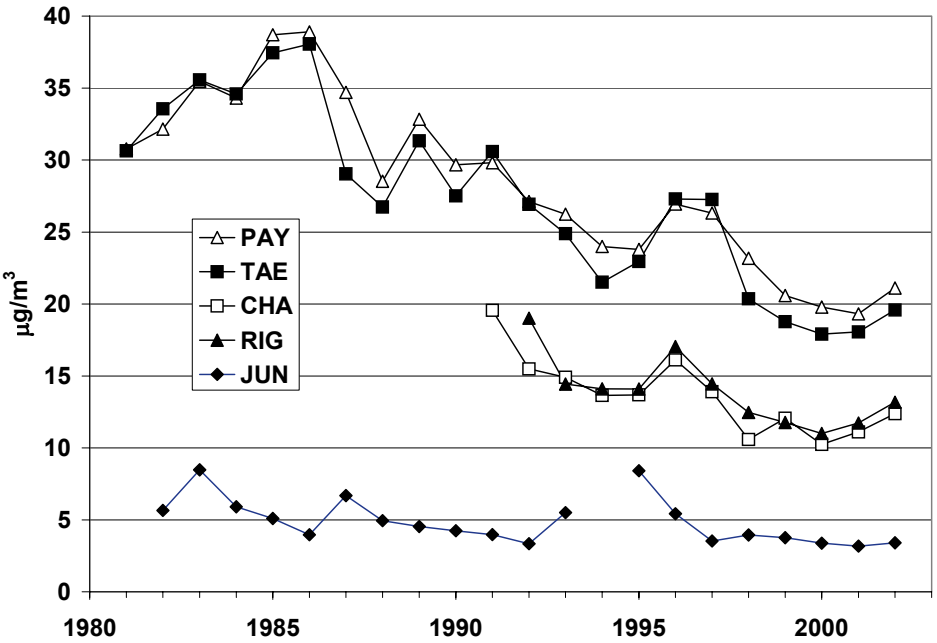



**Figure 9.5** Long-term PM10 trends at the Swiss EMEP sites (Ballaman et al., 2004).

The Swiss seasonal variation for $PM_{10}$ is different from that reported for 2000 from Birkenes in Norway, where low concentrations occur in winter, and higher concentrations in spring and autumn. The variations are likely to vary due to the contributing sources. Local soil dust could be expected to be low in northern Europe during the cold and wet season. However, local contributions from combustion would give the highest concentrations during the cold season. More data are needed to evaluate particle sources and is a necessary input for control strategies.

Particle mass concentrations have been an integral part of the measurement program at German EMEP sites since the start (Figure 9.6). Both the PM mass concentration and the concentration of

particulate sulphur show significant decreasing trends. The particle mass has decreased approximately by a factor of 2.5, and the particulate sulphur approximately by a factor of 3 since 1980.

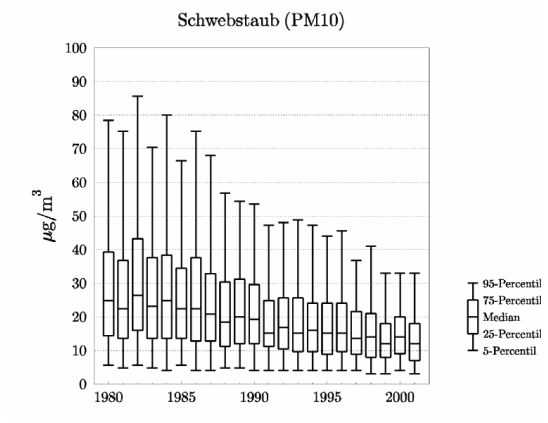



**Figure 9.6a** Particle concentrations. Results from 8 sites. (Wallasch 2004).

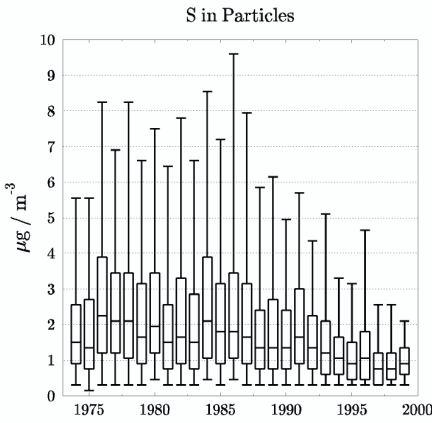



**Figure 9.6b** Sulfur in particles. Results from 8 German sites. Wallasch 2004).

TSP has been measured also in Spain (Fernández et al., 2004) where relatively high concentrations are observed compared to the levels in central and northern Europe. The high levels of TSP are due to dust contributions from the Saharan desert and other arid and semi-arid areas. The highest concentrations are seen at sites subject to natural dust sources. The natural contribution is also seen as a higher TSP/$PM_{10}$ ratio than at urban sites. No trend is observed, as could be expected, when natural sources dominate.

In the Slovak Republic (Mitošinková et al., 2004), the regional average concentrations of particles as TSP range between 15 and 45µg/$m^3$, depending on location and altitude. The highest levels are observed in the dry and windy lowlands. The influence of the local agricultural activities is obvious. The lowest concentrations are observed for the alpine sites.

In Sweden, a trend decreasing by a factor 2-3 between 1980 and 2000 is seen for the measured soot concentrations at the EMEP sites in southern Sweden (Figure 9.7). In northern Sweden the levels are frequently below the detection limit (Lövblad et alö., 2004).

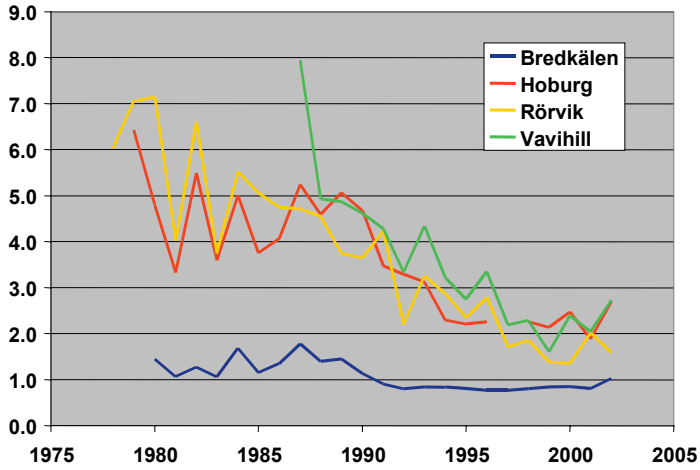



**Figure 9.7** Trends of particle pollution at four Swedish EMEP sites measured as soot (black smoke). Units: µg/$m^3$. (Lövblad et a., 2004)

## 9.4 Need for further work on particle pollution

Current levels of particulate matter (PM) pose a considerable threat to human health in Europe. Even in urban areas, long range transport of PM contributes strongly to PM levels. To protect human health it is necessary to abate emissions of PM and its gaseous precursors (SO2, NOx; NH3 and NMVOC) and decrease the levels in air. For this reason additional data are needed on particle emissions (including speciation) and concentrations in ambient air (measured with standardised methods to make comparisons between sites possible and meaningful). The EMEP model will be gradually extended and improved, e.g. by including additional process like formation of secondary organic aerosols from VOCs. In addition, measurements of different size fractions, chemical composition and other PM characteristics are needed to increase our knowledge about particluatre matter and its effects on health and climate.

## 9.5 Conclusions

- Significant downward trends are seen for particles in the air. In addition it is expected from earlier studies that a considerable decrease was achieved already before 1980.

- Long-range transported particles contribute considerably to the levels, also in urban areas, and are likely to contribute to the risk for human health.

- Efforts to quanitfy emissions of different PM fractions (including PM10 and PM2.5), ideally including chemical composition, have to be intensified.

- Quality assured and comparable data on particle levels are needed in a much greater extent than what is available today. Particles as $PM_{10}$, $PM_{2.5}$ and even $PM_1$ will be needed in the future EMEP programme. In addition, chemical characterisation, size distributions etc. are needed to learn more about the complex particles.

- The unified Eulerian EMEP model will be gradually improved and extended to allow a full mass closure.

## 9.6 References

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