

Contributions of Road Traffic to Ambient PM10 and PM2.5 Concentrations

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Abstract

Recent epidemiological studies have shown that elevated concentrations of fine suspended particles can cause adverse health effects for human beings. Therefore, the Swiss clean air act (Luftreinhalteverordnung, LRV) has set new limit values for PM₁₀ (particles with an aerodynamic diameter <10µm). Measurements performed within the framework of the Swiss Monitoring Network (NABEL) show that the limit values are not respected in large parts of Switzerland.

In order to develop effective reduction scenarios the sources of the fine particles need to be known as well as their contribution to the ambient air concentrations. Up to now the sources of PM₁₀ and PM_{2.5} were not sufficiently identified and quantified in Switzerland. It was, therefore, the main goal of this study to identify the most important sources of fine particles and to quantify their contributions to the ambient air concentrations.

The elevated concentrations of fine particles near busy roads indicate that road traffic is an important source for atmospheric particles. There are different causes for road traffic induced fine particles. Primary particles are emitted from tile pipes as well as gaseous precursors for secondary particles. In addition the road traffic contributes to the fine particle load of the atmosphere by abrasion and resuspension of particles (street dust, tyre wear, break wear).

A multivariate receptor model was applied to estimate the contribution of road traffic to ambient levels of fine particles (PM₁₀) at different locations in Switzerland. At two roadside sites with heavy local traffic, the road traffic was found to account for 46% and 64% of PM₁₀. At an urban background site, the estimated average road traffic contribution was 34%, whereas a slightly higher value was obtained at a suburban site (36%). This results are in good agreement with the findings of a recent study, where a conceptually different approach (dispersion modelling) was applied.

Keywords

Air pollution – Particles – PM₁₀ – PM_{2.5} – Swiss Transport Research Conference – STRC 2001 – Monte Verità

1. Introduction and goal of the study

There are indications from recent epidemiological studies that elevated concentrations of fine suspended particles can be responsible for adverse health effects for human beings [WHO/UN 1999]. These findings were taken into account in the last revision of the Swiss clean air act (Luftreinhalteverordnung, LRV) with new limit values for PM₁₀. The measurements within the framework of the Swiss National Air Monitoring Network (NABEL) show that the limit values are not respected in large parts of Switzerland. In addition, there is at present an increasing interest in the even finer particle fraction PM_{2.5}. Though there are no limit values for PM_{2.5} in Switzerland so far, there are clear indications for adverse health effects related to the PM_{2.5} fraction as well. In order to develop effective reduction scenarios the sources of the fine particles need to be known as well as their contribution to the ambient air concentrations. Up to now the sources of PM₁₀ and PM_{2.5} were not sufficiently identified and quantified in Switzerland. It was, therefore, the main goal of this study to identify the most important sources of fine particles and to quantify their contributions to the ambient air concentrations.

The elevated concentrations of fine particles near busy roads as measured in the NABEL network show that road traffic is an important source for atmospheric particles. The total emissions of road traffic are caused by different contributions. From tile pipes primary particles are emitted as well as gaseous precursors for secondary particles. In addition the road traffic contributes to the fine particle load of the atmosphere by abrasion and resuspension of particles (street dust, tyre wear, break wear).

Due to the very complex composition of atmospheric particles the measurement techniques are demanding. The conventional sampling methods (e.g. the manual gravimetric procedure used in this study) are subject to sampling artefacts as the loss of semivolatile constituents during sampling, transport, storage and conditioning. This can be in particular the case with ammonium nitrate, organic compounds and adsorbed humidity. On the other hand, there can be artefact formation of particle mass on the filters by reaction with gaseous substances. Quantitatively, this effect is less pronounced than the losses. From a scientific point of view these artefacts are of course not desirable. For the main purpose of this study, however, this problem is not relevant because the epidemiologic studies and the air quality standards derived from those studies are based on data which were obtained with the same sampling procedures and, therefore, were subject to the same artefacts.

2. The key results at a glance

The main goal of the study was to quantify the contribution of road traffic to the ambient air concentrations of PM10 and PM2.5 at four carefully selected sites in Switzerland which represent important air pollution scenarios. For this purpose, the chemical composition of ambient particle samples were analyzed in detail and the influencing factors (sources) were determined by means of multivariate statistics (receptor model). In addition, the contribution of the heavy duty traffic to the ambient fine particle concentrations was investigated by comparing parallel continuous monitoring data from a site directly at a very busy road and a nearby urban background site. It turned out that at moderately polluted urban and suburban sites PM10 consists roughly of one third carbonaceous aerosols (soot and organic compounds) one third secondary inorganic aerosols (sulphate, nitrate and ammonium) and one third mineral particles and humidity. Table 1 shows the contributions of the road traffic (incl. the traffic related part of the secondary aerosols and the presumably small contributions of the offroad traffic at these sites):

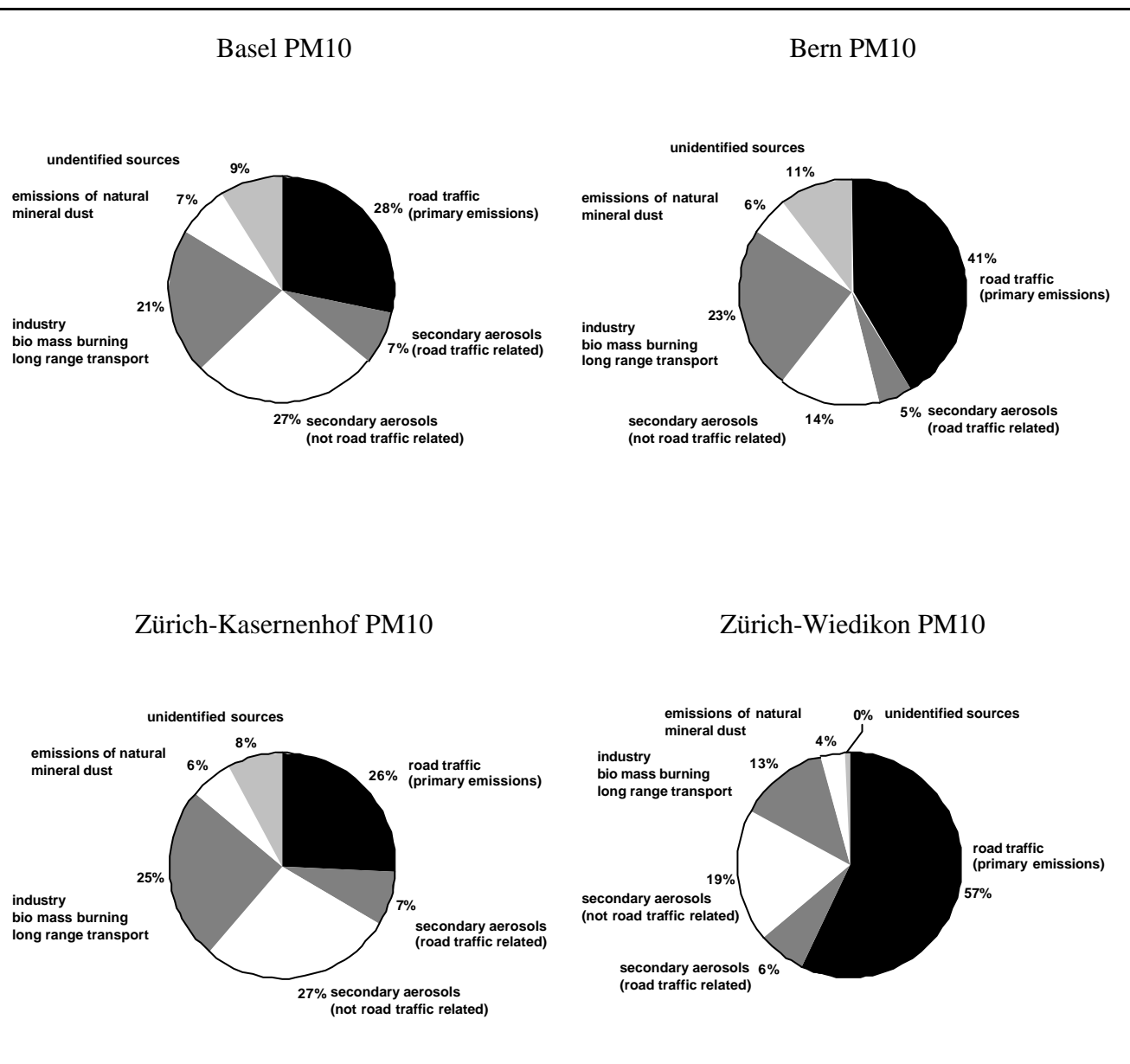
Table 1: Annual mean road traffic contribution to PM10 and PM2.5 at the investigated sites

Site	Parameter	Site characteristics	PM Measurement ($\mu\text{g}/\text{m}^3$)	Mean road traffic contribution ($\mu\text{g}/\text{m}^3$)	(%)
Basel	PM10	suburban	25.0	9.5	35.3
Bern	PM10	city centre, kerbside	39.6	18.9	45.7
Zürich-Kasernenhof	PM10	city centre, court yard	24.5	8.6	33.3
Zürich-Wiedikon	PM10	city centre, kerbside	42.6	26.8	62.9
Bern	PM2.5	city centre, kerbside	24.1	9.2	36.8
Zurich-Kasernenhof	PM2.5	city centre, court yard	20.7	5.4	26.0

A differentiation of these contributions according to direct tail pipe emissions and abrasion/resuspension emissions was not possible. Tracer analyses showed that, with the exception of kerbside sites, tyre wear is quantitatively not important. However, additional campaign measurements at Zürich showed the importance of the heavy duty traffic. With 8% heavy duty

vehicles at the investigated site a contribution of roughly 50% to the local primary PM10 road traffic emissions resulted. Figure 1 gives an overview of the contributions of different sources and source categories to the ambient air concentrations of PM10.

Figure 1: Contributions of the identified sources and source categories to PM10 at measuring sites with different pollution levels.



3. The method of source attribution with receptor models

The source attribution of the ambient air concentrations of fine particles was conducted with a receptor model. Application of receptor models requires knowledge of the chemical composition of atmospheric particles at a certain measuring site (receptor), i.e. the concentrations of a large number of chemical constituents in a sufficient number of samples are needed. All receptor models are based on the assumption of mass conservation. However, there are two different realisations of this approach: chemical mass balance (CMB) and multivariate receptor models [Hopke 1991]. In addition to data on the chemical composition of atmospheric particles at the receptor, the chemical mass balance uses quantitative information of the chemical composition of PM_{10} emitted by the sources (source profiles) to solve the mass balance equation, i.e., to estimate the source contributions. In contrast, multivariate receptor models use only the chemical composition data of atmospheric particles at the receptor to estimate both, the source profiles and the source contribution. The basic idea is that chemical constituents from the same source will be correlated and the correlation structure of the data set can be used to solve the mass balance equation. The solution of the mass balance equation by multivariate receptor modelling can be considered as a linear coordinate transformation, where the dimensionality of the data set (number of observed chemical species) is reduced to a smaller number of so-called factors. These factors might then be interpreted in terms of source profiles. In this work the multivariate receptor model approach was selected. The applied model was developed at the Seminar for Statistics at the Federal Institute of Technology (ETH) in Zurich.

Secondary atmospheric particles which are formed by chemical transformation of gaseous precursors cannot be included in receptor models. Therefore, source contributions to secondary PM_{10} were assessed by combined evaluation of transboundary import export budgets [EMEP 1999] and an emission inventory of the precursor gases [CORINAIR 1997].

4. Measurement programme, methods and selection of sites

For a successful application of a receptor model for the identification of the emission sources of fine particles a sufficient number of measurements of suitable constituents (indicator variables) are needed. Within the scope of this study samples of PM₁₀ and PM_{2.5} from the different measurement sites were extensively analysed.

The following analysis were performed (in parenthesis, the applied analytical method is indicated):

- Water soluble inorganic ions NH_4^+ , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , Cl^- (Ionchromatography). The sum of the concentrations of NH_4^+ , NO_3^- and SO_4^{2-} represents the secondary inorganic particle fraction.
- Elemental and organic carbon EC and OC (coulometric carbon analysis [VDI 1996]).
- Trace elements Al, As, Ba, Br, Ca, Ce, Cd, Cu, Fe, Ga, K, La, Mg, Mn, Mo, Nd, Ni, Pb, Rh, Sb, Se, Tl, V, Y (plasma mass spectrometry, ICP-MS).
- Polycyclic aromatic compounds (Laser Two-Step Mass spectrometry, L2MS).
- Biogenic particles (bio assays, microscopy)
- Tyre wear (GC-MS)

In addition to the investigations with the receptor model special measurements with high time resolution were performed at the highly traffic related site Zurich-Wiedikon. From these measurements the contribution of the local road traffic to PM₁₀ was determined and information could be obtained about the partitioning of the traffic contributions into light and heavy duty traffic.

The sampling was performed within the scope of the particle measurements of the Swiss National Air Pollution Network (NABEL). During the period from 01.02.98 to 31.03.99 a sufficient number of PM₁₀ and PM_{2.5} samples from the NABEL sites Basel, Bern, Chaumont and Zürich-Kasernenhof was chemically analysed in detail. The sites were selected according to important and representative pollution scenarios in Switzerland (Table 2).

During two measurement campaigns in July/August 1998 and January/February 1999 PM₁₀ samples from an extremely traffic-influenced site of the Amt für Gesundheit und Umwelt of

the city of Zürich in Zürich-Wiedikon were collected and analysed. In addition in spring and autumn 1999 PM10 samples from the NABEL site Payerne were analysed. Payerne represents a rural situation of the Swiss plateau.

High volume samplers (Digitel DHA-80) with a PM10 or PM2.5 sampling inlet (impactor) were used for sampling. During 24 hours 720 m³ of ambient air were drawn through binder-free quartz fibre filters (Schleicher und Schuell QF20, 150mm diameter). This sampling procedure is compatible with the reference method according to EN 12341.

Table 2: Overview of the investigated sites

Site	Site characteristics	Parameter	Number of samples
Basel	suburban	PM10/PM2.5	120/82
Bern	city centre, kerbside	PM10/PM2.5	120/82
Chaumont	rural, above 1000 m asl.	PM10/PM2.5	120/82
Zürich-Kasernenhof	city centre, in courtyard	PM10/PM2.5	121/85
Zürich-Wiedikon	city centre, kerbside	PM10	59
Payerne	rural, below 1000 m asl.	PM10	17

5. Chemical composition of the fine particles

5.1 Analysed constituents and mass balance

The measurement programme mentioned above comprised all components of the fine particle fraction which contribute considerably to the mass. The sum of all analysed components should therefore equal the gravimetrically determined mass. Figure 2 shows the composition of PM10 and PM2.5 for 3 typical exposition types.

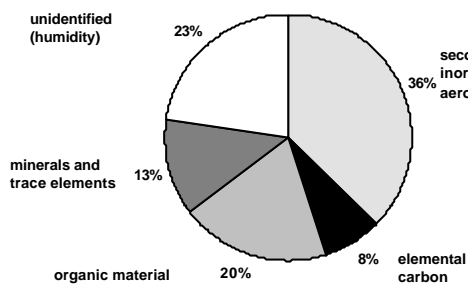
5.2 Water content and biogenic particles

The unidentified part (see figure 2) can mainly be attributed to the water content of the particles which is weighed in the gravimetric procedure but not involved in the analytical programme and to biogenic particles which are not always correctly quantified with the used coulometric method for carbonaceous aerosols. Spot measurements of the humidity of selected samples showed a mean water content of 16% in the warm season and 6% in the cold season. Specific analysis of the mass of biogenic particles turned out to be very difficult. The used methods did not allow for clear quantifications. The Bradford assay had a too high detection limit, the protein contents of all investigated samples were below the limit. From the known detection limit and the assumption of an average protein content of biogenic particles it can be concluded that the concentrations of biogenic particles in the investigated samples were always below $3.5 \mu\text{g}/\text{m}^3$.

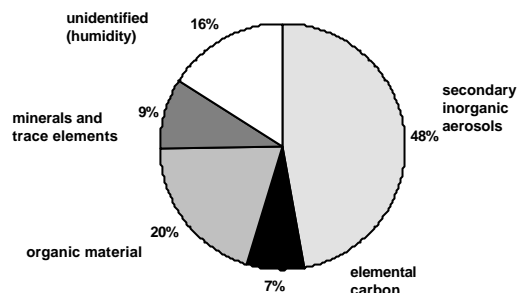
However, microscopic analysis of a limited number of samples (8 samples per site) showed clearly the presence of biogenic particles. As expected, the number of the identified biogenic particles was higher during the vegetation period. Conversion of particle numbers to particle mass are generally very difficult because a lot of uncertain assumptions about shape and density of the particles have to be made. A rough estimate leads to a mean mass concentration for biogenic particles of approximately $1\text{--}1.5 \mu\text{g}/\text{m}^3$, and a maximum daily value of $6 \mu\text{g}/\text{m}^3$

Figure 2: Chemical composition of PM10 and PM2.5 at Basel (suburban), Bern (kerbside) and Chaumont (rural)

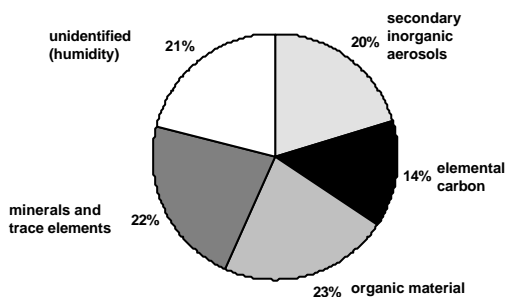
Basel PM10



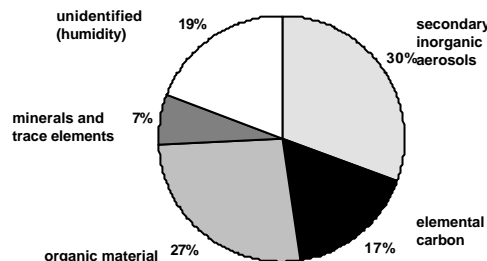
Basel PM2.5



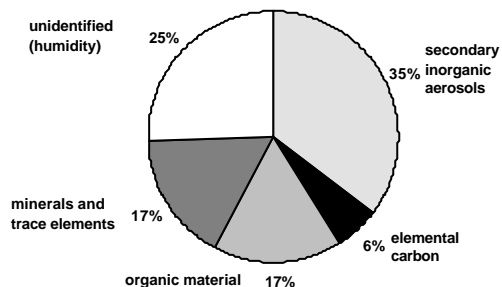
Bern PM10



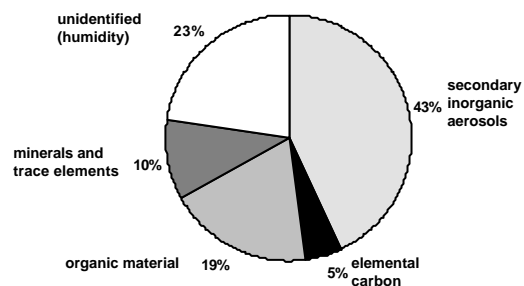
Bern PM2.5



Chaumont PM10



Chaumont PM2.5



6. Source attribution of PM10 and PM2.5

6.1 Source attribution for primary aerosols

The attribution of the primary aerosols was performed with a receptor model. As a result the applied model gives factors which explain a certain amount of the total variance of the constituents of the aerosols. From the knowledge of typical combinations of constituents for certain emission sources and the comparison with the composition of the found factors, sources or groups of sources can be attributed to the statistically obtained factors. In addition to these so called source profiles, also the source activities can be estimated. From the relation of the source activities with different weekdays or seasons and from the enrichment factors additional informations for the identification of sources can be obtained.

With this method the following sources and source groups could be attributed to the factors which were obtained from the receptor model: Waste incineration and bio mass burning, thallium containing dust (cement production?), road traffic (mainly tail pipe emissions), street dust, street dust containing salt. It is advisable to summarize the latter three factors to a common source "road traffic" because a complete separation of the factors seems to be unlikely due to the high correlation between these different road traffic contributions. Figure 1 gives an overview of the estimated mean annual contributions of all identified sources and source groups for the investigated sites.

6.2 Source attribution of secondary inorganic aerosols

The source attribution of the secondary inorganic aerosols is based on the data of the European emission inventory for the gaseous precursors NH_3 , NO_x and SO_2 [CORINAIR 1997] and the long range transport modeling of the secondary inorganic aerosol fraction within the scope of the EMEP programme [EMEP 1999]. Agriculture is the main source of ammonia and contributes therefore considerably to the ambient air concentrations of ammonium. Road traffic is the main cause for nitrate formation. Emissions of fossil power plants and other industrial and non-industrial combustion processes as well as biogenic emissions are important emitters of SO_2 and contribute therefore to the sulphate formation. Based on the data of precursor emissions the contribution to the secondary inorganic PM10 can be estimated for the investigated sites.

The model results of EMEP agree quite well with the measurements performed in this study. For Switzerland a annual mean value for the concentrations of secondary inorganic aerosols of 10-15 $\mu\text{g}/\text{m}^3$ was calculated. Our measured values at Basel, Bern and Zürich are somewhat below 10 $\mu\text{g}/\text{m}^3$. It has, however, to be kept in mind that the measurements underestimate the true ammonium and nitrate concentrations due to sampling losses.

6.3 Secondary organic aerosols

The measurements performed within the scope of this study show that secondary organic aerosols do not contribute significantly to the PM10 (and PM2.5) mass considering long term means. Elevated concentrations can be observed on specific days with high photochemical activity. From a detailed analysis of a photochemically active episode in Zürich a contribution of secondary organic aerosols of up to 3 $\mu\text{g}/\text{m}^3$ could be found. No attempt to attribute these generally small amounts to certain sources was made.

6.4 Contribution of tyre wear

In tyres organic compounds like N-Cyclohexyl-2-benzothiazolamin (NCBA) can be found which stem more or less exclusively from vulcanized rubber. These compounds are not easy to analyse, but they are very specific tracers for tyre wear. The contribution of tyre wear can directly be estimated from the measured concentrations in aerosol samples if the mean content of the tracer in the tyres is known.

Analysis of the contents of NCBA in a selection of 12 frequently used tyre brands were compared with the contents of NCBA in PM10 samples from Bern, Zürich, Tänikon and Jungfrauoch. Table 3 gives the results of the estimations for the contribution of tyre wear in PM10. It shows also quite a high uncertainty for the contributions of tyre wear to PM10. This is due to a high variation of the NCBA content in the different tyres. In addition, only a relatively small number of samples could be analysed due to the very complex and expensive analytical procedure. Therefore, the results have to be considered as indicative estimates. Nevertheless, they show that tyre wear is quantitatively only important at sites which are directly influenced by nearby road traffic (kerbside sites). In the mass balance for PM10 the tyre wear contributions are contained in EC and OC, for the source attributions in the factors road traffic and street dust.

Table 3: Contribution of tyre wear in PM10

Site	Number of analysed samples	Tyre wear in PM10 (%)	95%-confidence intervall (%)
Bern	6	7.5	5.1-14.0
Zürich-Kasernenhof	7	1.9	1.3-3.6
Tänikon	4	< 1	
Jungfraujoch ^a	4	< 1	

^a Total suspended particulate (TSP) measurements instead of PM10

6.5 Overview of the source attribution for ambient air particle concentrations

Table 4 shows the attribution of sources of the fine particle mass (PM10) to the factors obtained from the receptor model. The receptor model does not give exactly the same factors (resp. separation of the sources) for all sites, which makes the direct comparison of the sites somewhat uncomfortable. Though for Chaumont only daily samples with $PM_{10} > 8 \mu g/m^3$ were included into the statistical analysis no detailed separation of the factors could be obtained. Therefore, the contribution of the road traffic could not be quantified at this site.

Within the limits of uncertainty the sums of the calculated contributions of the factors, indicated in the 2nd line compare quite well with the measured values (1st line in the table).

The factors in connection with road traffic are added to get the total contribution of road traffic. An overview of the obtained results (including the traffic related contribution to the secondary aerosols) can be found in Table 1.

With the applied receptor model it was not possible to separate the contributions of different traffic categories. It is almost certain that the road traffic contributions obtained with the receptor model include also contributions of other traffic sources like offroad traffic and small amounts of emissions from air and ship traffic. The unsatisfactory separation of sources is a general problem involved with the use of receptor models for source attribution. In order to obtain a complete separation of the source contributions, specific tracers would be needed for all sources which in addition must be stable during the transport from the source to the re-

ceptor. Such tracers, however, are very difficult to find and often even more difficult to analyse.

Table 4: Source attribution of the mean PM10 ambient air concentrations ($\mu\text{g}/\text{m}^3$) to the factors obtained with the receptor model. For the site Chaumont only days with $\text{PM}_{10} > 8 \mu\text{g}/\text{m}^3$ were used

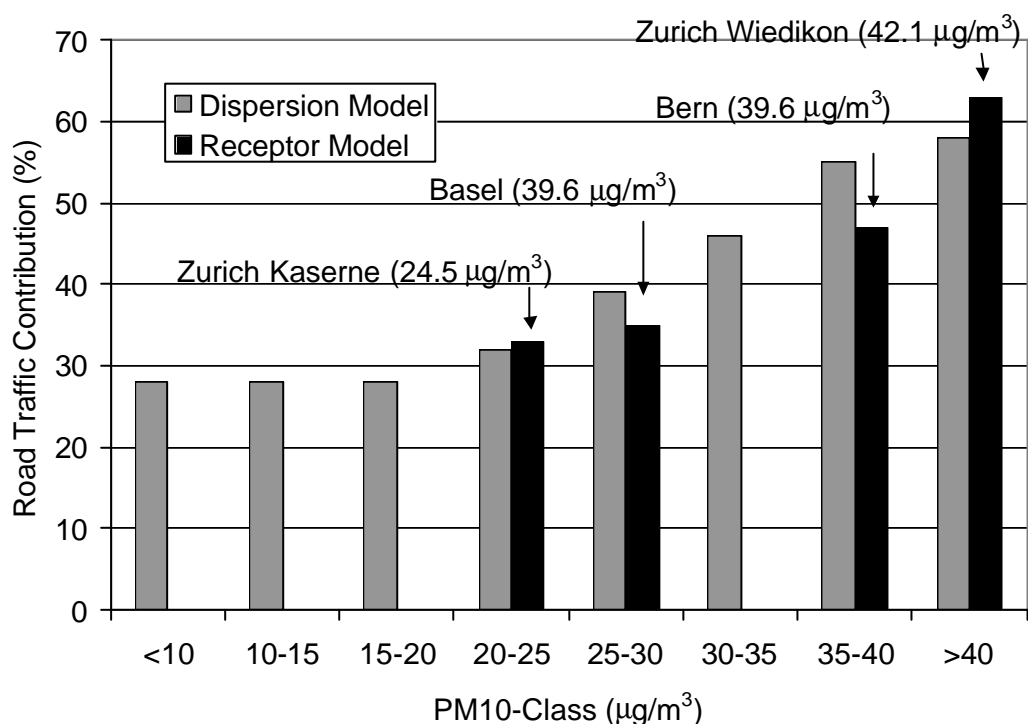
	Basel	Bern	Chaumont	Zürich- Kasernenhof	Zürich- Wiedikon
Total PM10 (measured)	25.0	39.6	16.5	24.5	42.6
Total PM10 (receptor model)	26.9	41.4	16.4	25.8	42.6
Tail pipe emissions of road traffic (incl. offroad)	4.9	7.6		3.4	17.8
Road dust (as far as not contained in the factors tail pipe emissions and road salt)	1.8	6.4		2.5	3.2
Road salt (incl. simultaneously re- suspended road dust)	0.9	3.1	0.3	0.8	3.3
Waste incineration, biomass burn- ing	2.7				
Waste incineration, biomass burn- ing, long range transport		8.2		4.8	5.4
Long range transport, Tl-factor ^a	2.9				
Tl-factor ^a		1.4		1.6	
Mineral dust (not traffic related))	2.0	2.3	2.7	1.6	1.6
Diverse anthropogenic sources			4.3		
Other non identified sources	2.4	4.4	3.6	2.0	0.2
Secondary aerosols	9.3	8.0	5.5	9.1	11.1
traffic related part	1.9	1.8	0.5	1.9	2.5

^a Probably from cement production

6.6 Comparison of the results of the receptor model with dispersion modeling

Source contributions of air pollutants can be obtained with two different approaches, with receptor models and with dispersion modeling. In a recent BUWAL study a dispersion model for ambient PM₁₀ was presented [BUWAL 1999; Filliger et al. 1999]. It allows to estimate the road traffic contributions at sites with different PM₁₀ levels. Figure 3 compares the results of the dispersion model with our findings from the receptor model. It shows a good comparability of the receptor and the dispersion model

Figure 3: Comparison of total road traffic contributions to PM₁₀ as determined by two independent methods: Dispersion modelling [BUWAL 1999; Filliger et al. 1999] and receptor modelling (this work).



7. Light and heavy motor vehicles

The quality of the separation of the different sources which can be reached with the receptor model is not sufficient to allow for a differentiation of the emissions of light motor vehicles (passenger cars and light duty vehicles) and heavy motor vehicles (heavy duty vehicles and buses). In order to close this knowledge gap, additional measurement campaigns with high time resolution were performed at the two urban sites Zürich-Kasernenhof and Zürich-Wiedikon (Schimmelstrasse). Zürich-Kasernenhof is situated in a large courtyard of a former arsenal and represents an urban background situation which is not directly influenced by dominating local sources. The site Zürich-Wiedikon is at the kerb of a very busy transit road and represents a strongly traffic influenced situation. Besides air pollution monitoring, the measurements involved also counting of the light and heavy motor vehicle numbers (induction counter). The differences of the parallel measurements at the two sites give a good approximation of the contribution of the local traffic at the Schimmelstrasse.

It was found that the PM₁₀ emissions of the road traffic on the Schimmelstrasse are considerably lower on Sundays than on weekdays. This can be explained with the absence of heavy duty traffic on Sundays. The few counts of heavy motor vehicles on Sundays are most probably passenger buses. From comparison of the road traffic contributions on Sundays and on weekdays and from the traffic frequencies the relative contribution of the heavy duty traffic to the primary PM₁₀ emissions of the total traffic can be estimated to be 53% at this site.

On the basis of the gravimetric daily PM₁₀ values at the sites Zürich-Wiedikon and Zürich-Kasernenhof a similar estimation can be made. The mean difference of the PM₁₀ concentrations at the two sites ($PM_{10}^{Wiedikon} - PM_{10}^{Kasernenhof}$) is 19.7 $\mu\text{g}/\text{m}^3$ for weekdays and 8.9 $\mu\text{g}/\text{m}^3$ for Sundays. In good agreement with the estimations from the monitoring campaigns a relative contribution of heavy duty traffic to the primary PM₁₀ emissions from total traffic of 54% results. The differences ($PM_{10}^{Wiedikon} - PM_{10}^{Kasernenhof}$) are approximately normally distributed, a t-test gives a 95% confidence interval of 49% bis 64%.

8. Conclusions and open problems

For some typical Swiss pollution scenarios the traffic related contributions to the PM₁₀ and partly also to PM_{2.5} was quantified by receptor modeling. The selection of rather polluted sites (in the range from extremely traffic influenced to urban background and suburban) led to the characterization of scenarios which are potentially harmful. For rural sites with low pollution levels (Chaumont) the receptor model did not allow to quantify the traffic contributions with a sufficient reliability. The values presented in this study are therefore representative for the exposition of an urban and suburban population but do not represent a Swiss mean exposition. The limitations of the receptor model became visible. A detailed speciation of the source contributions (e.g. distinction between light and heavy duty vehicles) was not possible. Also the determination of the contributions of the different industrial sources was only possible in a rather summaric way.

With the additional measurement campaigns with high time resolution at the extremely traffic-exposed site Zürich-Wiedikon it was, however, possible to separately quantify the contributions of light and heavy motor vehicles. It could be shown that with the evaluation of differences (local pollution minus background pollution) it is also possible to estimate emission factors for realistic traffic scenarios. There is an urgent need for such informations because the available emission factors from test bench measurements do not provide information on the emissions from abrasion and resuspension processes. In order to close this knowledge gap and to get emission factors for different typical traffic scenarios a new project was recently started. With an optimised concept measurements on both sides of selected roads will be performed in order to determine emission factors for light and heavy motor vehicles and for the involved abrasion and resuspension processes.

As an open problem remain the contributions of the offroad emissions in general. Due to the very similar chemical composition these emissions are not separated from the traffic related factors in the receptor model. It can be assumed, however, that at the selected sites the offroad contributions are quantitatively not important because no typical offroad sources as heavy construction machines, tractors or diesel powered rail engines were active near the sites. But this does not mean that offroad emissions are in general not important as they can cause locally very high particle concentrations.

The contributions of domestic heating (oil and gas) could also not be separated due to the

similar chemical composition. Nevertheless, it can be assumed with high certainty that these emissions are not important at all for the particle concentrations at the selected sites because the emission factors of these heating devices are well known to be very low. The more problematic particle emissions of wood combustion are contained in the factor "biomass burning".

More details concerning methods, results and the technical details of the receptor models can be found in the full project report [Hüglin 2000].

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