Inventorying PM emissions

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1. Introduction

Transfer of material into the atmosphere is a key factor influencing atmospheric concentrations of trace constituents. Emission inventories provide the information needed to understand and quantify the involved processes. Both regulatory interests and scientific needs require high quality emission data. In contrast to inventories of gaseous pollutants, which are well-established, inventories of particulate matter (PM) suffer from inadequate reliability. While considerable efforts for improvements are under way, the major obstacles are in the specific properties of PM. PM is not a unique substance, but an internal and/or external mixture of a multitude of compounds from very different origin, different effects on substrates, and different size. Quantification of emission rates is hampered by the large number of distinct natural and anthropogenic, direct and fugitive sources, as well as their strong dependencies on measuring conditions (temperature, lapse time) and process state (start-up vs. equilibrium). Efforts in Austria resulted in some of the first nation-wide inventories in Europe, which to a considerable extent were supported by country-specific information on emission patterns.

2. Method to set up emission inventories

Emission inventories compile the information for many different sources. For most sources, emissions are calculated as the product of an activity (describing the extent at which an activity occurs) and an emission factor representing the intensity of emissions. It is common practice to rely on existing guidelines which provide recommendations of potentially important source sectors. Following these recommendations first of all allows comparability to similar activities in other areas. Moreover, it safeguards applicability of the emission factors suggested in these guidelines.

For Austria, a first PM inventory was established in 2001 (Winiwarter et al. 2001, 2002). A revision focussing on the open topics as identified in the first study was prepared in a second assessment (Winiwarter et al. 2007, 2008) which also took advantage of the developments made in the scientific literature since, especially with regard to fugitive emissions. A sizable set of specific emission-relevant information could be obtained from measurements obtained in Austria. In the following, we will detail the methods used by source sector, with consideration of fugitive sources (very low emission rates over a large area provide sizable contributions to the total) vs. combustion sources which in general are released via an exhaust duct (stack, chimney, exhaust pipe).

Combustion in industry and power plants

For large installations, direct source measurements are available. Due to the high efficiency and large degree of implementation of abatement devices like electrostatic precipitators, overall emissions are small even for large point sources operated on solid fuels.

Industry (fugitive emissions)

Fugitive emissions from industry derive from fractionating processes and/or mechanical handling of "dusty" material which leaves it airborne. The handling steps (loading / dropping / moving operations) are quite well understood and listed in VDI methods. Theoretical considerations allow developing abatement strategies for fugitive dust emissions (see this compendium, pp. 16 ff).

Very little information is available on construction (e.g., demolishing of buildings) or quarries. The sheer amount of material extracted in quarries in Austria makes this a source to be considered, irrespectively of the actual release rate applied.

Domestic heating

Traditionally, woody biomass is often used for space heating in Austria, especially in the forested areas of the country. Since about 1995, highly efficient stoves are on the market and being installed in houses. It is now estimated that more than 30% of the biomass is burnt efficiently at low emission rates in such new stoves. Particle emission factors of the older types of Austrian stoves are available from measurements on in-use stoves performed 1997/98 (Spitzer et al. 1998) as well as from new stoves (Obernberger and Brunner 2005).

Table 1 presents details on sets of focused Austrian source measurements. The detailed chemical composition of particles emitted from combustion of logwood in a tile stove (Schmidl et al. 2008a) were performed with the major wood types of Austria. These data do not yield emission rates, but allow improved interpretation of ambient air data with respect to source apportionment. In a similar way the smoke of the combustion of garden leaves was chemically characterised (Schmidl et al. 2008b). Recently emission tests of PM10 from modern stoves (pellet stoves and "chimney type" logwood stoves) were performed including detailed chemical composition (Schmidl et al. 2011, Kistler et al. 2012).

Table 1: Emission factors derived from stove measurements (PM emission determined after a dilution step, except where marked).

	PM10 (mg/MJ)
*) Austrian test 1997/98 logwood stoves (Spitzer et al. 1998)	148
*) Austrian standard 1998 for logwood stoves (Beech wood/standardised)	60
2 "Chimney stoves" 6kW, 10 kW, 3 Austrian wood types+Briquetts (Schmidl et al. 2011)	83
"6 kW Chimney stove" 12 Central Europ. wood types+Briquetts (Kistler et al. 2012)	67
"6 kW Chimney stove" 4 Portugese wood types (Gonçalves et al. 2010)	114
2 Pellet stoves / wood pellets (Schmidl et al. 2011)	
"fullload"	8-11
"start up"	8-34
"maloperation"	3-21
Pellet stove (Schmidl et al. 2011)	
miscanthus pellets	8
triticale pellets	120
6kW Pellet stove / wood pellets (Kistler et al. 2012)	21

* PM determination from the undiluted "hot" exhaust stream.

Schmidl et al. (2011) compared emissions from two pellet stoves and two logwood fired "chimney" stoves. While the main components of the particulate emissions of "chimney" stoves are organic material and soot, the emissions of pellet stoves comprise predominantly inorganic ions (sodium, potassium, magnesium, calcium, chloride, nitrate and sulfate). These authors also investigated the emission behaviour of "new types" of agricultural biofuels, such as pellets from elephant grass (miscanthus), and triticale pellets, a type of "energy corn". While elephant grass pellets emissions of PM10 were even lower than for wood pellets, triticale pellets were "high emitters", with a factor of 9 higher PM10 emissions than from wood pellets.

Kistler et al. (2012) used different wood types from Alpine Central Europe and Central European plains on the same pellet and "chimney" stoves. While the Central Alpine forests are dominated by Spruce and Beech, with common occurrence (>5% of the forest area) of Fir, Larch, and Mountain Pines (Scots Pine, Black Pine), the forests in the Pannonian plain are dominated by Black

Locust, Hornbeam, several Oak types and Pines. Surprisingly large differences in the emission behaviour of PM10 for different wood types were observed. This indicates that it may be necessary for the emission inventories to consider the different emission behaviour for different countries of Europe. E.g. Sessile Oak turned out to be a "high" emitter of PM10, with a ten times higher emission rate compared to the "low emitters" Poplar and Larch. Similarily, in a joint study of the University of Aveiro (Portugal) and the TU Vienna, large emission differences of PM10 were obtained for the four major wood types with common occurrence in Portugal: Eucalyptus, Cork Oak, Acacia ("Golden Wattle") and Maritime Pine.

Kistler et al. (2012) report odour emissions in addition to the PM10 emissions, which can be used in dispersion models to estimate the impact of odour nuisance from wood combustion. The authors conclude that frequent short term occurrence of wood smoke odour is likely for many communities with traditional wood stove use ("wood burning communities").

As has been shown in US (Ward et al. 2006) and Scandinavian studies before, extensive use of fuel wood in "wood burning communities" may lead to violations of PM2.5 or benzo(a)pyrene (BaP) ambient air quality standards also in Austria. Exceeding the EU BaP target value due to wood smoke impact is evident in Zederhaus, Salzburg (Spangl and Nagl 2010). Thus, the information about the chemical composition of wood smoke from different wood types and stoves is also important for inventorying the emissions of the substances regulated by ambient air quality legislation (PM10, PM2.5; Pb, Cd, As, Ni; BaP) and for the components under the ECE reporting commitments (e.g. selected PAH and other POP's).

Road transport

Probably the best investigated PM source in general is the road transport. Emission factors for exhaust gases are available from a multitude of dynamometric test bench experiments. Here the main problem is to obtain reliable measurement results that also hold for interpretation in the real atmosphere – particles leaving the exhaust pipe are extremely small and still in the process of growth, as surrounded with high concentrations of condensable gases. The nationally characteristic high share of diesel cars adds to the importance of this source category.

Emission rates from automotive traffic are assessed for individual vehicles on dynamometer test facilities. Such investigations are performed in Austria at TU Graz and TU Vienna. The data are used for the official emission inventory of automotive traffic, the "handbook of Traffic emissions", edited for the countries Austria, Germany and Switzerland, in the current version HBEFA Version 3.1 including all EURO emission classes including EURO 5 and EURO 6, and the chemical components HC, CO, NO_x, PM, CO_2, CH_4, NMHC, Pb, SO_2, N_2O, NH_3, and Benzene, as well as "primary NO_2", particle numbers and PM mass of passenger cars.

As some parameters are uncertain or difficult to access from dynamometer studies (e.g. the influence of malfunctioning cars, so called "super emitters", the "primary NO_2 emission" and non-exhaust PM emissions from abrasion and re-suspension) validation tests of the HBEFA emission rates are performed from on road measurements in tunnels or on streets and highways. Such "real world" tests have been performed in Styria, Carinthia and Salzburg by teams from TU Graz, TU Vienna and the local environmental authorities (Hausberger at al. 2003). Scientific evaluations were also reported for the Tauerntunnel, where three tests took place within three decades. The results of the first two tests demonstrate the decadal improvement of traffic emissions (Schmid et al. 2001), and a relatively good agreement between measured and modelled emissions (unpublished data). Non-exhaust emissions were investigated in two studies in the Kaisermuehlentunnel (a city highway tunnel), yielding different results in the two runs considered to reflect different driving conditions and different silt loadings of the highway surface (Laschober et al. 2004, Handler et al. 2008).

At the time of the first Austrian inventory (Winiwarter et al. 2001) available information on fugitive emissions from road transport (due to suspension of road dust, brake wear or tyre wear) was very unreliable. Since that time, measurements have been performed, some of which also cover activities in Austria (Ketzel et al. 2007). These measurements now indicate fugitive emissions to be about as important as exhaust emissions in terms of PM10 mass.

Off-road transport

Emission factors for exhaust emissions are available, which are characterized by the typically low turnover in off-road vehicles. Engines tend to be old, new standards are implemented into the fleet very slowly. Very little information is available on fugitive emissions. This is a potentially important source which requires further efforts to be spent.

Agriculture

Indoor air in animal houses is known to be loaded with particulate matter. Simple extrapolation of air exchange rate would indicate this to be a considerable source, but the respective biogenic material can not be observed in the ambient air. Here again more information is urgently needed. Emission estimates due to agricultural field operations (plowing and packing, harrowing, disking, cultivating) are based on (few) European measurements, again with

Austrian participation (Öttl and Funk 2007, Funk et al. 2008). These studies differentiate strongly different emissions, with huge increases observed when soil is completely dry. The distinction between soil dryness seems to be a guiding parameter to establish an inventory.

Natural emissions

Except for wind-blown dust, which never was explicitly investigated in Austria, the most important natural source in terms of PM mass seems to be the release of fungal spores. A handful of ambient air measurements in the Vienna area indicate concentrations of fungal spores to be clearly larger than those of cellulose (marker compound for plant debris, which we regard to be likewise stable in the atmosphere). Several data series over Europe allow to compare ambient air winter and summertime ratios of cellulose to a few anthropogenic PM species. Assuming that emission ratios should match concentration ratios, we are able to provide an emission range for plant debris, and extending the same logics also the emission range for fungal spores. Adding these species provides a first quantification of Primary Biogenic Aerosol Particles (PBAP) emissions (Winiwarter et al. 2009a, Puxbaum et al. 2009). While this species provides about 5% (annual average) of the ambient PM10 observed, it is also important as natural cloud condensation nuclei or compound affecting the atmosphere's radiative properties.

Following an event of volcanic dust impacts, a very recent study with Austrian participation estimated emission strengths of emissions from the Icelandic volcano Eyjafjallajökull. Stohl et al. (2011) apply an inversion model to satellite observations of aerosol to arrive at total emission of airborne particles due to this volcanic eruption in the year 2010 over 40 days of 8.3 +/- 4.2 Tg, which is the same order of magnitude as the total annual anthropogenic PM10 emissions over Europe of 6 Tg (see Winiwarter et al. 2009a).

3. Some key results of Austrian emission inventories

Describing source sectors individually allows to identify a pattern in emission reports. For very different source sectors, huge discrepancies between reported amounts of material available for atmospheric release and their actual atmospheric occurrence are evident. We have developed the concept of "potential emissions" to deal with the problem. "Potential emissions" as derived from the availability of particles to be released are the upper limits to possible release fluxes, but they require confirmation from atmospheric measurements before being accepted to the inventory. When no such confirmation was available, we used emissions calculated according to the lower of the available emission factors. This way TSP emissions for Austria were less than half of the emissions that could have been estimated when considering the potential emissions. These differences occur for fugitive emissions, they become dominating the large size fractions as well as TSP emissions.

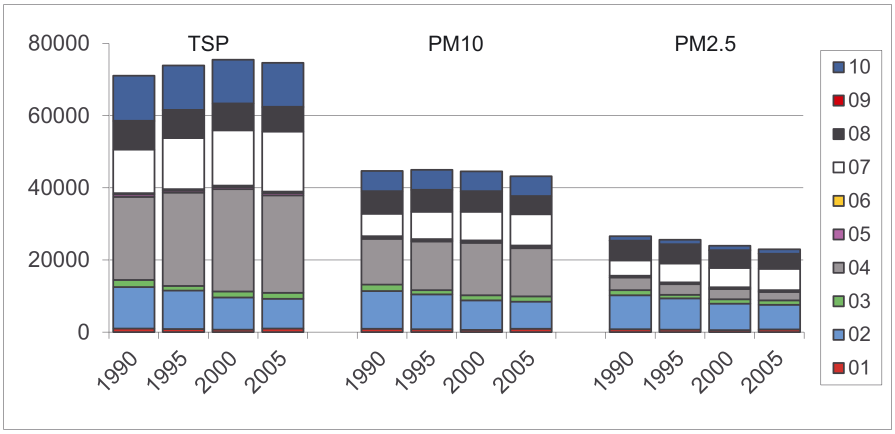

Figure 1: Time series (Winiwarter et al. 2007) for Austrian PM emission estimates (in tons per year). Results are presented by CORINAIR source categories: (01) Combustion in energy and transformation industry; (02) Non-industrial combustion plants; (03) Combustion in manufacturing industry; (04) Production processes; (05) Extraction and distribution of fossil fuels; (06) Solvent and other product use; (07) Road transport; (08) Other mobile sources and machinery; (09) Waste; (10) Agriculture.

The resulting emissions of TSP and the size fractions PM10 and PM2.5 are presented in Figure 1. According to these results, the emissions of TSP in Austria derive from:

- industrial processes (about one third; here potential emissions – and their associated uncertainty – may contribute significantly to the number, as it was not possible to obtain a lower estimate)
- road transport (about a quarter)
- agriculture
- small heating installations

Smaller size fractions (PM10, and especially PM2.5) are dominated by combustion emissions – small heating installations, road transport and other transport make up for most of the PM2.5 emissions.

The trend between 1990 and 2005 indicates increased emissions from road transport, despite of technology improvements, due to the increased transport activity. For small heating installations, even increases in firewood consumptions have been more than matched by the introduction of efficient burners over the last decade, such that overall emissions have decreased.

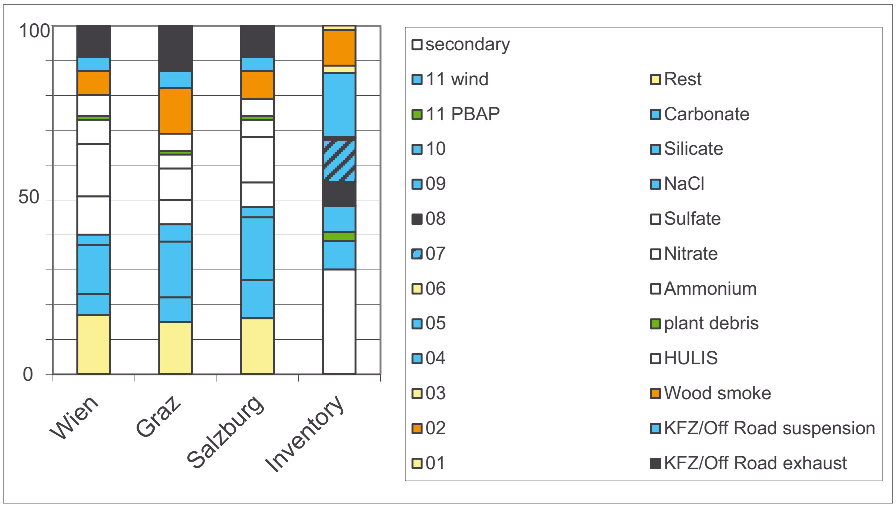

Figure 2: Comparison of AQUELLA source analysis (Bauer et al. 2007) – sources described in right hand column of the legend – to the Austrian inventory, CORINAIR categories in the left column as listed in Figure 1 and include (11) "natural sources" as well as a fraction of 30% secondary aerosol as derived by the source analysis. Colours were assigned to help comparison, black represents soot, orange combustion products, blue stands for mineral compounds, green for organic material and white is secondary aerosol.

Validation of an emission inventory requires an independent set of data. The most independent way to assess emissions is ambient measurements. Figure 2 compares results obtained from ambient measurements using source apportionment (Bauer et al. 2007) to the inventory. While this approach currently is not very detailed, it indicates at least the general applicability of the approach taken.

4. Austrian activities referring to the European situation

Some of the early activities to derive emissions for Europe as a whole have been developed in Austria (Lükewille et al. 2001). This assessment took advantage of the activity information available in the RAINS/GAINS database (see this compendium, pp. 121 ff) and added size-fractionated PM as a new component. Spatial resolution was by countries, results were available in 5 year intervals from 1990 to 2030. The database has recently been expanded to cover also simple chemical speciation: black carbon, organic carbon (Kupiainen and Klimont 2004, 2007), which i.a. have been used to estimate PBAP emissions.

As part of a contribution to COST action 633 "Particulate Matter and Health", a review on PM emission inventories in Europe has been prepared (Winiwarter et al. 2009b). A large body of scientific literature is presented in this review dealing with multiple aspects of PM emissions in Europe. The overview clearly separates well-investigated areas (especially vehicle emissions, but for transport even the fugitive emissions now have been reasonably well assessed for Europe) and determines the data gaps. Not surprisingly, gaps for Europe refer to the same sources as for Austria: Information on fugitive emissions (caused by wind shear, material transfer processes or other mechanical forces from non-point sources) is sparse. Especially activities in industry (quarries), agriculture, but also natural particle formation like sea salt and wind blown dust will require further attention. PM is an intensely studied area, and improvements may be expected with validation methods in place as shown in Figure 2 for Austria.

5. Conclusions

Improving the Austrian emission inventory would require additional measurements of those sources where potential emissions may play a role. Currently, industrial processes (quarries) are the only source where emissions have been calculated from their potential emissions, i.e. the maximum available estimate. This source may be overestimated, but no other information has been available. Combustion emission estimates would strongly benefit from better activity estimates for domestic heating, especially concerning the quantity of wood burnt in modern installations.

Most promising abatement options regard phase-out of outdated technology. This includes old Diesel engines used in "Other transport", as well as old, inefficient equipment to burn wood. Possibly measures in limestone quarries could help reduce fugitive emissions.

Comparison with alternate emission estimates and results of source apportionment studies show no major discrepancies between these two independent approaches. While further, more detailed and focused comparison exercises are needed to arrive at a comprehensive conclusion, this outcome supports the results and the reliability of the new inventory.

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