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**Modeling emissions for three-dimensional atmospheric  
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# Modeling emissions for three-dimensional atmospheric chemistry transport models

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## Introduction

Many regions in the world suffer from serious air pollution problems. Although many countries in Europe and North America significantly reduced their emissions of harmful gases and particles, air pollution remains a serious threat for human health. EU states that air pollution is one of the most severe environmental problems in Europe (European Environment Agency, 2016) and the World Health Organization (WHO) published air quality guidelines (WHO, 2006) and limit values, which are still exceeded in many regions of the world (Henschel et al., 2013, Prüss-Üstün et al., 2016).

Atmospheric chemistry transport models are used to better understand the relationship between emissions from different sources – like natural and anthropogenic emissions – and concentration levels of harmful substances as well as their spatial and temporal distribution. In order to deliver the best possible description of the fate of air pollutants in the atmosphere, these models need accurate information about chemical reactions, the physical state of the atmosphere and about the flux of certain trace gases and particles into the atmosphere. This flux is typically called ‘emission’. Many studies have emphasized that a profound knowledge about how, where and when substances are released (‘emitted’) into the atmosphere is crucial

for the reliability of air quality predictions because transport and chemical transformation heavily depends on meteorological variables that also vary in space and time (see e.g. Frost et al., 2013, Fuzzi et al., 2015, Bergström et al., 2012, Denier van der Gon et al., 2015, Fountoukis et al., 2014, Im et al., 2014, Kühlwein et al., 2002, Vedrenne et al., 2016). In addition, chemical reactions, adsorption, desorption and coagulation processes depend on the presence of possible reaction or collision partners.

Emission data for anthropogenic emissions are provided in the form of emission inventories, which typically contain annual national totals for certain emission sectors, like industry, transportation and households, sometimes on predefined grids. A temporal distribution of the emissions is typically not given. However, in 3D chemistry transport models (CTMs), the emission information needs to be available with sufficiently high temporal resolution and on the same spatial grid used by the CTM. This paper reviews the models and methods that are used around the world in order to transform the information given in global and regional emission inventories into temporally and spatially resolved data that can directly be used in 3D CTMs.

Emissions from natural sources like sea spray, desert dust or biogenic particles and volatile organics depend to a large extent on meteorological conditions, in particular on wind speed (for sea spray and desert dust), temperature (biogenic particles and semivolatile species) and radiation (volatile organics). Therefore they are often not part of inventories but computed inline within the CTMs using adequate parameterizations. Typically meteorological data as well as land use data are used in these parameterizations.

We start with a summary of the most important and most widely used regional and global emission inventories. We look at the chemical species and the emission sectors that are covered in these inventories. Then we summarize the methods to create emission inventories. This involves top-down approaches to disaggregate bulk emissions spatially and temporally or to redistribute emissions that are available as gridded data as well as methods to calculate emissions from single sources, described as a bottom-up approach. Models that treat natural emissions or semi-natural emissions like vegetation fires are presented in the following chapter. Finally we give an outlook on new directions in emission modeling and the possibilities to significantly improve emission data for CTMs when new and large data sources like traffic information become available for emission models.

## **Emission Inventories**

Emission inventories are collections of emission data from many sources combined into one integrated data set. They usually contain annual total emissions of certain substances in a specified region, e.g. per country. The collection of the emission data is often done in a very detailed way for a high number of individual sources such as power plants, industrial plants or motor vehicles. Each individual facility is categorized into so called emission sectors and sometimes split into subsectors. The methods of emission compilation vary from region to region and from country to country. Often, the data collection starts on the level of rather small political units like counties in the US and is then progressively aggregated to larger political units. In Europe, for example, every country reports annual total emissions per emission sector to the European Monitoring and Evaluation Program (EMEP). When brought onto a map, the data may be given as the sum of all sources in a certain sector of this country. Because this is hardly usable in three dimensional emission models, many emission inventories are, in addition to country totals, also given on a grid. This gridding is done with methods described in one of the following sections. Often it needs to be repeated in order to map the emissions onto the specific grid needed for a certain model application. This may cause specific problems like emissions being placed into regions without sources or the reduction of emission gradients.

### Emission sectors

Major anthropogenic emission sources are often reported individually. However, there are numerous small and medium scale sources of air pollutants and it is not feasible to describe every single source separately when compiling a comprehensive source inventory. Because of this, emission sources are typically aggregated to emission sectors which have a varying degree of detail ranging from as few as 10 sectors up to several hundred subsectors. The most commonly used systems are Nomenclature for Reporting / Common Reporting Format (NRF/CRF) of United Nations Framework Convention on Climate Change (UNFCCC) also known as IPCC sectors, Selected Nomenclature for Air Pollutants (SNAP), Statistical Classification of Economic Activities in the European Community (NACE) and the National

Emission Inventory (NEI) emission sectors used in North America. These sectors are defined based on similarities of sources with regards to used material (e.g. fuel type), process characteristics (e.g. combustion processes, volatilization, product use), or products or purpose of activities causing emissions (e.g. power generation, transport). Due to the manifold ways of aggregating emission sources into sectors it is often problematic to establish an unambiguous mapping between emission sectors of different inventories. In spite of this, sources within a sector are usually characterized by comparable temporal and spatial distribution patterns as well as chemical composition of the emission flux, and this facilitates the development of sector-wise disaggregation methods for top-down emission models.

European inventories typically use the SNAP sectors, which contains 11 different sectors (10 anthropogenic and 1 natural emission sector) that are further subdivided into subsectors. Emissions are reported by the European countries following the detailed NFR source sector classification system. This system contains several dozen categories for which several thousand emission factors exist (European Environment Agency, 2016a) and can be used in the national emission reports.

In the US, the NEI consists of eight major source sectors with a total of 59 subsectors. The sectors are in some way similar to what is used in Europe but not all sectors can be found in European inventories. Nevertheless, the approach is similar and the categories can be mapped to each other. The NEI is documented in detail in a technical support document provided by the Environmental Protection Agency (EPA) of the United States (USEPA, 2017).

In other regions of the world much less detailed information about sectoral emissions exists. For example, the Asian inventory REAS (Ohara et al., 2007) used three main sectors, each having two subsectors for the anthropogenic emissions in the first version of the inventory. In REAS 2 (Kurokawa et al., 2013) the sector road transport was added.

## Emitted substances

The substances included in emission inventories can be divided into the ‘classical’ pollutants, greenhouse gases (GHG), and toxic substances. The ‘classical’ pollutants are SO<sub>2</sub>, NO<sub>x</sub>, CO, NMVOC, NH<sub>3</sub> and PM. These species have to be considered by every comprehensive CTM for two reasons. Firstly, they are the so called criteria pollutants or their precursors. Criteria

pollutants are substances that can harm human health and the environment and cause damage to buildings. For these substances ( $O_3$ ,  $NO_2$ , CO,  $SO_2$ , PM) air quality standards and concentration limits are enforced in all developed countries. Secondly, the ‘classical’ pollutants are necessary to determine the oxidative state of the atmosphere and the particle number, mass, and surface area available for physico-chemical interactions. Thus they induce a direct feedback on the lifetime and transport patterns of most air pollutants. Greenhouse gases are sometimes reported together with the classical pollutants in the same inventory (e.g. in the RCPs and in EDGAR). To a large extent, they have the same sources but GHGs are not very reactive and therefore they are not further discussed here. The term toxic substances subsumes chemicals that have a known toxic or ecotoxic effect. They comprise a large and heterogeneous group of persistent organic pollutants (e.g. PAH, PCB, dioxins), VOCs (e.g. formaldehyde, acrolein) and heavy metals (e.g. Hg, Pb, Cd).

Furthermore, some emission species are actually mixtures of substances and need to be split into components for the use in CTMs.  $NO_x$  includes NO and  $NO_2$  and almost all sources of nitrogen oxides contain both compounds. An emission split has to be prescribed. PM is typically subdivided into two size classes, PM<sub>10</sub> and PM<sub>2.5</sub>, with particle diameters smaller than 10 $\mu$ m and 2.5 $\mu$ m, respectively. Both PM<sub>10</sub> and PM<sub>2.5</sub> are then further disaggregated into particulate species with different properties such as organic aerosols, elemental carbon, sulfate, nitrate and ammonium particles. NMVOCs consist of numerous hydrocarbons with different chemical reactivity and a varying tendency for the formation of secondary particles. To reduce the number of NMVOC species so called photochemical mechanisms have been developed where chemical compounds are lumped together according to their chemical properties. In regional model applications, the most commonly used are the carbon bond mechanisms (Gery et al., 1989, Yarwood et al., 2005, Yarwood, 2010), the Statewide Air Pollution Research Center (SAPRC) mechanisms (Carter, 1988, Carter, 2010, Carter and Heo, 2013) and the Regional Atmospheric Chemistry Mechanisms (RACM) and its predecessors (Stockwell et al., 1990, Stockwell et al., 1997). Global CTMs mostly use specific, slightly simplified chemistry mechanisms.

#### Global and continental inventories

A number of global and regional emission inventories exist. Most of them contain mainly anthropogenic emissions and emissions from biomass burning. Others are dedicated to single

sources, like vegetation fires, desert dust or biogenic emissions. They cover very different timescales, ranging from single years to more than a century. An overview of many of the available inventories can be found at the ECCAD (Emissions of atmospheric Compounds & Compilation of Ancillary Data) home page (ECCAD, 2017). A collection of some of the most frequently used global inventories for anthropogenic emissions together with some details about the covered time span, the species given in the inventory and the horizontal resolution can be seen in Table 1 (see also Granier et al., 2011). The table only contains inventories of more than one reactive substance from more than one sector that cover more than one year from 2000 onwards. It is not meant to be a complete list of all existing global inventories. These inventories are supposed to be available for scientists around the world to be used in chemistry transport model applications.

EDGAR (Olivier et al., 1995, Olivier et al., 2002, Janssens-Maenhout et al., 2010, Crippa et al., 2016, EDGAR2017) is one of the most used global emission inventories (e.g. Solazzo et al., 2012, Bergström et al., 2012, Im et al., 2015). The inventory is provided for a number of emission sectors following the IPCC sectors and as annual data sets, either on a grid map or as country totals. Monthly emissions are available for the year 2010. The data is produced in a bottom-up approach and relies on energy statistics from IEA and agricultural information from FAO. A major advantage of the data set is that the emissions for all years are calculated with the same consistent methodology. On the other hand, this may ignore more detailed country specific information that could improve the emission estimates and their spatial distribution (Janssens-Maenhout et al., 2015).

To overcome this problem the HTAP\_v2.2 inventory (Janssens-Maenhout et al., 2015) followed a hybrid approach where regional inventories for Europe (TNO-MACCII, Kuenen et al., 2014), East Asia (MIX-Asia, Li et al. 2017) and Asia (REAS2.1, Kurokawa et al., 2013) and North America (US EPA, 2016) are combined with global data from EDGAR v4.3. See Table 2 for details about the regional inventories. The drawback of the hybrid approach is that artificial gradients between neighboring regions appear sometimes at the boundary between the global and the regional inventory and that producing time series may be difficult as not all inventories are available for the same years.

MACCcity (Granier et al., 2011; Diehl et al., 2012) is an extended version of the ACCMIP historical emissions dataset that has been developed by Lamarque et al. (2010). Essentially, it is a temporal interpolation of the ACCMIP data set with the RCP8.5 scenario (van Vuuren et al., 2011, Riahi et al., 2007) developed for the IPCC. ACCMIP aimed at creating a best

estimate emission data set for the year 2000 combining global and regional data sets. It has then been extended to years before 2000 by combining the RETRO (Schultz et al., 2007, 2008) and EDGAR-HYDE (van Ardenne et al., 2001) data sets.

The IIASA developed the GAINS model which includes a bottom-up methodology to calculate national emissions by source sector and fuel type (Amann et al., 2011, 2012, 2013). Moreover, a number of worldwide emission scenarios for most of the relevant greenhouse gases and air pollutants, including, the IPCC RCP 8.5 scenario are developed at IIASA (Riahi et al., 2007). The GAINS model explores cost-effective emission control strategies that simultaneously tackle local air quality and greenhouse gases so as to maximize benefits at all scales. GAINS is now implemented for the whole world, distinguishing 165 regions including 48 European countries and 46 provinces/states in China and India. However, as it focusses on policy support and (costs of) control technologies no gridded data are prepared. Within the European ECLIPSE project, global emission fields for the past as well as for the future have been constructed. Three different scenarios for the future exist, which are a baseline scenario, a mitigation scenario and a “no further control” scenario. One set of monthly varying emission patterns is given but there is no further information on modified temporal patterns available for the scenarios. More details can be found in Klimont et al. (2017) and Stohl et al. (2015).

Peking University published a number of emission data sets for various species (Wang et al., 2014, Huang et al., 2014, Huang et al., 2015, Meng et al., 2017, Huang et al., 2017), see Table 1 where they are listed as the PKU inventory. They are mainly based on a global fuel consumption inventory for the year 2007 (Wang et al., 2013), which is available on a  $0.1^\circ \times 0.1^\circ$  grid. For some species the time series has been extended to other years, covering the period from 1960 to approximately 2010, but it is not gridded. Non-combustion emissions are not included in this inventory.

The goal of the newly developed global emission inventory Community Emissions Data System (CEDS) (Hoesly et al., 2017) is to avoid gradients between regions that might appear in hybrid inventories like HTAP\_v2.2 as far as possible. CEDS is based on energy statistics data from IEA and emission factors from GAINS and EDGAR. The idea behind CEDS is to create a community-based emission data system where national or regional inventory developers will be able to improve the quality of the data base. More details can be found in Hoesly et al., 2017. The system provides gridded monthly emissions based on spatial proxies from EDGAR and VOC speciation.



Most of the global inventories are built using a bottom-up approach combining activity data with source specific emission factors. The advantage of such a methodology is that technological developments that influence emission factors can be investigated independently from modified activities. However, the activity data is often based on global statistical data like fuel consumption that is only available on national level and needs to be spatially distributed in a top down approach with appropriate proxy data.

All currently available global emission data sets have their merits and demerits. In fact, many of them follow similar approaches, e.g. they make use of IEA fuel use statistics for emissions related to combustion and then use different sets of emission factors. Several inventories are hybrids, i.e. they are composed of regional inventories that are complemented with global data sets or they use data from other inventories for important substances (e.g. NH<sub>3</sub> or NMVOCs from non-combustion sources) or sectors (e.g. shipping and aviation).

It certainly depends on the actual application which one would be a suitable choice. For investigations of changes in the atmospheric composition over several decades it is necessary to build upon a homogeneous data set that covers the entire time period. However, such a data set might not be the best choice when single years or even shorter time series shall be produced in selected regions. Hybrid data sets will most likely give more accurate emission information in the area where regional inventories are included. However, they might suffer from inconsistencies at the boundaries between the different data sets and the time series are often not consistent. The higher the spatial and the temporal resolution of the emission data set and the more information on speciation (e.g. VOC and PM), as well as emission sectors, is provided the more suitable it is for a number of applications with various CTMs. In general, more details are beneficial for a number of applications. The drawback is that either the file size increases rapidly or a postprocessing of the data is necessary, e.g. to include time profiles or a region dependent VOC speciation. Clearly, the global data sets are missing details in temporal and spatial resolution that are necessary for many regional CTM applications. None of the inventories comes with a time resolution shorter than one month or provides temporal profiles for specific source sectors.

For shipping and aircraft emissions, global inventories exist that treat just these sectors. Because both transport modes are inherently international, they are often not treated well in regional emission inventories. Corbett et al. (2007) and Wang et al. (2008) published global shipping emission inventories, while Eyring et al. (2010) is another widely used inventory and one of the first to utilize a bottom-up approach from ship movement data. Jalkanen et al.

(2009, 2012) and Aulinger et al. (2016) developed regional shipping emission inventories for the Baltic Sea and the North Sea that are based on Automatic Identification System (AIS) ship position data. The same has been done for the Chinese coast by Liu et al. (2016). This approach allows for the construction of a very detailed and rather complete bottom-up ship emission inventory. Corbett et al. (2010) specifically investigated shipping emissions in the Arctic. Recently, a global shipping emission inventory based on AIS ship position data was published by Johansson et al. (2017).

Global emissions from aircraft have been published by Lee et al. (2009) for the year 2005. They also summarize other inventories, e.g. from Sausen and Schumann (2000) and Kim et al. (2007), but they do not provide any spatial maps. A key aspect of aviation emission inventories is emission height. For regional air quality studies the emissions during Landing and Take-off (LTO) are most relevant. LTO includes all emissions below 1 km (3000 feet). For global atmospheric composition and climate applications, aircraft cruise emissions are also relevant. Wilkerson et al. (2010) published gridded data for the years 2004 and 2006 on  $0.5^\circ \times 0.5^\circ$  horizontal and 0.5 km vertical resolution. More recently Wasiuk et al. (2016) published spatially resolved global inventories for the years 2005-2011, extending the time series for 2000 – 2005 from Kim et al. (2007).

## Regional inventories

Some of the widely used regional emission inventories and their characteristics are listed in Table 2. Regional inventories often provide more detailed information about emissions sources, their spatial distribution and also about temporal cycles of the emissions. There are large differences between the individual regional inventories with respect to calculation methods, temporal coverage and number of pollutants included.

Emission data for the European continent is available from the EMEP Centre on Emission Inventories and Projections (CEIP) (CEIP, 2017). CEIP collects the official reported data by the European parties to the convention on Long-Range Transboundary Air Pollution (LRTAP) but also provides gap-filled data for AQ modelling purposes. This gap-filled data (expert emission estimates) is consistent in a way that gaps in the officially reported data have been filled by emission experts. Although the default country emission estimates will be

based on the use of the EMEP/EEA air pollutant emission inventory guidebook (European Environment Agency, 2016a), the methods how the emissions are calculated may vary from country to country. Emission data is available either as gridded data or as country totals. Time profiles to calculate hourly data can be downloaded from EMEP-CTM (2017).

TNO-MACC\_III is another European emission inventory for UNECE-Europe developed in support of the Air Quality forecasting under the Copernicus Monitoring and Atmospheric Composition and Climate programme (GMES 2017, Marécal et al., 2015). TNO-MACC\_III is an extended and improved version of the previous TNO-MACC and TNO-MACC\_II emission datasets (Kuenen et al., 2014; Pouliot et al., 2012; 2015). It uses the officially reported emissions by parties under the CLRTAP to EMEP to the extent possible. In addition, source sector-specific data are used in a harmonized way as well as a consistent gridding methodology at a high resolution. This ensures that patterns across borders do not show sudden changes or jumps. PM10 and PM2.5 emissions are split by source sector and country into its five components; EC, OC, SO<sub>4</sub>, Na and other minerals (Kuenen et al., 2014).

Emission Data for the United States is provided by the United States Environmental Protection Agency (US EPA) (US EPA, 2016) in the US NEI (US EPA, 2017). The NEI is based primarily upon data provided by state, local, and tribal air agencies for sources in their jurisdictions and supplemented by data developed by the US EPA. This information is insufficient for air quality modeling since these models require data that is hourly, gridded, and speciated to a chemical mechanism. The US EPA provides this information in the form of an emission modeling platform. An emissions modeling platform is the full set of emissions inventories, ancillary data files, software tools, and scripts that process the emissions into the form needed for air quality modeling. A Technical Support Document is released with the modeling platform that provides detailed information about temporal allocation, spatial allocation, and the methods used to create a set of modeling files for base and future year modeling (US EPA 2017). Additional details on how the US data was merged are provided in Pouliot et al. (2014) and Pouliot et al. (2015).

REAS is an emission inventory that covers the entire Asia (Kurokawa et al., 2013). It is a bottom-up inventory that contains all pollutants important for CTM applications. Statistical data to derive the activity data is taken from IEA and FAO, but also from national statistics. For Japan, Taiwan and South Korea recent national inventories were considered. Emissions from aviation and shipping are taken from EDGAR. Kurokawa et al. (2013) performed comparisons between REAS 2.1 and other inventories. They reported typical differences in

the order of 25% for annual totals of most emitted pollutants in China. For CO the differences were more than 50% between EDGAR and REAS.

MEIC is a national emission inventory for China developed and maintained by Tsinghua University Beijing (MEIC, 2017). It is a bottom up inventory described in several publications (Wang et al., 2012, Li et al., 2014, Zheng et al., 2014, Liu et al., 2015). It can deliver data in various spatial resolutions via an online portal (Li et al., 2017), but this portal is until now only available in Chinese. Emissions for India and China were reported by Lu et al. (2011). They include SO<sub>2</sub> and primary carbonaceous aerosol emissions from the sectors power generation, industry, transport and residential heating. The inventory is built bottom up from activity data for the four sectors, a spatial gridding is done using proxy data. More details are given in Table 2, where the inventory is included as ANL. Together with MEIC, it is part of the MIX-Asia inventory (Li et al. 2017) that covers entire Asia. Similar to the approach followed for the HTAP v2.2 global inventory, MIX-Asia is a hybrid inventory that combines data from several regional inventories. In addition to REAS, MEIC, and ANL (for India only) national inventories for Japan and South Korea are fed into a larger data set. The inventory gives separate information for the five main anthropogenic emission sectors: power generation, industry, residential heating, traffic and agriculture. Li et al. (2017) performed a comparison between MIX-Asia, EDGAR v4.2 and REAS 2.1. The main differences between MIX-Asia and REAS were found for the industrial sector and for traffic. For most pollutant/sector combinations, REAS revealed higher emissions than MIX-Asia while EDGAR was typically lower. The relative differences for the annual totals can easily be in the order of 50%, irrespective which pollutant or sector is considered. Saikawa et al. (2017) performed a similar exercise with 5 different emission inventories for China. They also found large differences in the total emissions, in particular for CO and when they were broken down to source sectors and regions.

This clearly shows that emission totals are still hard to determine, in particular in rapidly developing countries like China where the annual economic growth can be substantial (leading to increased emissions) and where new emission control technologies may be implemented in a short time (leading to reduced emissions). Inventories in these regions are difficult to keep up-to-date. For other regions of the world, like Africa or South America, hardly any regional inventory exists. Liousse et al. (2014) constructed an inventory for combustion emissions in Africa for the year 2005 and made projections for 2030. When

compared to global RCP and ECLIPSE inventories differences for individual pollutants for the entire continent were in the order of 30 -50 % for 2005.

In addition to the regional inventories, many countries provide emission inventories on very high spatial resolution (down to 1 x 1 km<sup>2</sup>), among them are the United Kingdom (Jones et al., 2017), Spain (Guevara et al., 2013), and Germany (Schneider et al., 2016). These inventories are well suited for small scale air quality studies, however, they are also lacking information about the temporal distribution of the emissions. These inventories are not discussed here.

With the exception of the US NEI, all regional inventories considered here are delivered as gridded data for a number of pollutants and often also for several emission sectors, see the overview in Table 2. Many regional model applications need high resolution emission data, both in time and space. This is one reason why the US EPA developed its emission modeling platform. Until now this approach is not followed anywhere else.

Therefore, additional emission models are needed to further process emission data from inventories and provide them as ‘model-ready’ data. One of these emission models is the SMOKE model developed by the US-EPA (Houyoux, 1998, Houyoux et al., 2000). In recent years there are also a European version of this model called SMOKE for Europe (Bieser et al., 2011) and a version for south east Asia called SMOKE-PRD (Wang et al., 2011). In other cases, the spatial and temporal distribution of the emissions is closely connected with the CTM itself like in the EMEP model (Simpson et al., 2012).

### Point source inventories

Emissions from point sources are often collected in individual registers. This is because the owners of big power plants or industrial plants are required by law to report their emissions if they are above a certain threshold. In Europe this information is collected in the European pollutant release and transfer register (E-PRTR, EEA, 2016). Currently, E-PRTR contains emission information from more than 33000 facilities for the time period from 2007 to 2014. The latter means, that when using the register, it cannot be said if an individual point source is still in operation or if the emissions changed since they were reported. This might not be a big problem when looking at a larger area, however, if small regions are under investigation, it

needs to be verified if the data about the point sources in the selected region are still valid. In addition, emissions that are below the reporting threshold are not included in the register although they might be relevant. They will be included in national emission totals but they won't be allocated to the specific point source.

Point source emissions need to be treated in a special way because the emission height may often be significantly above ground level. In addition, the exhaust gas leaves the stack with a certain exit velocity and at a temperature far above the ambient temperature. This needs to be taken into account in order to assign the emissions to the correct vertical atmospheric layer. Moreover, because of the high temperature and the high concentrations of pollutants chemical reactions inside the plume will differ from those in the highly diluted surrounding environment. Therefore it needs to be taken into account how the reported amount of the emitted pollutants has been derived. For example, if they were measured at high temperatures some of the condensable pollutants may still be in the gas phase although they will rapidly form particles in the atmosphere. Others will quickly be oxidized when they enter the atmosphere which means that the chemical composition will be quickly modified.

The US EPA NEI provides stack information for each release point in the inventory. This information can be used with a plume rise algorithm such as the Briggs formulation (Briggs, 1969, 1971, 1972) found in SMOKE (SMOKE, 2017), to estimate emissions at each layer of the modeling domain using hourly meteorological conditions. Plume rise may also be calculated inside CTMs, however, the necessary information about the individual stack still needs to be provided from the emission inventory. Information about stack properties is rare. E-PRTR only contains information about the location of the source but not about the source properties. None of the global or regional inventories listed in Tables 1 and 2 contains stack properties for point sources.

## **Spatial Disaggregation**

Horizontal distribution

Most of the emission inventories are provided as gridded data sets on spatial resolutions between 0.1 x 0.1 degrees (e.g. EDGAR) to 0.5 x 0.5 degrees (e.g. the ECLIPSE emissions, see Tables 1 and 2). However, atmospheric chemistry transport models often use other spatial resolutions and other map projections. In such cases, the emission data needs to be re-gridded to fit to the actual model grid in use.

This re-gridding can be done in several ways. In HTAP\_v2.2 for example, the emission data from the regional inventories is first split into a much finer grid (e.g. by dividing a grid with 0.25 x 0.25 degrees into 25 grid cells with 0.05 x 0.05 degrees horizontal extension each) and then recombined to the target grid (e.g. by combining 2x2 grid cells to get a new grid resolution of 0.1 x 0.1 degrees). This can be regarded as a bilinear interpolation method where no new or additional information about the emission sources is taken into account. It can be applied in a similar way to produce emission fields on any required grid. However, it may neglect some obvious features of emissions from certain sectors, i.e. emissions may be mapped on areas without any sources.

Other methods use spatial information that is related to the emissions in order to distribute them onto a new grid. One example is related to emissions from residential heating. The assumption is that this depends on the number of people living in a specific area. Therefore, the emissions from this sector would first be aggregated for a larger area, e.g. for a specific country, and then newly distributed following population density maps on a very high resolution grid (down to 1 x 1 km<sup>2</sup>). Next, the emissions can be aggregated to the desired grid. This method ensures a much more realistic distribution of the emissions of a specific sector and a lower spread into emission free regions.

The inclusion of non-static spatially resolved data may add even more detail to the emissions of a certain sector. This could be done for residential heating emissions, which will depend on outside temperatures. Given that information about the heating demand in relation to the ambient temperature is given, emissions from this sector can be distributed according to spatially varying temperature data. This will be available for nearly every three dimensional chemistry transport model that is driven with meteorological data. Examples for this method are described in Aulinger et al. (2011) and Mues et al. (2014). This approach includes a temporal distribution of the emissions according to the temperature data.

The spatial redistribution of emissions from a certain sector is a common practice. See for example the methods described by Backes et al. (2016) for agricultural emissions. Here,

spatially distributed information about land use, animal density and again meteorological information has been used to distribute the total ammonia emissions in space and in time. The agricultural emissions were split into those from manure management and from agricultural soils. Emissions from manure were further split into animal types and housing types. This allowed using animal density maps and specific emission factors depending on the types of barns and on meteorological conditions. Soil emissions were spatially distributed according to vegetation information. After splitting the agricultural emissions like illustrated in Fig. 1 the emissions were redistributed considering specific characteristics of each of these subsectors and using meteorological parameters to calculate the emission flux. This concerned a spatial and a temporal distribution of the emissions at the same time. This is a consequence of taking meteorological data into account that varies simultaneously in time and space.

When smaller regions are modelled, it is easier to consider detailed information about the location of specific emission sources or to include information on fleet composition on specific streets. This can modify the emission inventories and subsequently the CTM results quite substantially (Vedrenne et al., 2016).

### Vertical distribution

Although it has a large influence on the dispersion of the emissions, the emission height is often unknown and therefore not well considered in emission data sets. This is very important for point source emissions from high stacks where, due to exit velocity and the exhaust gas temperature, effective emission heights can reach several hundred meters. Therefore, the emissions can be well above the top of the planetary boundary layer resulting in efficient long range transport and only small effects on the local environment. This depends on the meteorological conditions and large differences between daytime and nighttime as well as between seasons can be expected. The vertical development of an emission plume can be calculated as a function of stack characteristics, often called stack parameters (e.g. exit velocity, stack height, stack diameter, flue gas temperature), and ambient meteorological conditions (Briggs, 1969, 1975, 1984, Raffort et al., 2015).

Based on plume rise calculations implemented in the SMOKE model (Houyoux, 1998, 2000) Bieser et al. (2011a) calculated a set of more than 40,000 vertical emission profiles for six



pollutants taking source sector, climatic or political region, seasons and day- and nighttime into account. These profiles were then combined into 73 different groups by means of hierarchical cluster analysis for further use in CTMs. The advantage is that individual plume rise calculations for each stack, which are computationally expensive in the aggregate, can be avoided without neglecting the high temporal and spatial variability of vertical emission profiles. Bieser et al. (2011) showed that the difference in SO<sub>2</sub> and SO<sub>4</sub> concentrations at ground level were 1-6% and 1-2%, respectively, when the average vertical profiles were used or a full plume rise calculation was applied.

Fig 2 shows the range of emission heights for 5 SNAP sectors and compares the emission heights derived with SMOKE with the formerly used heights from the EMEP model. The EMEP profiles are based on stack data estimated for selected stacks in Zagreb, Croatia (Vidic, 2010). They may not be representative for other European regions. Their vertical resolution with 6 layers between 92 m and 1100 m is too coarse for regional CTMs which have 20–40 vertical layers with near surface layer heights between 20 m and 60 m. In addition they are annual averages that do not consider diurnal and seasonal cycles. Bieser et al. (2011) could show that the difference in SO<sub>2</sub> and SO<sub>4</sub> concentrations at ground level were 1-6% and 1-2%, respectively, when the new vertical profiles were used or a full plume rise calculation was applied.

Guevara et al (2014) investigated the effect of plume rise calculation with the Community Multiscale Air Quality (CMAQ) model on effective emission heights for a number of stacks in Spain and found that online calculations improve the modeled concentration fields. Their effective emission heights were comparable to those found by Bieser et al. (2011a). However, they pointed out that individual stack data like height, exit velocity and exit temperature should be available. This is usually not the case but a database containing this information could help modelers around the world to improve their simulation results.

Typically, high stacks are at fixed places and therefore it makes sense to characterize the vertical emission height by political or climatic region. With the upcoming bottom-up emission inventories for shipping (e.g. Jalkanen et al., 2009, 2012, Aulinger, 2016, Johansson et al., 2017), new methods need to be developed to vertically allocate the emissions from big ships. The stacks of big ocean going vessels can easily reach 50 m height or even more, therefore the effective emissions might reach the 3<sup>rd</sup> or 4<sup>th</sup> model layer in many CTMs. Mason et al. (2008) used a plume rise algorithm within SMOKE to calculate the effective emission heights for a 20 m stack and found that on average about 2/3 of the emissions were between

37m and 75m while 30% were between 75m and 150m and only few percent below. One of the difficulties in treating these emissions correctly is that they belong to a ‘moving stack’, which means that the ship velocity and the spatially varying environmental conditions need to be taken into account.

Emissions from aviation need special attention concerning their vertical distribution because the emission source itself changes its vertical position over time. In addition the exhaust gas is hot and will be subject to plume rise in the colder atmosphere. Until now, simple approaches have been established that vertically distribute the LTO emissions with most of them being in the lowest model layer and then decreasing their share with height.

### **Temporal Disaggregation**

One important goal of emission modeling is to improve the representation of the temporal distribution of the emissions and develop, in addition to spatial surrogates like population density, also temporal surrogates for accurately representing the observed changes in emissions on various time scales. In order to correctly account for the dispersion and the chemical conversion of the emitted pollutants, a correct timing of the emissions is crucial. For example, the height of the planetary boundary layer, wind speed, and radiation undergo significant changes between daytime and nighttime and all of them are highly relevant for the distribution of the emitted pollutants. In addition, many sources have strong temporal patterns, with variations ranging from hourly to seasonally.

Recent emission inventories like HTAP2.2, ECLIPSE or REAS 2.1 cover seasonal differences and provide monthly emission maps, but most of the inventories provide only annual totals. The temporal variation of specific source sectors is then taken into account by applying typical time profiles and thereby distributing the emission data in time. These are weighting factors that are derived from temporal activity profiles and the sum of which is 1. Among the frequently used time profiles are those disseminated and published by Denier van der Gon et al. (2011). They represent average time profiles for Europe. In Fig. 3 the time profile for the emissions from the 10 SNAP sectors can be seen. They are divided into monthly, weekly and daily variations. The daily variation of traffic emissions for example shows a strong morning rush hour peak and a less confined late afternoon peak when people return home. A similar

pattern but somewhat shifted in time can be seen for SNAP 2 (combustion for residential heating). Using this method of a three-step temporal disaggregation the daily profiles will be the same every day and the weekly profiles the same in every season. These simplifications are useful but certainly unrealistic. On weekends for example, the driving cycle changes significantly and differences between Saturday and Sunday need to be taken into account. Traffic on highways may have other time profiles and working hours may be different even within a country (e.g. between Northern and Southern regions in Italy and France). Therefore, more sophisticated methods need to be developed to account for the temporal emission variations. In recent years, those methods were developed for some emission sectors.

For example, recent publications propose new methods to improve the spatio-temporal allocation of ammonia emissions to model grid cells. These publications make use of animal density and vegetation maps, and take into account factors such as type of animal housing, fertilizer application schedules, and meteorological conditions, to develop grid specific time profiles (Skjoth et al., 2011, Backes et al., 2016, Hendriks et al., 2016). Backes et al. (2016) used the temperature and wind speed dependence of ammonia emissions from open and closed barns originally given by Skjoth et al. (2004) and combined them with meteorological data to derive temporally resolved  $\text{NH}_3$  emissions for Central Europe. In addition they extended the manure application model from Skjoth et al. (2011) by policy restrictions for closed periods, which differ from country to country. They found an improved annual time profile for  $\text{NH}_3$  emissions from agriculture that varies from grid cell to grid cell.

A similar approach was followed by Hendriks et al. (2016), who used the same temperature dependent functions from Skjoth et al. (2004) for ammonia emissions from animal house keeping and fertilizer application. In addition, they used manure transport data for 2007 – 2011 in Flanders/Belgium to improve the annual profile of  $\text{NH}_3$  emissions in the Netherlands, Belgium, North East France and West Germany. Fig 4 (left) demonstrates that the maximum of  $\text{NH}_3$  emissions from manure application is in April/May, which is about 4-6 weeks later than the standard annual time profiles suggest. The summer minimum and the late summer maximum are very similar to what was found by Backes et al. (DTP in Fig 4, right) and significantly different from the standard profiles used before (STP in Fig 4, right). Hendriks et al. also found significantly improved correlations for  $\text{NH}_3$  concentrations when they compared their LOTOS EUROS model results to observations.

Another example is residential heating, where regional preferences for certain fuel types (e.g. wood or coal) together with ambient temperatures can be taken into account for more realistic emission data than would be possible with static annual profiles for each model grid cell.

Aulinger et al. (2011) introduced such a method for emissions of benzo(a)pyrene from residential heating. They used the relationship between heat supply and ambient temperature in Hamburg to define a linear function that describes the increase in heating demand for temperatures below 18 °C. For temperatures higher than 18 °C, a minimum amount of emissions was set that depends on the probability that temperatures below 18 °C occur.

Bieser et al. (2011) generalized this approach for all emissions from the sector residential heating and implemented it into SMOKE for Europe. As an example, Fig. 5a) shows the spatially averaged temporal profile of CO emissions in Europe in a daily resolution. The default profile uses monthly varying factors overlaid with changes between working days and weekends. Significant changes between the last day of a month and the first day of the next month are clearly visible in the time series. These unrealistic variations are removed when the static monthly factors are replaced by temperature dependent factors that vary on a daily basis. With this method, the heating demand in the course of a year can be followed and different years will have different temporal profiles throughout the year (see Fig. 5b).

Mues et al. (2014) followed a very similar approach where every daily average temperature below a threshold of 18°C is taken in the annual heating demand for each grid cell. For each day and each grid cell this results in a daily emission scaling that depends on the daily temperatures in that particular grid cell. This approach may also introduce changes in the spatial distribution of the emissions. This is shown in Figure 6 for NO<sub>x</sub> and PM<sub>2.5</sub> in Europe in 2009. Both approaches can be transferred to other parts of world, provided that the share between emissions related to heating and those for hot water production stemming from the same heating system can be assessed.

Menut et al. (2012) looked at the impact of more detailed daily time profiles of NO<sub>x</sub> emissions from traffic in several European cities. They developed regionally differentiated daily variations of traffic emissions and quantified the changes in NO<sub>2</sub>, O<sub>3</sub> and PM concentrations with the Chimere model. Modelled NO<sub>2</sub> concentrations were significantly affected showing higher peak values and also higher mean values. Consequently, the ozone concentrations decreased slightly. PM<sub>10</sub> was on average slightly higher than in the case with

standard time profiles. In comparison to observations, the model bias for PM<sub>10</sub> and O<sub>3</sub> in selected cities was slightly reduced, but it remained high.

The effects of improved temporal profiles for traffic emissions in Germany have also been looked at by Mues et al. (2014). The correlation coefficients (based on both, hourly and daily concentration values) were improved for NO<sub>2</sub> concentrations at urban background stations but the effects on annual mean values were only minor.

Mues et al. (2014) also investigated the combined effects of improved temporal profiles for emissions from power generation (SNAP1), non-industrial combustion (SNAP2) and traffic (SNAP7). The largest improvements in correlation coefficients for NO<sub>2</sub>, SO<sub>2</sub> and PM<sub>10</sub> were found when new time profiles were used for all sectors simultaneously. This points to the fact that small improvements in several emission sectors can lead to cumulative and significant improvements in modelling concentration time series. Nevertheless, the modelled annual biases for these species remained almost constant.

These examples for the improvements for temporal profiles for selected source categories demonstrates the positive impact of a detailed and more realistic representation of the temporal variability of emissions on the modelled concentrations of the respective pollutants. In comparison to observations, the correlation coefficients can be significantly improved (Menut et al., 2012, Mues et al., 2014). In addition, model biases are often reduced as well, but to a minor extent. Peak values were more influenced than average values. This is not very surprising, because the total emissions remained constant in the model experiments. On the other hand, peak values are often of high importance, e.g. when compliance with threshold values is investigated.

## **Bottom-up Emission Calculations**

In a strict interpretation of bottom-up emission modeling, creating an emissions inventory requires calculating the emission flux of every single source, for example a combustion engine, including its temporal dynamics. Thus, the energy production of this engine must be available as a function of time, and further, energy specific emission factors that are dependent on the engine type, the fuel type and in many cases also on the engine state or

engine load. These emission factors are given in g/kWh. Instead of engine specific emission factors some models use fuel specific emission factors. In that case, the fuel consumption is calculated from the energy production of the engine and the emission factors are given in g/kg fuel.

Point source emissions from power plants can be calculated this way. The necessary data are its energy production per unit of time, the fuel used and the end-of-pipe technology installed (Adolph et al., 1997). Additional information about exhaust gas temperature, stack height and weather conditions need to be known, in order to simulate the rise and dispersion of the exhaust plume (Raffort et al., 2015). It is, then, possible to calculate the mass of fuel burned and to apply fuel specific emission factors to calculate emission fluxes of different pollutants. Such emission factors are available for all common fuels and for the most important pollutants.

For power plants, it is often justified to use emission factors that are independent of the load of the combustion device because combustion units for energy and heat production run under constant operating conditions. This is not the case for combustion engines where the emissions per energy demand depend strongly on operation modes such as traveling at constant speed, start-up or maneuvering. Models for calculating emissions of ship diesel engines, for example, use emission factors that are functions of the engine load (Larsen et al. 2015). As different engines have different power uptake characteristics, in sophisticated models these functions are separated according to fuel type, engine type, maximum engine power, year of build and ship size expressed in gross tonnage or TEU (Aulinger et al., 2016). The temporal evolution of a vessel's energy demand can be derived from its activity profile. This is usually divided into sailing, maneuvering and berthing and can easily be tracked in case the vessel broadcasts its position, movement status and speed via AIS (Jalkanen et al., 2009, Goldsworthy and Goldsworthy, 2015, Aulinger et al., 2016, Chen et al., 2016). Large and commercial vessels are obliged to send such signals for security reasons. Thus, it is possible to calculate emissions for individual ships at nearly arbitrary temporal resolution, which constitutes a typical low level bottom-up model. In even more detailed models, external factors that influence the power uptake, and thus energy demand, can also be taken into account (Jalkanen et al., 2012). These are resistance factors such as fouling, wind speed and direction, wave height and currents. However, these parameters are usually not available from measurements at the required temporal or spatial resolution and must therefore also be

modeled or estimated, which bears the risk of introducing large uncertainties into the emission model.

Estimating emissions from road traffic can be done in a bottom-up approach, in particular estimating local emissions with micro-scale traffic models (Wang and Fu, 2010, Rakha et al., 2004). It is, however, hardly possible – at a reasonable effort – to follow the movements of single vehicles in real time road traffic inclusive of their type and engine type. Therefore, the activity profiles of motor vehicles in daily traffic are calculated by models before they are used for emission calculations (Abou-Senna et al., 2013). This is still a bottom-up approach even if not at the lowest level. One way to derive activity profiles is to evaluate traffic statistics, which can be as detailed as determining the speed and driving characteristic of vehicles between two road segments (Zhang et al., 2016). Another way is to simulate the behavior of the different road users by analyzing the traffic demand (Alam et al., 2014). This can be done using assumptions on the delivery of goods between different industrial regions or metropolitan areas. Then, the share of these goods transported by light and heavy duty vehicles must be estimated. Another example for assumptions made in road user models is the share of cars used for commuting between living and industrial areas. The time profile of these activities is derived from business and working hours or evaluating traffic statistics (Cardelino, 1998, Chen et al., 2016). The results of these travel simulations can then be treated like following the activities of single vehicles. Considering also the types of roads used, the expected traffic density and their orographic profile, which influences the energy demand and, hence, fuel consumption of the vehicles (Jackson and Aultman-Hall, 2010) allows applying fuel specific emission factors for calculating pollutant emissions.

Area sources like ammonia emissions triggered by the application of fertilizers to arable land (Reidi et al., 2008) can be calculated in a bottom-up approach, as well. In this case, the amount and type of the fertilizer must be known, also the area of the treated soil, the time and duration of the treatment and the treatment method (Roelle and Aneja, 2005). The method of applying the fertilizer, such as spreading liquid manure with planes, tractors or injected directly into the soil, mostly determines the amount of ammonia that escapes into the atmosphere during application (Nyord et al., 2008). Weather conditions and type of crop grown determines the ratio of ammonia that evaporates following the fertilization. As it is hardly possible to monitor the fertilization at every single piece of arable land, most of the model parameters are subject to assumptions or estimations. The dates of fertilization, as an example, are derived from the local legislation, the climate and the types of crops grown

(Backes et al., 2016). This yields information about the activity of the area source whereas the emission strength or emission factors (in grams or moles per area and time) are estimated by evaporation models.

It is evident that bottom-up emission models can have different levels of detail or granularity. The reason is that often not all parameters needed for a pure bottom-up approach are known. Generally, when there are fewer variables which define the emissions process that are accessible to direct measurements, there are more assumptions about an emission source that must be made. On the one hand, it is desirable for sophisticated bottom-up emission modeling to include as many variables as possible. However, on the one hand, this bears the risk of over-fitting the model, which can happen, when data are included that are biased or bear large uncertainties (Hawkins, 2004). In that case, the uncertainty of a complex emission model increases in comparison to a simpler approach and it may be advisable to choose the simpler approach. Bottom-up and top-down modeling approaches are often combined, i.e. temporal profiles and spatial variability are modelled using detailed information, e.g. meteorological or traffic data, while the total amount of the emitted substance stems from national or continental statistics. Examples for this were given before (Aulinger et al., 2011, Backes et al., 2016, Mues et al., 2014).

## **Natural Emissions**

Natural emissions can be responsible for a large fraction of the total emissions of certain substances, e.g. for VOCs and PM. They need to be included in chemistry transport model calculations, not only because of their share in the emission totals but also because of their interaction with anthropogenic emissions. Consequently, the correct spatial and temporal distribution is of similar importance as it is for anthropogenic emissions. Natural emissions typically depend on meteorological conditions and on land use. Often, special models exist for biogenic and dust emissions.

### **Biogenic emissions**



Biogenic VOCs (BVOCs) are an important atmospheric constituent significantly affecting atmospheric chemistry. They contribute to both gas phase chemistry (Atkinson and Arey, 2003) and heterogeneous chemistry of aerosols and clouds (Hallquist et al., 2009) in the troposphere. BVOCs strongly influence ozone and secondary aerosol formation, both of which ultimately influence air quality and radiative forcing (Andreae and Crutzen 1997 , Kanakidou et al., 2005 , Arneth et al. 2010). In the boreal forest, 70-90% of the water-soluble organic aerosol can be attributed to secondary products of BVOC oxidation (Cavalli et al., 2006). CH<sub>2</sub>O and O<sub>3</sub> are among important secondary products in the gas phase (e.g. Curci et al., 2009). Terrestrial vegetation is the dominant source of all VOCs in the atmosphere accounting for about 90% of the total emission, and isoprene and monoterpenes are the most abundant species among the biogenic VOCs (Sinderalova et al., 2014). In the marine environment, isoprene is produced by both phytoplankton and seaweed. The total global oceanic emissions of isoprene are estimated to be in the range 0.27-1.7 TgC/yr (Arnold et al., 2009) which is at maximum 0.4 % of the estimated global terrestrial isoprene source of 400-750 TgC/yr (Müller et al., 2008, Guenther et al., 2006). In the urban environment BVOCs are usually less important than anthropogenic VOCs, but they might influence urban NO<sub>x</sub> and O<sub>3</sub> budgets under specific conditions (Lee et al., 2006, Churkina et al., 2017).

Quantitative estimates of BVOC emissions are needed to effectively model air quality. These estimations are challenging due to the large number of compounds and biological sources involved. The implementation of BVOC emission inventories in atmospheric CTMs is further complicated by the high complexity of in-canopy processes such as chemical reaction, aerosol formation, deposition and turbulent exchange. Due to their high reactivity, BVOC emitted from foliar biomass are partially converted to secondary gas-phase products and particulate products before actually entering the lower atmosphere. The vertical resolution of 3-D atmospheric chemistry models is usually too coarse (20-40 m) to fully capture the vertical variation in the production of secondary aerosol precursor species (Saylor, 2013). BVOC emissions and their biological and chemical diversity are comprehensively reviewed by Guenther (2013). The overview also offers strategies for improving BVOC emission modeling approaches by focusing on better representations of the underlying diversity. Quantitative emission models also need to account for all the processes that lead to and control the variability of emissions.

In order to quantify the terrestrial biogenic emission of isoprene on regional but also on the global scale and with high spatial resolution (~1 km) the Model of Emissions of Gases and

Aerosols from Nature (MEGAN) was compiled (Guenther et al. 2006). Via several steps this modelling system was extended and more compounds and processes have been considered. MEGAN 2.1 is now able to account for 149 known compounds (Guenther et al., 2012, Guenther, 2013). MEGAN is driven by land cover, weather, and atmospheric chemical composition but it does not consider secondary gas phase and aerosol products that are formed before they enter the lower atmosphere. MEGAN 2.1 components are shown in Figure 7. MEGAN can either run stand-alone for generating emission inventories or can be incorporated as an on-line component of chemistry transport models (prepared for CAMx and CMAQ, WRFchem; e.g., Pouliot and Pierce, 2009, Zhao et al., 2016). A 30-year global BVOC emission dataset based on MEGAN 2.1 is presented by Sindelarova et al. (2014).

The BEIS system, which has been a component of the CMAQ (Byun and Ching, 1999, Byun and Schere, 2006) modelling system for a long time (Pierce and Waldruff, 1991, Pierce et al., 2002, Byun and Schere, 2006, Appel et al., 2017), has been further developed. All BEIS-3 versions (Vukovich and Pierce, 2002, Schwede et al., 2005) are designed for use with the SMOKE system of CMAS. Bash et al. (2016) describe the latest version (BEIS3.6.5) and discuss a thorough evaluation of the system using BEIS emissions in CMAQ v5.02 for a Californian case study.

MEGAN and BEIS both estimate BVOC emissions following the empirical algorithm initially developed by Guenther et al. (2006). The emission factors between MEGAN and BEIS differ as MEGAN uses emission factors for 16 different global plant functional types (Guenther et al., 2012) while BEIS uses species- or species-group-specific emission factors where available and MODIS plant function types where no species-specific data are available (Bash et al. 2016). The variability in BEIS emission rates is greater than in MEGAN 2.1 (Guenther et al., 2012) due to the more detailed representation of vegetation species.

GloBEIS is a biogenic emissions modeling system based on the BEIS emission factors and algorithms but with an easier-to-use interface and compatibility with a wider range of input data sources and enhanced algorithms including canopy environment, leaf age, variable LAI, and the influence of antecedent temperature conditions (Yarwood et al., 1999; 2010).

GloBEIS was developed to allow users to estimate biogenic emissions of volatile organic compounds, carbon monoxide, and soil NO<sub>x</sub> emissions for any time scale and domain. Some reserachers select GloBEIS because it uses vegetation species specific emissions factors, requires minimal inputs and can be run on a desktop computer. The emission factors used in

GloBEIS are the same as those reported by Guenther et al (1995) and the MEGAN model, but expressed at the leaf level and not the canopy level (Drewniak et al. 2014).

A detailed description of the differences in model approaches of GloBEIS, BEIS and MEGAN and results of an emission comparison in the US are provided by Sakulyanontvittaya et al. (2012). The model results varied considerably for the various species, depending on model version, time and location. In an earlier study, Sakulyanontvittaya et al (2010) found that isoprene emissions between GloBEIS and MEGAN were comparable.

The first comprehensive regional BVOC emission inventory for Europe has been set up by Simpson et al. (1999) and implemented in the EMEP MSC-W chemical transport model. Simpson et al. (1999) established a national estimate of species distribution of 32 vegetation types in Europe by using statistical inputs at the national scale for 37 countries. More recently the BVOC emission inventory in the EMEP model has been updated (Simpson et al., 2012), using BVOC emission factors for forests created from the map of forest species generated by Köble and Seufert (2001). This work provided maps for 115 tree species in 30 European countries, based upon a compilation of data from the ICP-forest network (ICP FORESTS, 2017).

ICP tree maps were also used together with additional land use information on agriculture and other vegetation, to develop a BVOC emission inventory for Europe. This has been done with special regard to the plant-specific land use data for the use in chemistry transport models. The inventory and its evaluation as well as comparison with other inventories is described by Karl et al. (2009). Another BVOC emission inventory for Europe aiming for an improved seasonality and land-cover component was developed by Oderbolz et al. (2013). Further sensitivity experiments show that land surface schemes do influence the simulated BVOCs, but the impact is much smaller than that of vegetation distributions (Zhao et al., 2016).

Most atmospheric chemistry schemes include at most only a few BVOCs and may lump these together with other compounds which limits the advantages of a detailed emissions chemical speciation. The increased number of compounds is a disadvantage as there might be a significant increase in the computational resources associated with emissions parameterization, processing inputs, and emission calculations. Performance depends partly also on the region of interest, in which certain processes are more relevant than other or than BVOC categories used.

## Dust emissions

Atmospheric or wind-blown dust, also called *aeolian dust*, comes from arid regions, where near surface meteorological (i.e. mean wind velocity, turbulence intensity), and surface conditions (i.e. soil texture, particle size distribution crusting, soil moisture, vegetation protection) allow for an initial lifting process (e.g. Tegen et al., 2002; UNEP, WMO, UNCCD, 2016). About one third of the global land area is covered by dust-producing surfaces, mainly deserts and drylands (Jickells et al., 2005). Direct human actions like off-road vehicles or soil disturbance caused by grazing can contribute to enhanced dust emissions important on locale scales (e.g. Gilles et al., 2005, Neff et al., 2008, Tegen et al., 2004). Estimates of the potential dust emissions vary significantly, ranging between 500 to 4400 Tg per year (e.g. Engelstaedter et al., 2006, Posfai and Buseck, 2010, Huneus et al., 2011)

Dust influences a wide range of atmospheric physical, chemical and biogeochemical processes, including the marine and terrestrial biosphere through the transport of nutrients like iron (Mahowald et al., 2010) and phosphorus (Nenes et al., 2011) and can thereby after air-sea deposition modulate ocean carbon storage and finally atmospheric CO<sub>2</sub> concentration (Schulz et al., 2012). Furthermore, mineral aerosols are important for air quality assessments through their impact on visibility and concentration levels of PM (Huneus et al., 2011).

Dust in the atmosphere is predominantly produced by saltation (bouncing particles) and sandblasting of sand-sized grains. Once the wind speed reaches a certain critical value, termed the *impact* or *fluid threshold*, the drag and lift forces exerted by the fluid are sufficient to lift some particles from the surface. Typical Aeolian transport modes important for air quality and climate process modelling are the short term suspension (particles between ~ 20 and 70 micrometer) and long term suspension (particles < 20 micrometer), the latter of which can remain in the atmosphere up to several weeks (Kok et al., 2012).

### *Schemes, parameterizations and modelling*

To consider atmospheric dust concentrations in chemistry transport models the correct description of the spatial and temporal variability of dust emission occurrences and intensities is a prerequisite. Explicit dust emission models provide a physical description of the main processes involved in dust production. Key elements of model dust emission schemes are largely based on empirical data from wind tunnel experiments. Accurate surface and soil

databases and the use of most pertinent meteorological fields at the relevant scales are required to model mineral dust fluxes with acceptable quality. Uncertainties are often associated with the model's capacity to realistically reproduce the near surface meteorological conditions. Elements, parameterizations and input data retrieval for a process-oriented model are discussed by Laurent et al. (2009).

Many weather, climate and chemistry transport models use empirically derived parameterizations to account for dust emissions. As an example the more physically/numerical modelling based parameterization by Kok et al. (2014) can be mentioned. It is easy to implement into larger-scale models, since it depends only on the wind friction velocity, the soil's threshold friction velocity and the soil clay content. In addition, it takes into accounts a soil's increased ability to produce dust under saltation bombardment as it becomes more erodible and the increased scaling of the dust flux with wind speed as a soil becomes less erodible. These are features missing in many other parameterizations (Kok et al., 2014). The authors claim that their parameterization reduces the need for empirical dust source functions (e.g. Tegen et al., 2002) that are often used in dust modules of larger-scale models, like e.g. in the GOCART model (Ginoux et al., 2001). Evan et al. (2015) examine two dust simulations made with the Weather Research and Forecasting with Chemistry (WRF-Chem) model using the GOCART scheme and the Kok et al. parametrization, see also Zhao et al. (2013). The models show disagreement in the relative importance of identified emitting regions with respect to emitted amount and frequency of events, underlining existing uncertainties. Three different state-of-the-art dust emission schemes were evaluated with observed dust fluxes in a performance test using a box model described by Haustein et al. (2014). It was found that a key-parameter for mismatch is surface crusting that limits the availability of erosive material, even at higher wind speeds, the second-most important parameter is the soil size distribution. Three different dust parameterization schemes with different physical assumptions used in connection with WRF-Chem were assessed by Flaounas et al. (2017), these authors report also on tuning sensitivity studies to better match observed vertical profiles.

There are several studies that examine different aspects of dust emission modelling. For example, Zhao et al. (2013) used the WRF-Chem model with three different approaches to represent dust size distribution (8-bin, 4-bin, and 3-mode) to investigate its influence on different dust parameters. An uncertainty of a factor of 2 is quantified in dust emission estimation due to the different size parameterizations. The WRF-Chem sensitivity to vertical

resolution during a transport study of Saharan dust was investigated by Teixeira et al. (2016), dust emissions were based on GOCART (Ginoux et al., 2001). As might be expected, the best match with observed vertical profiles was observed when employing a high number of vertical layers in the troposphere (80 to 100).

The CMAQ model version 5.2 (CMAQ, 2017) contains an updated windblown dust algorithm that is described in Foroutan et al. (2017). The scheme incorporates a dynamic relationship for the surface roughness length relevant to small-scale dust generation processes. The effect of non-erodible elements on the local flow acceleration, drag partitioning, and surface coverage protection is modeled in a consistently formulated and physics based manner

#### *Data sets and forecast*

Dust mineral fractions at emission are made available for download by the Goddard Institute for Space Studies (NASA-GISS, 2017). Eight mineral species in five size ranges in a resolution of 5' longitude by 5' latitude are considered. The underlying modelling is described by Perlwitz et al. (2015a; 2015b). The Barcelona Dust Forecast Center (BDFC) offers regional dust forecast fields for Northern Africa, the Mediterranean and the Middle East using the NMMB/BSC-Dust model continuously on a daily basis. The model is described in Pérez et al. (2011) and Haustein et al. (2012). Output data contains besides the different dust related variables an emission flux. Historic data (starting 2000) is also available and can be downloaded upon request (BSC, 2017).

#### Emissions from soil

Soil is not only a source of dust particles but also of oxidized nitrogen compounds. Nitrification and denitrification of reactive nitrogen in soil by microorganisms leads to formation of nitrogen monoxide (NO) and nitrous oxide (N<sub>2</sub>O). They are emitted to the atmosphere and are responsible for up to 21 Tg N NO (Davidson and Kingslee, 1997) and 6.6 Tg N N<sub>2</sub>O (Syakila and Kroeze, 2011) annual global emissions. This accounts for about 15% of the NO and 37% of the N<sub>2</sub>O total emissions. N<sub>2</sub>O is an important GHG but because of its low reactivity, N<sub>2</sub>O is often neglected in chemistry transport modelling.

Two main approaches exist in soil emission modeling. One approach is a simple parameterization based on macroscale variables like land use classification and temperature such as presented by Yienger and Levy (1995) and implemented in standard emission models like BEIS. Yienger and Levy presented an emission function that is an exponential function of temperature fitted to the results of chamber soil emission measurements with biome-specific parameters. It can be described by temporally resolved model variables such as temperature and land use classification and may be altered and modified with functions for rain-pulsing emissions, fertilization, fire-fertilization and canopy reduction.

The other approach is a more sophisticated nutrient load simulation model that calculates nitrification- and denitrification rates and resulting gaseous nitrogen emissions. Examples for those models are EPIC (Williams et al., 1998) and DNDC (Li et al., 1992). They are used as separate preprocessors for emission inventories and calculate the emission rates based on temporally resolved information such as meteorological conditions, runoff, fertilizer application, growing- and harvesting times as well as stationary information such as crop type, plant type, root depth and soil properties.

Depending on the data available, NO emissions from soil can be calculated in a preprocessor or inline in the chemical transport model during runtime.

## Sea spray

Sea spray aerosol (SSA) is emitted from the ocean surface primarily by bubble bursting (Blanchard and Woodcock, 1980; Monahan et al., 1986). The emitted droplets contain not only sea salt but also organic matter, that is torn from the surface micro layer (SML) (Gantt and Meskhidze, 2013a; Blanchard, 1964). In the open ocean, entrainment of air and resulting bubble bursting is primarily governed by wind speed. Additionally, sea surface temperature (SST), salinity, and the SML (Lewis and Schwartz, 2004) impact the size distribution. In the coastal surf zone, the wind speed becomes less important and the key driving factors are morphological features such as water depth, slope, and wave breakers (deLeeuw et al., 2000).

Sea spray and, as the most important part of it, sea salt emissions have been a subject of research since the 1960s (Blanchard and Woodcock, 1980; references therein). In the 1980s, Monahan and Muircheartaigh (1980) described a power law relation between wind speed and

white cap coverage (metric for bursting bubbles), and derived a wind-dependent sea salt emission parameterization (Monahan et al., 1986). Lewis and Schwarz (2004) and de Leeuw et al. (2011) give a detailed overview of further research performed on sea salt emission parameterizations in the 1990s and 2000s. In the current releases of the major chemistry transport models, mainly three sea salt emission parameterizations are utilized. Those are an extension of the Monahan model to sub-micron droplet sizes by Gong (2003), an extension of the Gong model to include SST-dependence by Jaegle et al. (2011) and a model with combined parameterizations for ultrafine (according to Martensson et al.), fine and coarse (according to Monahan, 1986), and even larger particles (Smith et al., 1993). Recently, Ovadnevaite et al. (2014) published a new sea salt emission parameterization that considers wind, wave, and salinity data.

Surf zone emissions and emissions from ice are still not sufficiently or not at all represented in modern CTMs. The barrier for not improving surf zone emissions is the lack of detailed coastal morphological data and, additionally, the difficulty to incorporate these data into models that have a coarse spatial resolution. CMAQ and CAMx are two of the few CTMs with surf zone emissions (Gantt et al., 2015a). Several recent model studies exist, showing the relevance of SSA emissions by snow and frost flowers from ice (Levine et al., 2014; Legrand et al., 2016; Xu et al., 2016; Huang and Jaegle, 2017, Rhodes et al., 2017). However, it is still under discussion to what extent snow or frost flowers drive SSA emissions.

Bubble bursting also leads to emissions of marine primary organic aerosol (POA). Marine POA have different chemical and optical properties than pure SSA and facilitate cloud formation (O'Dowd et al., 2004; Wilson et al., 2015). Marine POA emissions have been coupled to sea salt emissions in recent modeling studies (Gantt et al., 2012; Gantt et al., 2015).

## Vegetation fires

Wildfires around the world have increasingly affected human values, assets and ecosystem services, among which air quality is of common interest (Moritz et al., 2014). These vegetation fires are an important source of pollutants, gases and aerosol particles (Langmann et al., 2009). Globally, fire emissions are responsible for 5 to 8% of the annual premature



deaths from poor air quality, and across much of the tropics fire is the primary cause of elevated mortality from air pollution (Lelieveld et al., 2015). Thus, for many regions emissions from vegetation fires should be considered in air quality studies.

The methods for calculating emissions from vegetation fires or biomass burning are comparable to the bottom-up methods for generating emissions from combustion engines. Necessary parameters are the mass of fuel burned per unit of time and area and the type of fuel, i.e. the vegetation types that determine the emission factors for the different pollutants. Seiler and Crutzen (1980) proposed a basic formula:

$$E_i = A(x, t) \times B(x) \times FB \times ef_i \quad (1)$$

This describes the emissions  $E_i$  of substance  $i$  as the product of the area burnt  $A$  at time  $t$  and location  $x$ , the fuel load  $B$ , i.e. the biomass per area, at location  $x$ , the fraction of the biomass that is actually combusted by the fire  $FB$  and the emission factor  $ef_i$  for substance  $i$ . While the area burnt is usually derived from satellite observations, the fuel loads and the fraction burned is derived from land use and vegetation type maps, which are also mostly taken from satellite observations. In order to create emissions in high temporal resolutions many authors recommend the usage of burnt area in combination with active fire detection products (Lioussé et al., 2004; Michel et al., 2005; Hoelzemann et al., 2004; van der Werf et al., 2006; Mieville et al.; Ichoku and Kaufman, 2005). An example is the MODIS Thermal Anomalies Product (Giglio et al., 2006) used by Wiedinmyer et al., (2006; 2011) to create a global inventory with daily emission estimates in 1 km<sup>2</sup> resolution, the Fire Inventory from NCAR (FINN). It is available for download at <http://bai.acom.ucar.edu/Data/fire/>. A large collection of emission factors for all relevant gaseous and particulate species distinguished by vegetation types that can be found in literature is also provided (e.g. Stockwell, 2014; 2015; Yokelson, 2013).

Sofiev et al. (2009) presented an alternative method for calculating emissions of particulate matter from satellite products. They established a direct relation between the 4 μm Brightness temperature anomaly (TA) and fire radiative power (FRP) and empirical emission factors for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively. The emission factors are calibrated by using the fire emissions in a dispersion model (SILAM), comparing the thus calculated concentrations to observations and adapting the emission factors so that the deviations between observed and modelled pollutant concentrations are minimized (Soares and Sofiev, 2014). Emissions of other compounds in the particulate and gaseous phase were calculated using scaling factors for

different vegetation types (Andreae and Merlet, 2001). Annual fire emission inventories produced with this method for the years 2000 – 2015 are available on the internet (IS4FIRES, 2017).

Another recently updated source for emissions from vegetation fires is the Global Fire Emissions Database (GFED), which is now available in its 4<sup>th</sup> version (GFEDv4, Randerson et al. 2015). The database allows one to quantify global fire emissions patterns during 1997 to 2016. The Carnegie–Ames–Stanford Approach (CASA) biogeochemical model was used for its creation, input datasets are largely based on satellite products. The GFED dataset provides global estimates of monthly burned area, monthly emissions and fractional contributions of different fire types. Daily or 3-hourly fields are provided to allow for scaling monthly emissions to higher temporal resolutions. The data on all typical fire emission compounds are given at 0.25-degree latitude by 0.25-degree longitude spatial resolution and are available from June 1995 through 2016 as yearly totals by region, globally, and by fire source for each region (van der Werf et al. 2017).

Emission estimates for C, CO<sub>2</sub>, CO, CH<sub>4</sub>, H<sub>2</sub>, N<sub>2</sub>O, NO<sub>x</sub>, NMHC, OC, BC, PM<sub>2.5</sub>, TPM, SO<sub>2</sub>, and dry matter (DM), were derived by combining burned area data with results of the revised version of the biogeochemical model CASA-GFED, that estimates fuel loads and combustion completeness for each monthly time step (van der Werf et al. 2010; van der Werf et al. 2017). Daily and 3-hourly time series were derived by the disaggregation of the monthly MODIS data, mean diurnal cycles were constructed from Geostationary Operational Environmental Satellite (GOES) Wildfire Automated Biomass Burning Algorithm (WF\_ABBA) active fire observations (Mu et al., 2010). GFEDv4 vegetation fire emissions from 1995 through 2016 are available online (GFED, 2017).

A system which allows for near real time use of emission from vegetation fires is described by Kaiser et al. (2012). The Global Fire Assimilation System (GFAS) calculates biomass burning emissions by assimilating Fire Radiative Power (FRP) observations from the MODIS instruments onboard the Terra and Aqua satellites. GFAS makes use of the quantitative information on the combustion rate that is contained in the FRP observations from the satellites, and it detects fires in real time at high spatial and temporal resolution. Daily fire emissions for 40 gas-phase and aerosol trace species are compiled on a global 0.5 × 0.5 grid from 2003 to the present. For an improvement of the assimilation system see Di Giuseppe et al. (2017).

An additional challenge related to the creation of model ready fire emissions is the determination of the effective injection height. Depending on the heat content of the fire plume relative to the ambient air masses and the local weather conditions fire emissions could enter the model in the ground layer or far above the planetary boundary layer. Freitas et al (2006; 2001) proposed a mechanism to determine the effective injection height of fire emissions using a plume buoyancy approach. They calculated the plume rise with a 1-dimensional cloud resolving model for every model grid cell where a fire occurs.

## Volcanoes

Volcanoes are important natural sources of ash particles and a number of reactive gases including sulfur dioxide and halogens. They have the potential for huge impacts on the local and regional environment but this depends very much on the location of the volcano and the weather conditions during its eruption. Volcanic emissions are highly intermittent and almost impossible to forecast. Similar to dust emissions, the size spectrum of the emitted particles is subject to large variations, depending on details during the eruption process (Sparks et al., 1997). The amount of transportable particles is typically just a few percent of the total emitted mass (see e.g. Langmann et al., 2012 about the eruption of the Eyjafjallajökull in 2010) and it is hard to determine during the eruption how big this fraction really is and its size distribution. Observations combined with model simulations are needed for a reliable estimate of the ash concentrations and their regional distribution (Stohl et al., 2011, Matthias et al., 2012). In the future, online monitoring networks such as the Network for Observation of Volcanic and Atmospheric Change (NOVAC) (Galle et al., 2010) will help representing volcanic emissions in emission data sets.

Because volcanic emissions consider only one large source of aerosol particles that do not interact with other pollutants in the atmosphere, they are usually treated with Lagrangian models (e.g. Stohl et al., 2011, Mastin et al., 2009) and emissions from other sources are not considered. Therefore they are in most cases not included in emission data sets used for air quality simulations. In some regions, volcanoes are permanent sources of SO<sub>2</sub> and it might therefore be recommendable to consider these emissions. Neely and Schmidt (2016) compiled

an emission data base for SO<sub>2</sub> emissions from volcanoes for the time period from 1850 until today. However, it focuses on explosive emissions during which SO<sub>2</sub> enters the stratosphere. There, it might remain for a long time and contribute to the formation of small sulfuric acid droplets.

## Lightning

Von Liebig proposed already in 1827 that lightning might be a natural influence factor for the production of nitrogen oxides. The ionic lightning channel heats up to several thousand degrees Celsius. At these temperatures, molecular nitrogen and oxygen from ambient air react to form nitrogen oxides.

The range of emission estimates on the global scale varies from 1 Tg to 20 Tg (Schumann and Huntrieser 2007, Zhang et al., 2003, Lee et al., 1997). This is mainly because the emission estimate per flash varies. Following IPCC (2001) lightning NO<sub>x</sub> contributes about 5 Tg N to the global annual emissions, which is about 10%. Because the highest lightning density occurs in the Inter Tropical Convergence (ITC) zone, most emissions are located around the equator.

Emissions are usually calculated based on the flash counts. Three approaches how they can be derived are common: A parameterizations based on meteorological model data that predicts thunderstorms and consequently lightning activity. Alternatively, lightning detection network measurements like the National Lightning Detection Network (NLDN) in the US and optical satellite images like the Lightning Imaging Sensor (LIS) and the Optical Transient Detector (OTD) satellite instruments can be used to retrieve the number of flashes at a certain location. Usually the flash counts are converted into flash densities and then multiplied with an emission factor to derive the NO emissions. These emission factors are derived from field experiments and satellite data (see Ott et al., 2010 and Allen et al., 2012) and can be constant or based on flash data like flash energy and length. Such data is used to constrain models that calculate flash densities based on cloud top height, e.g. Murray et al. (2012) for GEOS-Chem and Wang et al. (2013) for CMAQ.

Some parameterizations distinguish between the in-cloud flashes and the cloud-to-ground flashes. The emissions are distributed according to the vertical lightning energy distribution.

Assuming a fixed ratio between the in-cloud and cloud-to-ground flashes (e.g. Fehr et al., 2004), the NO emissions are uniformly distributed between the ground and the cloud top.

Depending on which of the methods described above is used, lightning NO<sub>x</sub> can be calculated in a preprocessor or inline in the chemical transport model itself.

## **Summary and Outlook**

Detailed emissions data are critical for accurately predicting concentrations of atmospheric air pollutants through chemistry transport modeling. For three dimensional chemistry transport model systems, emission amounts as well as their spatial and temporal distribution need to be provided for a number of anthropogenically and naturally emitted pollutants. Over recent years greater emphasis has been devoted to this fact, partly due to the better source specific availability of emission data. Nowadays a number of emission inventories exist for the global and the regional scale and for time periods of several years up to decades. The inventories are constructed with different methods that can be roughly divided into top-down and bottom-up approaches. Bottom-up approaches are widely used for natural emissions. Most of the anthropogenic emission inventories are also bottom-up because they calculate emissions from activities and emission factors. However, the activities are often derived from national or global statistics (e.g. on fuel use) and spatially distributed using proxy data, which is a top-down approach. One main advantage of top-down approaches is that similar methods to distribute the emissions in space can be applied all over the globe. This helps avoiding unrealistic gradients, e.g. between countries that use different methods to calculate emissions with bottom-up methods. Satellite data products are frequently used to construct emission inventories in a top-down approach. However, the number of pollutants that can be treated this way was limited until now. Bottom-up approaches are typically more accurate in the temporal and spatial distribution of the emissions because they take detailed information about the emission sources into account. Though, if the number of individual sources gets very large, it can be extremely laborious, if not impossible for practical reasons, to account for every single emission source that is highly variable in time and space. Large point sources are typically considered in a bottom-up calculation. Shipping emissions are another, more recent example. Their emissions can be calculated based on ship position data and technical specifications that are available for almost all bigger ships in the world.

In order to better account for the temporal and spatial variation of emissions, a number of methods have been developed that use detailed source specific information to distribute e.g. annual national emission totals. This can be seen as a hybrid approach where temporal and spatial distribution functions are calculated as in a bottom-up emission calculation and these functions are subsequently used to distribute national emission totals of a certain sector in time and space. This can include spatially varying temporal emission profiles. Such a method has e.g. been used for NH<sub>3</sub> emissions from agriculture and for emissions from residential heating, both of which at least partly depend on meteorological parameters like ambient temperature or wind speed. Such an approach could possibly be applied for other emission sectors as well, although the data needed to model it properly may differ between sources. Emissions from power plants for example depend on weather. First, because the energy demand for heating and cooling depends on it and second, because the amount of renewable energy in the electricity grid depends on it. The latter may lead to lower emissions from traditional coal or gas fired power plants.

The amount of detail in the emission data that is necessary to improve the CTM calculations depends on the area and the time period under investigation. The smaller the area and the shorter the time period, the more detailed information is necessary. Concerning the spatial resolution, it is evident that for a small area more details about, for example, traffic on specific streets, is necessary. For industrial plants in the area one has to know if they were really in use in the modelled time period. Comparisons between emission inventories for China (Li et al., 2017, Saikawa et al., 2017) showed that most likely regional inventories are more accurate than global ones for the same area. However, in some regions of the world regional emission inventories simply do not exist and global inventories are the only source of information.

Improving the temporal and spatial allocation of emissions in the future could involve large data sets that exist but have not been used for emission modeling up to now. Emissions from street traffic could be improved if the real variability including traffic jams and the composition of the fleet could be considered. Such data is collected by authorities, companies like Google, or within road toll systems in several countries. In the future, on-board units might even measure actual vehicle emissions in order to prove their compliance to existing regulations. The same will apply to ships. This information could ideally be used for emission data sets.

Emissions from agriculture can be made more accurate if application dates and amount of fertilizers would be reported. Satellite data products could help here by monitoring changes in surface properties that are caused by agricultural activities (e.g. high spatial resolution images from Sentinel 2, both in the visible and IR wavelength range, or Landsat land cover products). At the same time, off-road vehicle emissions including agricultural machinery could be estimated. Emissions from industry and power plants could also be reported in more detail than today and subsequently be used in emission models. Exhaust gas properties like temperature, exit velocity and concentrations of pollutants are often measured but the data is not accessible in high temporal resolution or not reported if it falls below a certain threshold. The same holds for stack heights and diameters, not only from industrial plants but also from large ships.

Satellite data is already used to provide daily information on fire activity (GFAS, 2017) all over the globe. In addition, it could be of great benefit, e.g. to better determine biogenic emissions. Spatial distributions of plant type, plant growth and leaf area (e.g. from MODIS), exact dates for blooming periods, harvesting, and leaf stripping, might be taken into account. The same for holds for land use changes caused by human activities like building new settlements, large dams or deforestation, which all change emission sources, even if not on very high time resolution.

Biogenic and most types of natural emissions, e.g. dust, sea spray and soil emissions depend on meteorological conditions, which means that they are highly variable in space and time. While their temporal variation can be calculated using the best available meteorological data, the total amount of emitted gas or particles are still connected with large uncertainties. This holds in particular for the size distribution of dust or sea salt particles and here for the fraction of small particles that can be transported over large distances. Sea spray may contain significant amounts of organic aerosol in times of high biological productivity in the oceans. This is seldom or only rudimentary considered in emission models.

Although there is still much space for the improvement of emission data that is used in 3D chemistry transport model systems, it should also be born in mind that more details being considered in the emission models does not necessarily improve the data. When processes are taken into account that could have large effects but are not well known, this could introduce high uncertainties and in the end worsen the emission data.

In summary, emission data is arguably the most important input for chemistry transport model systems and it needs to be prepared and checked carefully. It needs to be well adapted to the purpose of the model exercise and to the scientific question being asked. The smaller the model domain and the shorter the modelled time period, the more detail is necessary. A number of improvements have been achieved in recent years concerning more sophisticated temporal and spatial allocation of emissions. With the upcoming availability of huge data sets gathered through the use of information technologies in almost all areas of daily life, emission inventories can be further improved.

## Glossary

**Activity data:** Quantity describing the activity relevant for the emissions of a certain source, e.g. amount of fuel used, distance travelled, or amount of energy converted.

**Activity profile:** Temporal development of the activity of a certain emission source or a group of sources (e.g. fuel use per hour for one truck or all cars on a certain highway).

**Anthropogenic emissions:** emissions resulting from anthropogenic activities, such as vehicle and power plant emissions.

**Area source:** spatially aggregated emission sources that are uniformly distributed in a model grid cell. Emission sources summarized into area sources are commonly high in number and have low emissions per individual source.

**Biogenic emissions:** Emissions from vegetation.

**Bottom-up approach:** Method to calculate emission totals for a larger region or an emission sector by summing up all emissions from singular sources.

**Bulk emissions:** spatially and/or temporally aggregated emissions.

**Classical air pollutant:** SO<sub>2</sub>, NO<sub>x</sub>, CO, NMVOC, NH<sub>3</sub> and PM.

**Dust:** particulate matter emitted from land surface mainly by wind.



**Emission:** Flux of a substance X entering the atmosphere.

**Emission inventory:** Collection of emission amounts given as a sum over a longer period (typically 1 year) together with 2D spatial information (either on a grid or for administrative units like countries, regions and cities).

**Emission data set:** Emissions that have been processed in order to be directly used in 3D chemistry transport models. They are given as 4 D data including vertical information and temporal information on hourly level.

**Emission factor:** Source specific quantity describing the emissions of a substance X per activity unit.

**Emission sector:** Collection of emission sources characterized by similar activities (e.g. heating of houses and flats) or having similar purposes (e.g. electricity production, transport).

**Natural emissions:** Emissions from non-anthropogenic sources, such as dust and sea spray.

**Photochemical mechanism:** Set of reactions and reaction constants, which describe photochemical reactions of atmospheric compounds (individual chemicals or groups of chemicals)

**Planetary boundary layer (PBL):** Also known as atmospheric boundary layer; lower part of the atmosphere that is affected by the planetary surface. Air masses in the PBL have contact with the surface every 1-2 hours.

**PM<sub>2.5</sub>:** Particulate matter with a diameter  $\leq 2.5 \mu\text{m}$ . Concentrations are typically given as mass per volume.

**PM<sub>10</sub>:** Particulate matter with a diameter  $\leq 10 \mu\text{m}$ . Concentrations are typically given as mass per volume.

**Point source:** Individual emission source, typically with high emissions, such as power plants.

**Residential heating:** Heating of residential/apartment buildings.

**Sea spray:** Water droplets emitted from the ocean surface containing sea salt or marine organic compounds.

**Top-down approach:** Method to distribute emission totals for a larger region (e.g. a country) or for entire emission sectors (e.g. transport) into smaller units (e.g. model grid cells) by using appropriate proxy data (e.g. street maps).

**Volatile organic compounds (VOCs):** Group of organic compounds with a high vapor pressure at room temperature: majority of the molecules of VOCs is found in the gas phase. Usually, methane is not included in this group of substances. It is then named Non-Methane VOCs (NMVOCs).

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