4th International Workshop
on Uncertainty in Atmospheric Emissions
7-9 October 2015, Krakow, Poland

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About the Workshop

The assessment of greenhouse gases and air pollutants (indirect GHGs) emitted to and removed from the atmosphere is high on the political and scientific agendas. Building on the UN climate process, the international community strives to address the long-term challenge of climate change collectively and comprehensively, and to take concrete and timely action that proves sustainable and robust in the future. Under the umbrella of the UN Framework Convention on Climate Change, mainly developed country parties to the Convention have, since the mid-1990s, published annual or periodic inventories of emissions and removals, and continued to do so after the Kyoto Protocol to the Convention ceased in 2012. Policymakers use these inventories to develop strategies and policies for emission reductions and to track the progress of those strategies and policies. Where formal commitments to limit emissions exist, regulatory agencies and corporations rely on emission inventories to establish compliance records.

However, as increasing international concern and cooperation aim at policy-oriented solutions to the climate change problem, a number of issues circulating around uncertainty have come to the fore, which were undervalued or left unmentioned at the time of the Kyoto Protocol but require adequate recognition under a workable and legislated successor agreement. Accounting and verification of emissions in space and time, compliance with emission reduction commitments, risk of exceeding future temperature targets, evaluating effects of mitigation versus adaptation versus intensity of induced impacts at home and elsewhere, and accounting of traded emission permits are to name but a few.

The *4th International Workshop on Uncertainty in Atmospheric Emissions* is jointly organized by the *Systems Research Institute of the Polish Academy of Sciences*, the Austrian-based *International Institute for Applied Systems Analysis*, and the *Lviv Polytechnic National University*. The 4th Uncertainty Workshop follows up and expands on the scope of the earlier Uncertainty Workshops – the *1st Workshop* in 2004 in Warsaw, Poland; the *2nd Workshop* in 2007 in Laxenburg, Austria; and the *3rd Workshop* in 2010 in Lviv, Ukraine.

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Global anthropogenic particle number emissions and their size distributions

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Abstract

Aerosol particle number concentrations and size distributions affect our climate by determining the formation of cloud droplets and thus altering the cloud reflective properties. The aerosol-cloud interactions are one of the main uncertainties in estimating the future climate change. One of the weaknesses in current climate modelling is the description of number emissions and size distributions of particles. Here, we present the first global results of implementing particle number emission factors to GAINS emission scenario model and discuss the related uncertainties. The uncertainties for different source sectors vary significantly, causing a steep difference in total uncertainties in different parts of the world. The reason for these uncertainties is the scarcity of data on particle number size distributions for certain sources. The implemented particle number emission factors, however, are expected to be a significant improvement over previously applied particle number emissions estimates in climate modelling.

Keywords: particle number emissions, number size distribution, emission scenario model, aerosol-cloud interactions

1. Introduction

One of the main uncertainties in our understanding of the future climate change arises from the aerosol-cloud interactions [1]. One factor to these uncertainties is the inadequate description of aerosol number emissions from anthropogenic sources. The number of cloud droplets, which reflect solar radiation back to space, depends on the number concentrations of particles in cloud condensation nuclei -size range (CCN, diameters d_P close to or over 0.1 μm). These particles are emitted to the atmosphere directly from anthropogenic sources or formed in atmosphere due to the growth of ultrafine particles (UFP, with diameters below 0.1 μm), which may be of either biogenic or anthropogenic origin. As a source of CCN, the biogenic growth of UFP is roughly as significant a source of CCN as direct anthropogenic emissions [2]. On the other hand, UFP have severe adverse health effects, which are different to those of particulate mass [3]. Also the main anthropogenic sources of UFP, which typically dominate particle number concentrations (PN), are different to the main sources of particulate mass [4].

Here, we present the first results of the implementation of aerosol number emission factors (EF_{PN}) in the global emission scenario model GAINS (Greenhouse gas - Air pollutant Interactions and Synergies [5]) and discuss the related uncertainties.

2. Methods

The GAINS model (Greenhouse gas – Air pollutant Interactions and Synergies [5]) is an integrated assessment model, which brings together information on the sources

and impacts of air pollutant and greenhouse gas emissions and their interactions. GAINS combines data on economic development, the structure, control potential and costs of emission sources, the formation and dispersion of pollutants in the atmosphere and an assessment of environmental impacts of pollution. The political scenarios in GAINS allow for researchers and modellers to study the future global emissions and their spatial distribution and for decision makers to compare the costs and outcomes of regulations and investments on new technologies. GAINS describes the inter-relations between the effects and emissions of various pollutants (SO_2, NO_x, PM, NMVOC, NH_3, CO_2, CH_4, N_2O, F-gases) that contribute to these effects. GAINS assesses more than 1000 measures to control the emissions to the atmosphere for each of its 168 regions (mainly countries, of which some divided in regions, e.g. China consists of 32 regions, and some grouped, e.g. Middle East). In its optimization mode, GAINS identifies the least-cost balance of emission control measures across pollutants, economic sectors and countries that meet user-specified air quality and climate targets.

The annual emissions E in a country or a region i are calculated with

$$E_i = \sum_{jkm} E_{ijkm} = \sum_{jkm} A_{ijkm} X_{ijkm} \mathrm{EF}_{ijkm} , \quad (1)$$

where the indices and symbols refer to

j	Source sector (e.g. domestic single house heating boilers)
k	Fuel (e.g. firewood, coal)
m	Abatement technology (e.g. pellet boilers, boilers with electrostatic precipitator)
A	Volume of annual activity (typically annual energy consumption in sector j with fuel k)
X	Share of abatement technology of the activity m (so that $\sum_m X_m=1$)
EF	Emission factors for each sector-fuel-technology –combination (emissions per activity unit)

We have recently introduced aerosol particle number emission factors (EF_{PN}) with corresponding particle (number) size distributions (PSD) to GAINS [4]. For road transport PN_{EF}:s and PSDs are based on the latest version of TRANSPHORM database [6]. For the global analysis these were extended with separate PN_{EF}:s and PSDs for different fuel sulphur contents. For other sources, emission factors are obtained from the literature and from the emission inventory by TNO [7,8]. The implemented emission factors and size distributions represent the emissions of both primary and secondary particles immediately after cooling and dilution to the surrounding air. Particle size distributions in size range 3-1000 nm are described with 8 size bins, facilitating their application in air quality and climate modelling.

3. Results

Global PN emissions are dominated by emissions of UFP, which form close to 80% of the total global emissions (lower panel of Fig. 1).

Figure 1 shows, for year 2010, the shares of different sources in global continental total particle number emissions (PN_{tot}), in number emissions of ultrafine particles (UFP) and non-UFP (d_P >100 nm), as well as in mass emissions of particles with $d_P < 1\mu m$ (PM_1). The main source of UFP is road transport, representing 40 % of the total UFP emissions and thus being the largest contributor to total aerosol particle number emissions. Power production contributes to the UFP emissions with a 20 % share, while

residential combustion has a 17 % share. The shares of residential combustion and road transport in non-UFP number emissions are quite similar, roughly 30 % each, whereas the PM_1 mass emissions are clearly dominated by residential combustion (> 50 %). The vast differences between the number and mass emission shares, especially from road transport and residential combustion, indicate the need for assessing the size segregated number emissions of aerosols in addition to mass emissions.

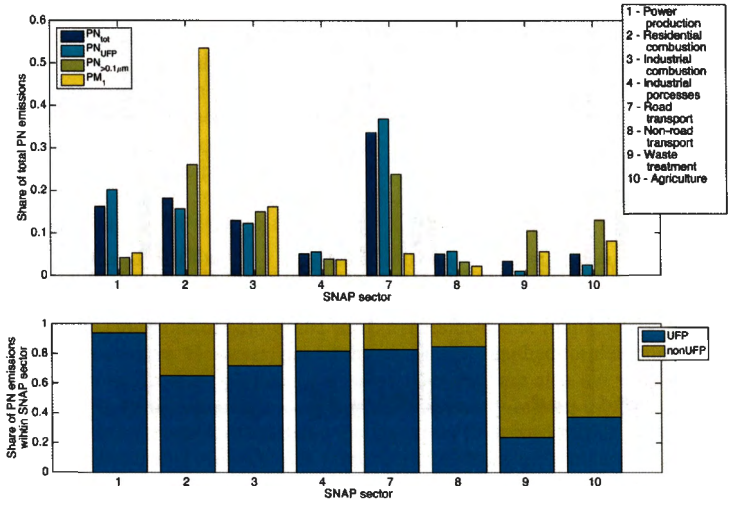

Figure 1. Upper panel: shares of different source sectors in aerosol number emissions of all (PN_{tot}), ultrafine (PN_{UFP}) and non-ultrafine (PN_{nonUFP}) particles and aerosol mass emissions of particles with diameters below 1 μm (PM) on 2010. Lower panel: shares of ultrafine and non-ultrafine particle in PN emissions for each SNAP-sector (see legend for clarification of SNAP sector codes).

The annual PN emissions and their estimated future trend in each source sector on different continents, with Eurasian continent divided to major countries and the rest of Europe and rest of Asia, are depicted in Figure 2. The future trend is based on the current legislation baseline scenario (ETP_CLE_v5) compiled in the ECLIPSE project [9]. In 2010, China emitted clearly the most aerosol particles due to high emissions from power production (especially from coke production), residential coal combustion and industrial combustion, followed by Asia (excl. China, India and Russia) and Europe (excl. Russia). In most parts of the world road transportation is the major source of particles.

The actions determined in current legislation are foreseen to decrease the PN emissions in China substantially due to decreases in emissions from coke production and residential coal combustion. This is partly related to the increase in the electricity network, replacing the coal fired cooking stoves. In Europe, North- and South-America and Australia due to the drastic decrease foreseen in traffic emissions due to improving particle emission abatement technologies, especially particle filters. On the contrary, especially in India and Russia, the increase in activities in industrial processes and combustion (the latter mainly in India) and combustion in gas pipeline compressors (in Russia) causes increases in total emissions. In Asia and Africa, the increase in road

transportation seems to overrule the benefits of improving emission abatement technologies, but also the emissions from other source sectors are estimated to increase. The global sum of continental anthropogenic emissions is predicted to decrease roughly by 15 % from 2010 to 2020 (from $1.5x10^{28}$ to $1.3x10^{28}$ particles/year), but expected to remain quite constant from 2020 to 2030.

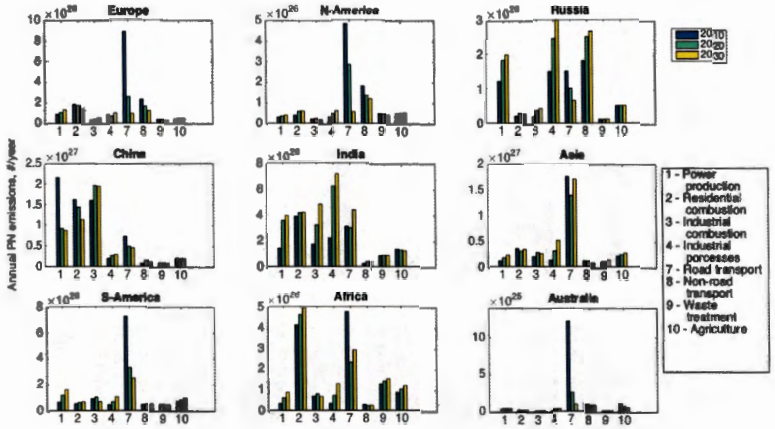

Figure 2. Contributions of different source sectors to particle number emissions in different parts of the world, from 2010 to 2030.

4. Uncertainties in the PN emissions

The uncertainties in the emissions arise from uncertainties specific for the different factors in Equation (1). Here we concentrate, however, only on the uncertainties related to the particle number emission factors EF_{PN}, because they can be estimated to be the main source of uncertainties in PN emissions due to the following reasons. The variation in particle numbers behaves typically in logarithmic scales and thus their concentrations and emissions can vary in orders of magnitude. Furthermore, e.g. Wang et al. [12] have shown that the emissions of the traditional pollutants (SO_2, NO_x, $PM_{2.5}$ and PM_{10}) calculated with GAINS compare well with those based on measurements (for the studied pollutants the major relative error was found to be factor of 2.5), which suggests that the activities and shares of abatement technologies in Eq. (1) represent the reality reasonably well. Finally, since the emission factors for the traditional pollutants have been revised several times due to restrictions in their emissions and concentrations, but much less efforts have been put to determining particle number emissions, which are restricted only in the latest EURO standards for road traffic, it is quite obvious that the emission factors are the most uncertain part of the emission calculation for PN.

The uncertainty levels related to different sources vary significantly. The EF_{PN} and PSD applied here for road traffic have been determined in a long sequence of EU funded projects were compiled, revised and reviewed in the project TRANSPHORM. However, even in this source sector there remain uncertainties related e.g. to the varying

driving conditions, effects of varying ethanol concentration in fuels and differences between laboratory and real-world emissions. The road traffic emission factors can still be estimated to be among the best analysed ones together with the heating stoves, boilers and fireplaces fuelled with wood in Western countries, due to the various research articles on their EF_{PN}:s. On the other end, the emission factors for coke production plants [10] and coal combustion in residential (mainly cooking) stoves [11] are highly uncertain, because both are based on only one article. For these sources, among several others having a minor part in European emission, but potentially much larger in other parts of the world, the EF_{PN}:s for different technologies cannot be determined from the literature, and thus we have set the effects of emission abatement technologies on EF_{PN} to be similar to EF for particulate matter mass emissions. This most certainly decreases the reliability of the emissions from these sources when newer technologies become more popular. Also the biomass, i.e. wood, agricultural residues and dung, combustion in residential sector especially in India and Africa are not fully representative, due to the lack of references for the typical burning equipment and conditions.

Another factor for uncertainty is often weak presentation of the smallest particles, d_P<0.01 µm, in the emission factors and PSDs. High sulphur contents in the fuel lead typically to high emissions of below 0.01 µm particles before or immediately after the emissions to atmosphere. However, not all the instruments applied for determining the emission factors detect these particles and sometimes they are also too volatile to be detected with the used technologies. Additional uncertainty related to these smallest particles yields from the lack of emission factors, apart from road traffic, for different fuel sulphur contents and technologies removing SO_2 from the exhaust e.g. in coal plants or industrial combustion.

The variations in source sector specific uncertainties in emission factors described above cause steep differences in the spatial distribution of the uncertainties. In the areas dominated by traffic emissions (see Figure 2), the total uncertainty is the smallest, whereas the emissions in China, India and Russia can be considered much higher.

5. Final remarks

The particle number and mass emissions are typically dominated by different sources sectors and individual sources contribute very differently to these measures. Thus, despite all the above mentioned uncertainties, the PN emissions in GAINS can be expected to describe better the real world emissions than those estimated by converting mass emissions to number emissions with source sector –specific size distributions and mass-to-number factors. For reducing these uncertainties, it is necessary to conduct particle number emission and size distribution measurements for the indicated source sectors.

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