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Size-resolved global emission inventory of primary particulate matter from energy-related combustion sources☆

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HIGHLIGHTS

- Mass-based size distribution of emissions of energy-related combustion sources.
- Method to estimate the size distribution of components of the combustion sector.
- Global PM_{10} emissions showing single-mode size distribution with peak around 700 nm.
- Discussion of uncertainties in global size distribution emission estimation.
- Investigation of mass size distribution changes with emission reduction scenarios.

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ABSTRACT

Current emission inventories provide information about the mass emissions of different chemical species from different emitting sources without information concerning the size distribution of primary particulate matter (PM). The size distribution information, however, is an important input into chemical transport models that determine the fate of PM and its impacts on climate and public health. At present, models usually make rather rudimentary assumptions about the size distribution of primary PM emissions in their model inputs. In this study, we develop a global and regional, size-resolved, mass emission inventory of primary PM emissions from source-specific combustion components of the residential, industrial, power, and transportation sectors for the year 2010. Uncertainties in the emission profiles are also provided. The global size-resolved PM emissions show a distribution with a single peak and the majority of the mass of particles in size ranges smaller than 1 μm. The PM size distributions for different sectors and world regions vary considerably, due to the different combustion characteristics. Typically, the sizes of particles decrease in the order: power sector > industrial sector > residential sector > transportation sector. Three emission scenarios are applied to the baseline distributions to study the likely changes in size distribution of emissions as clean technologies are implemented.

1. Introduction

Knowledge of the size distribution of aerosols is essential to understanding climate and human health effects. Aerosol optical properties depend strongly on the size distribution (Haywood and Boucher, 2000; Yu et al., 2006); and sub-micrometer aerosols, which have longer atmospheric lifetimes, scatter more light per unit mass (Seinfeld and Pandis, 2006). Smaller carbonaceous aerosols lead to larger, more negative, direct and indirect aerosol forcing (Bauer et al., 2010). The number of cloud condensation nuclei (CCN) per mass of aerosol depends on the chemical composition of aerosols as a function of size (Anttila et al., 2012; Feingold, 2003; McFiggans et al., 2006). In addition, it is well known that the size distribution of particulate matter (PM) determines the potential for human health effects; small particles can

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readily penetrate into the deep lung and initiate cardiopulmonary disease (Delfino et al., 2005; Pope and Dockery, 2006). Therefore, it is critical to know the size distribution of PM in the atmosphere, which is dependent both on the size distribution of primary PM and of secondary PM formed in the atmosphere.

Most current climate and chemical transport models have the ability to represent the temporal and spatial variability of the aerosol particle mass distribution but must assume a size distribution for the primary PM emissions in order to calculate radiative effects (Bauer et al., 2010) and transport. Most widely-used PM emission inventories (Bond et al., 2004; Cooke et al., 2002, 1999; Reff et al., 2009; Zhang et al., 2007) have very little size resolution and typically provide only mass emission information. Emission inventories for the U.S. (e.g., the National Emission Inventory (NEI)) and for Europe (e.g., European Monitoring and Evaluation Programme (EMEP)), as well as a few other countries, such as China (e.g., Zhang et al., 2007), provide some size information, but none of them provide continuous size distributions. They contain only very broad size bins such as total suspended particles (TSP), PM_{10}, and $PM_{2.5}$. Thus, modelers have to choose what sizes to assign for emission fluxes most of the time. The assumed size distributions may introduce a large amount of uncertainty in prediction of CCN concentrations (Pierce and Adams, 2009; Pierce et al., 2007; Reddington et al., 2013; Spracklen et al., 2011) and estimates of climate forcing (Bauer et al., 2010; Spracklen et al., 2011).

In this study, we develop the first global emission inventory of PM_{10} with detailed particle size distribution, especially for submicron particles, and thereby advance our understanding of the effects of particle size. We acknowledge that number size distribution is also important and provides different information from mass size distribution. In this study, we provide size-resolved emissions by mass, since it can be applied directly to currently available PM emission inventories (Bond et al., 2004; Streets et al., 2004; Yan et al., 2014b, 2011; GAINS, 2014) without any modification to their calculation methodologies. We tabulate size distribution profiles from the literature that contain measured size distributions of particle emissions from each source category. Section 2 introduces the methodology used to parameterize the size distributions. Section 3 discusses the size distributions by region, energy sector, and designed scenarios. Conclusions and future work are summarized in Section 4.

2. Methodology and datasets

The size-resolved mass emission inventory in this work is built upon previous work (Bond et al., 2004) that involved coarse or no size resolution. In that work, a technology-based model was constructed to estimate present-day global emissions of black and organic carbon particles. This model determines emissions by apportioning fuel use among different emitting technologies. Such an approach has also been used for historical and future emissions estimates (Bond et al., 2007; Streets et al., 2004; Yan et al., 2014b, 2011). The technology-based model allows us to discriminate size distributions among sources with different technologies, as well as to keep track of their impacts. In this work, we use updated combinations of fuels, combustion technologies, and emission control technologies based on Bond et al. (2004) and apply appropriate size distributions for each combination.

Fig. 1 shows the framework for building a size-resolved emission inventory. First, this work enhances existing compilations through literature review and update. Particle size distributions by sector, fuel, and technology are collected from the literature, as detailed in the Supporting information (SI). Second, these distributions are parameterized by unimodal or multimodal lognormal distributions, depending on the sample size of each distribution and the availability of data, and presented as mass median diameters (MMD), geometric standard deviation (GSD), and mass mixing ratio or weight (w) within each mode, if the distribution has more than one mode, as shown in Section 2.2. The modeled size distributions do not reproduce the measurements exactly, so there is uncertainty in the derived parameters, as described in Section 2.3. Third, these size distributions from different data sources are merged. There are insufficient data available to use region-specific measurements. We choose to combine data from all regions to generate a more robust representation of the distribution for a particular technology. When more than one distribution for a single technology is included, these distributions are treated equally and the average of the distributions is used. Finally, the estimated size distributions of each technology are combined with the corresponding mass-based particle emission factors and a fuel consumption activity database to develop a size-resolved emission inventory. The resulting emission inventory gives a continuous mass distribution by particle size. The distribution of total PM_{10} emissions (Em) along the logarithm of particle diameter ($\ln D_p$) for each sector k in a specific year i is given by:

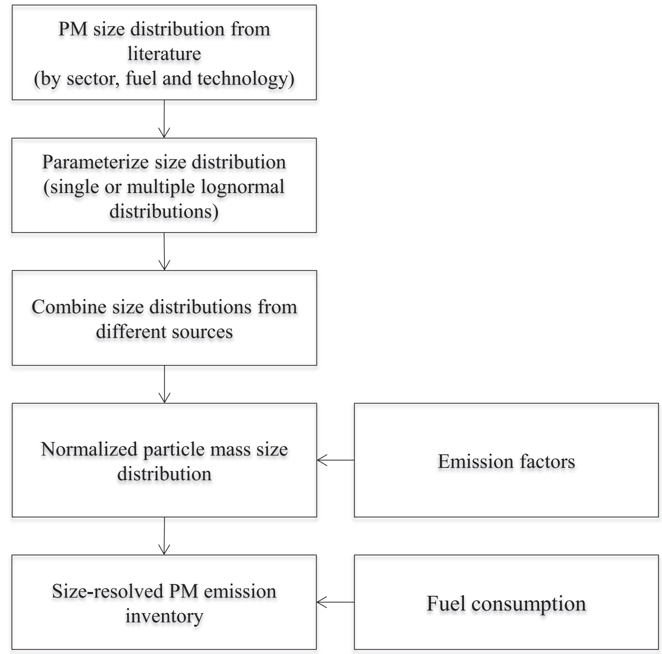

Fig. 1. Framework for building a size-resolved PM emission inventory.

$$Em_{i,k}(\ln D_p) = \sum_j \sum_l \sum_m FC_{i,j,k,l,m} EF_{k,l,m} g_{k,l,m}(\ln D_p) \quad (1)$$

where i, j, k, l, and m represent year, region, sector, fuel, and fuel/technology combination, respectively. Em is emission, FC is fuel consumption (kg/year), EF is emission factor (g/kg fuel) specific to a fuel/technology combination (including the effect of any control devices), D_p is particle diameter, and g ($\ln Dp$) is the particle size distribution of PM_{10} in the form of a single- or multi-lognormal distribution (see Section 2.2).

Mass fractions within certain size bins (e.g., PM_1, $PM_{1-2.5}$, and $PM_{2.5-10}$) can also be computed by the integrals of the distribution. Using $PM_{1-2.5}$ as an example, it can be estimated by: