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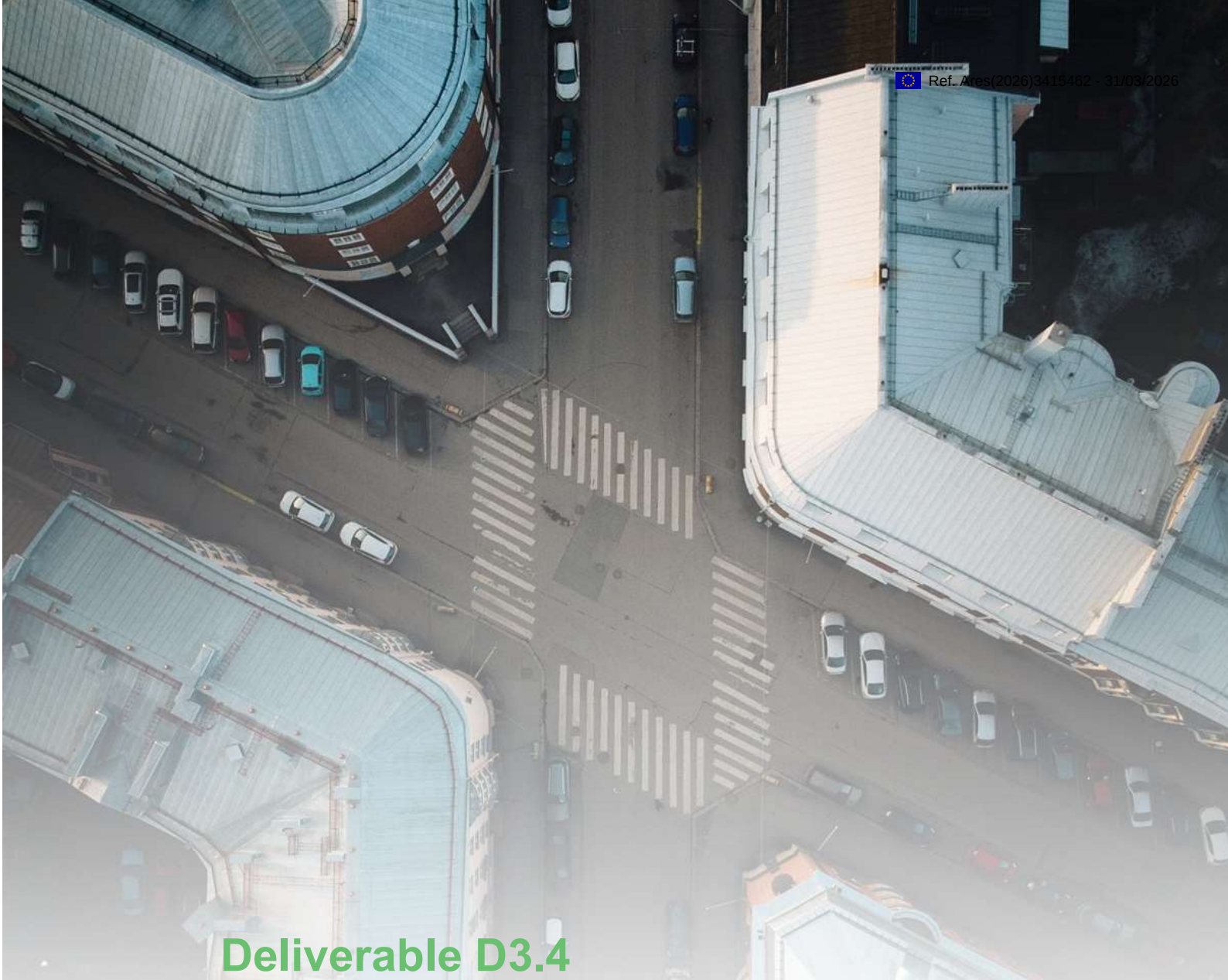

Deliverable D3.4

Digital equivalent total particles emissions index (ePMI) module software

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PAREMPI
Particle emission prevention and impact: from real-world emissions of traffic to secondary PM of urban air

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PAREMPI Particle emission prevention and impact: from real-world emissions of traffic to secondary PM of urban air

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List of abbreviations

BC	Black Carbon
BVOC	Biogenic Volatile Organic Compounds
CH_4	Methane
CO	Carbon monoxide
CTM	Chemistry Transport Model
EC-Earth	European Community Earth System Model
ePMI	Digital equivalent total particles emissions index
ESM	Earth System Model
MAE	Mean Absolute Error
MSA	Methanesulfonic acid
NEE	Non-exhaust emissions
NH_3	Ammonia
NH_4NO_3	Ammonium nitrate
NH_4^+	Ammonium
NMVOCs	Non-Methane Volatile Organic Compounds
NO_3^-	Nitrate
NO_x	Nitrogen oxides (NO and NO_2)
OC	Organic Carbon
O_3	Ozone
PM	Particulate Matter
$PM_{2.5}$	Particulate Matter smaller than 2.5 µm in diameter
PM_{10}	Particulate Matter smaller than 10 µm in diameter
PN	Particle Number
POM	Primary Organic Matter
RMSE	Root Mean Square Error
SOA	Secondary Organic Aerosols
SO_2	Sulfur dioxide
SO_4	Sulfate
UFP	Ultrafine Particles, i.e. particles smaller than 100 nm in diameter
VOC	Volatile Organic Compounds

1. Executive Summary

This technical note describes the digital equivalent total particles emissions index (ePMI) module software and its implementation in the global chemistry transport model TM5. ePMI predicts the primary and secondary particulate matter (PM), including $PM_{2.5}$ and PM_{10}, and ultrafine particle (UFP) formation on a limited number of emissions and meteorological input variables used by regional and global scale chemistry transport models (CTM). A standalone, easy to use, ePMI module version has been developed and integrated into a 1-dimensional atmospheric column model version of TM5. This so-called TM5-ePMI software can be used to estimate how anthropogenic emissions of aerosol particles and gases, from various sectors, influences the primary and secondary aerosol particle concentrations (number, surface area and mass) in a region (location) over the course of a day. It can also be used as a platform for initial testing and integration of new aerosol processes and parameterizations for regional and global scale CTMs or Earth System Models (ESMs), such as the European Community Earth System Model (EC-Earth).

2. Introduction

Due to the complex chemistry of the atmosphere, the effect of reducing emissions is not always straight forward. Therefore, it is not always clear which sectors, or species should be targeted by policy makers looking to improve air quality in their local region. Here, we present an easy-to-use software that can be used to quickly assess how changing emissions affect particle number and mass concentration.

The current beta-version needs to be compiled by the user, which requires some technical knowledge; however, in the future we aim to provide an executable (static binary) that can be run on any computer.

3. The ePMI software

The ePMI is intended to be used as a computationally efficient easy to use standalone software for assessment of anthropogenic particulate and gaseous emissions "total" impact on the particulate matter (PM) levels in a region of interest, accounting both for primary aerosol emissions, secondary aerosol formation, deposition and vertical mixing within the atmospheric boundary layer. ePMI is also intended to be used as an integrated aerosol module in global and regionals scale CTMs and ESMs. In the Horizon Europe project PAREMPI we have implemented and used ePMI in the global chemistry transport model TM5 (see https://github.com/AugustThomasson/traffics-affect-on-AQ-Europe for some preliminary results about traffics effect on $PM_{2.5}$ and particle number (PN) concentrations in Europe). The ePMI module includes simplified parameterizations of biogenic and anthropogenic SOA and UFP formation rates based on a limited number of input variables.

The principal idea behind ePMI is that air quality at a location is affected both by local and upwind emissions. We can greatly reduce the computational cost to assess how the local emissions, e.g. from road traffic, affect the total aerosol mass, surface area and number concentrations at a location if we assume that the local emissions only influence the local air quality at reasonable short timescales after being emitted and neglect horizontal transport (advection) within this time. The assumption of negligible influence from horizontal transport is of course not correct but could be justified on relatively short time scales (within less than one day) and for conditions with low horizontal wind speed. Under this assumption, we can initialize the model with concentrations of trace species representative of the background concentrations and run the model only for a short period while letting local emissions and atmospheric chemistry change the concentrations. The aim of the standalone ePMI software is not to be able to represent any specific real values, but rather to quickly assess the local equivalent total particle emissions and concentrations of (primary and secondary aerosols) originating from an anthropogenic emission sector in a region of interest. A secondary aim is to provide a platform to easily test model developments/parameterizations that later can be implemented in regional or global CTMs and ESMs.

The TM5-ePMI software is a column version of the global chemistry transport model TM5-MP (Bergman et al., 2022; Williams et al., 2017). The software inherits the following components from TM5-MP:

1. Gas-phase chemistry (modified version of Carbon Bond 5 (Williams et al., 2017, 2013; Yarwood et al., 2005))
2. Photolysis with aerosol feedback (Aan de Brugh et al., 2011; Williams et al., 2012)
3. The M7 aerosol dynamics model (Vignati et al., 2004), which represents aerosols dynamics (nucleation, coagulation, growth, and aging) in 7 log-normal modes. This version also includes Secondary Organic Aerosol (SOA) (Bergman et al., 2022) in addition to sulfate (SO_4), Black Carbon (BC), Primary Organic Matter (POM), dust and sea salt.
4. The EQuilibrium Simplified Aerosol Model EQSAM (Metzger et al., 2002; Williams et al., 2017) for gas/particle partitioning of inorganics. The result is a bulk representation of nitrate (NO_3^-), ammonium (NH_4^+) and methanesulfonic acid (MSA).
5. Vertical mixing in the boundary layer (Holtslag and Boville, 1993)
6. Dry deposition and sedimentation scheme (Aan de Brugh et al., 2011)

7. Symmetrical operation splitting (but without any advection): within each time step the model does half a time step that apply vertical mixing (V), sources and sinks (S) and the chemistry (C). In the next half timestep, the order is reversed: C - S - V.
8. Code structure, so that changes to the above-mentioned components could be easily tested in ePMI before being implemented in the full TM5-MP model

In addition, we have implemented the following components which are not part of TM5-MP

1. A novel SOA parameterization for aromatics
2. An air ion mediated ammonia – sulfuric acid new particle formation lookup table (Svenhag et al., 2024)
3. Sub-10 nm primary particle emissions from urban road traffic (Lintusaari et al., 2023)
4. The inclusion of road traffic non-exhaust emissions (NEE) into the dust tracers

The model is initialized with concentrations of each tracer representative of the background. These can either be user specified, but for simplicity we also provide a file that contains seasonal averages of each tracer in each vertical level on a 1°x1° grid in Europe from a full TM5-MP run of 2018. Due to the resolution of this data, it is representative of a rural background (and urban background for large enough cities to be resolved).

The user also specifies emissions of selected species from different sectors. Currently, only anthropogenic emissions of SO_2, NO_x, Non-Methane Volatile Organic Compounds (NMVOCs), NH_3, BC and OC are implemented for the following sectors: Road transport, Industry, Energy, Domestic Combustion, Shipping and Others. In addition, fine and coarse NEE from road transport can be specified. Each value should represent a daily average in $kg/m^2/s$. For some sectors, a temporal profile is applied to mimic changes to emissions throughout the day (Guevara et al., 2021). The emissions are added to different vertical levels in the model depending on the sector as in TM5-MP.

We assume very idealized meteorology, fixed throughout the simulation, and everything can be specified by the user. However, we do provide seasonal averages on the same 1°x1° European 2018 grid for vertical profiles of temperature and specific humidity, surface pressure and orography as a starting point. The only temporally dynamic meteorological variables are surface solar radiation and planetary boundary layer height which follow idealized parametrization based on solar zenith angle and user specified max/min values (see Figure 1 for examples). To modify the dry deposition and sedimentation, the user can specify 10 meter windspeed, surface roughness, leaf-area index. The vertical mixing in the boundary layer is determined by atmospheric stability, mostly modified by the surface roughness and surface latent and sensible heat flux. These can also be changed, but we recommend using one of the three predefined stability cases: unstable, neutral, stable. If the user does not specify any values, the model will use default values representing a mid-latitude urban environment.

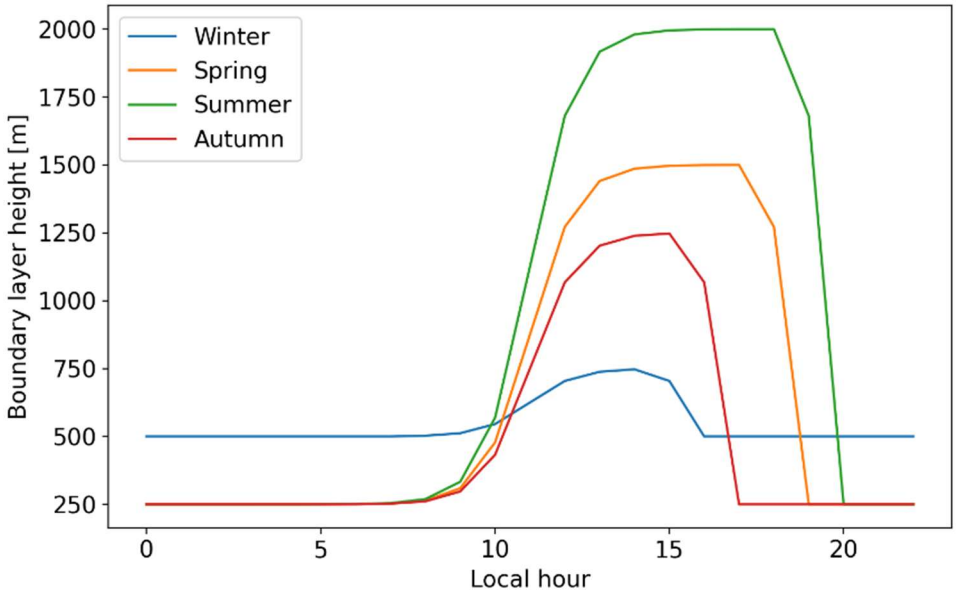

Figure 1: Example of idealized boundary layer heights based on different min/max heights and solar zenith angle per seasons

With this approach there are some limitations:

1. The standalone TM5-ePMI column model does not model the advection of tracers. Instead, the model is initialized with values representative of the background concentrations and runs as a stationary column with local emissions. It is assumed that the local emissions stay within the local region throughout the simulation. Therefore, we recommend only running the model for a short period (24 hours or less), otherwise tracers may accumulate unrealistically.
2. The idealized meteorology means that the model cannot reproduce any specific observations/measurements exactly. But this is not the aim of the software.
3. The model currently only considers clear sky meteorology (no cloud cover to modulate photolysis) and does not model wet deposition.
4. Currently, TM5-ePMI is designed to investigate the effect of urban emissions on local urban air quality. Therefore, it only considers anthropogenic emissions of selected short-lived secondary aerosol precursors and gases, and primary particles. The background concentrations do contain natural and wildfire emissions, but since there is no advection, some tracers which interact with local emissions might be depleted faster than expected, such as O_3 and BVOCs. There is an option to use fixed O_3, CO, CH_4 and BVOC vertical concentration profiles.

The results are presented as two tables in the standard output (in the terminal, or in a log file if specified in the settings.rc file) of the model. Both tables consist of a column showing initial (background) values, one column with the average and max contributions from the TM5-ePMI

simulation, and one with the total average and max (background + TM5-ePMI). The first table contains particle number concentrations (all UFPs), i.e. particles smaller than 100 nm in diameter) and the second table contains particle mass concentrations of $PM_{2.5}$ and its components. An example of the model output is presented in Figure 2.

```
ePMI/TM5_Model_Done: start
 ------------------------------------------------------------------------
     RESULTING PARTICLE NUMBER (PN)
 ------------------------------------------------------------------------
                          |     From ePMI     |       Total       |
 Size (cm-3)     |  Init  |   Mean  |   Max   |   Mean  |   Max   |
 ------------------------------------------------------------------------
 All             | 2767.7 |  1923.4 |  4236.7 |  4691.1 |  7004.4 |
 UFP (d < 100nm) | 1839.4 |   989.2 |  2814.4 |  2828.7 |  4653.8 |
 ------------------------------------------------------------------------
 ------------------------------------------------------------------------
     RESULTING PARTICLE MASS (PM)
 ------------------------------------------------------------------------
 Size & specie   |  Init  |     From ePMI     |       Total       |
    ug/m3        |        |  Mean   |   Max   |  Mean   |   Max   |
 ------------------------------------------------------------------------
 PM2.5           |  4.292 |  3.819  |  6.902  |  8.110  | 11.194  |
    Primary      |  1.020 |  1.247  |  2.345  |  2.267  |  1.273  |
       BC        |  0.134 |  0.626  |  1.139  |  0.759  |  1.273  |
       POA       |  0.699 |  0.390  |  0.805  |  1.089  |  1.504  |
       DUST/NEE  |  0.188 |  0.231  |  0.401  |  0.419  |  0.589  |
    Secondary    |  2.361 |  2.825  |  5.746  |  5.186  |  1.273  |
       SOA       |  0.593 |  0.647  |  1.146  |  1.240  |  1.740  |
       NH4       |  0.488 |  0.627  |  1.187  |  1.115  |  1.676  |
       NO3       |  0.000 |  0.022  |  0.443  |  0.022  |  0.443  |
       SO4       |  1.279 |  1.529  |  2.969  |  2.808  |  4.248  |
 ------------------------------------------------------------------------
ePMI/TM5_Model_Done: end
```

Figure 2: Example of the standard output from the model

In addition to the standard output, the model can write full model time series of the entire column of most trace gases and aerosol components and some additional variables and meteorology into a .nc file. Use the setting “output.general : T” for this.

3.1 Model validation

To validate that the model produces realistic results we here present two sets of validation simulations. In 3.1.1, the model is validated against urban traffic measurements of $PM_{2.5}$ and NO_x in Sweden (provided by SMHI: https://datavardluft.smhi.se/portal/). In 3.1.2, the model is compared to Urban BC measurements from the European RI-Urbans infrastructure (Savadkoohi et al., 2023) (accessed at https://ebas-data.nilu.no/). In both cases, the average measurements for the seasons in 2018 are compared to the background values and the background values + ePMI. For each site, TM5-ePMI is run once per season initialized with the background concentrations at the given location. For emissions, seasonal averages from CAMS-GLOB-ANT v5.3 (Soulie et al., 2024) (except from industry which was found to have too large values in some cities) and HTAPv3 (Crippa et al., 2023) for fine and coarse non-exhaust emissions, both at 0.1°x0.1°, is used to represent the local emissions at the site. For the vertical pollution dispersion, the boundary layer height is represented according to Figure 1 and vertical mixing is turned on, since unstable or neutral conditions are more common than stable.

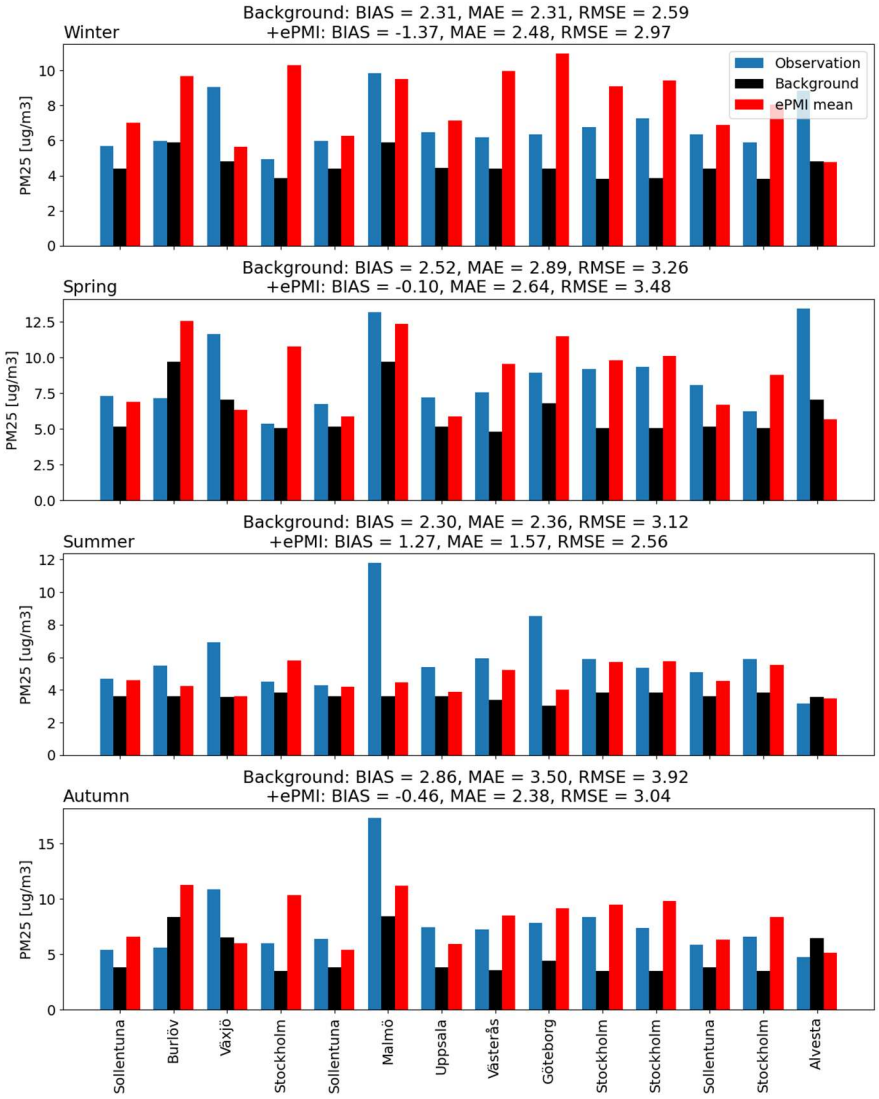

Figure 3: Observed seasonal averages of $PM_{2.5}$ (blue), background (black) and TM5-ePMI (red) for several Swedish urban roadside measurements. The rows show different seasons. Bias, mean absolute error and root mean squared error is annotated.

3.1.1 PM2.5 at urban traffic sites in Sweden

Figure 3 presents the comparison of observed seasonal averaged $PM_{2.5}$ measurements along several urbans roads in Swedish cites and corresponding model results. We see that the results from TM5-ePMI (red) in most cases recover some of the difference between background (black) and observations (blue), indicating that ePMI is producing realistic results. The bias, mean absolute error (MAE) and root mean squared error (RMSE) is generally improved (lower) when adding the ePMI results to the background. We see that ePMI overestimates mostly in winter and to a lesser extent in spring and autumn. During the winter the fraction of semi-volatile secondary inorganics (NH_4NO_3) in $PM_{2.5}$ is higher due to the lower temperatures and high humidity. Semi-volatile $PM_{2.5}$ components might not always be well represented by ambient observations as they can evaporate when heating and drying the samples. It is also possible that the idealised boundary layer, temperatures or clear sky assumptions are not truly representative of the seasonal average in winter especially. As mentioned

previously, the aim of TM5-ePMI is not to accurately reproduce specific observations/conditions, but to understand the general burden of traffic and other anthropogenic emissions, and to separate its influence on urban particulate loads.

3.1.2 BC at urban sites in Europe

Here we show that TM5-ePMI can recover the difference between background and observed seasonal average BC concentrations at European urban sites for 2018, indicating that ePMI is producing realistic values despite the idealized assumptions. Figure 4 shows the seasonal observed averages (blue), background (black) and TM5-ePMI (red) results for several sites. For some sites, TM5-ePMI still underestimates observations (s.a. ES0020U, NL0017U) which might be due to the idealised vertical mixing, but overall, we see an improved bias, MAE and RMSE indicating that ePMI produces realistic values.

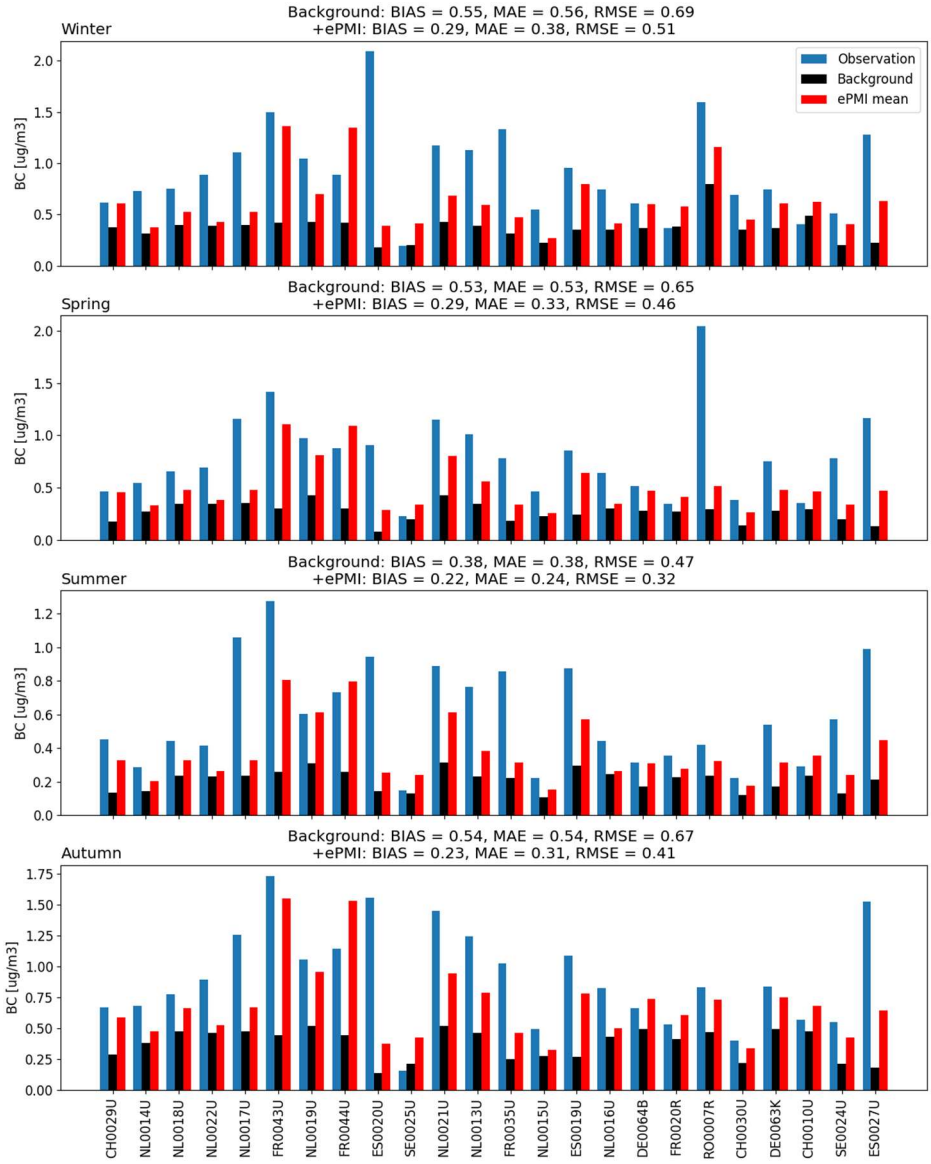

Figure 4: Observed seasonal averages black carbon (blue), background (black) and TM5-ePMI (red) for urban measurement sites in Europe. The rows show different seasons. Bias, mean absolute error and root mean squared error is annotated

3.2 Equivalent total particle emission examples

Below follows a few examples of the intended use of TM5-ePMI: finding the equivalent total particle number and mass concentrations for example emissions. The presented results in section 3.2.1 are for runs at Malmö, Sweden, assuming unstable boundary layer conditions and without fixed ozone concentrations in TM5-ePMI.

3.2.1 Equivalent total particles from traffic

We run TM5-ePMI with emissions from all anthropogenic emission sectors and without road transport emissions, under the assumptions that emissions from all sectors interact instantly. Both runs use boundary layer heights as in Figure 1 and full vertical mixing. The equivalent total particle mass and number concentrations from traffic is inferred to be the average difference between the runs with and without road transport emissions.

The modelled equivalent total particle mass concentration contributions are similar during winter and autumn with road traffic contributing to 2.2-2.7 µg/m^3 $PM_{2.5}$ (Figure 5). Around half of this is due to secondary aerosols, primarily inorganics (nitrate and ammonium) while the other half is due to primary particles, of which 80% are from non-exhaust emissions. During these seasons, equivalent total particle number concentrations due to traffic are around 1000-1200 cm^{-3}, 60-70% of which are ultra fine.

In spring, the secondary inorganics dominate total traffic $PM_{2.5}$ due to reduced primary non-exhaust emissions from resuspended road dust. Summer equivalent total particle mass from road traffic is near zero in the model. The primary particles still contribute to increased $PM_{2.5}$ from traffic, but with lower non-exhaust particles. There is a clear negative contribution from the secondary particles in TM5-ePMI. This means that the model suggests that removing traffic in summer would slightly increase the secondary $PM_{2.5}$ mass, highlighting the complex non-linear chemistry. In the example this is primarily a result of reduced OH concentrations and slower oxidation of SO_2 to sulfate. The particle number concentration contribution is still positive, with around 400 cm^{-3} UFP from road traffic.

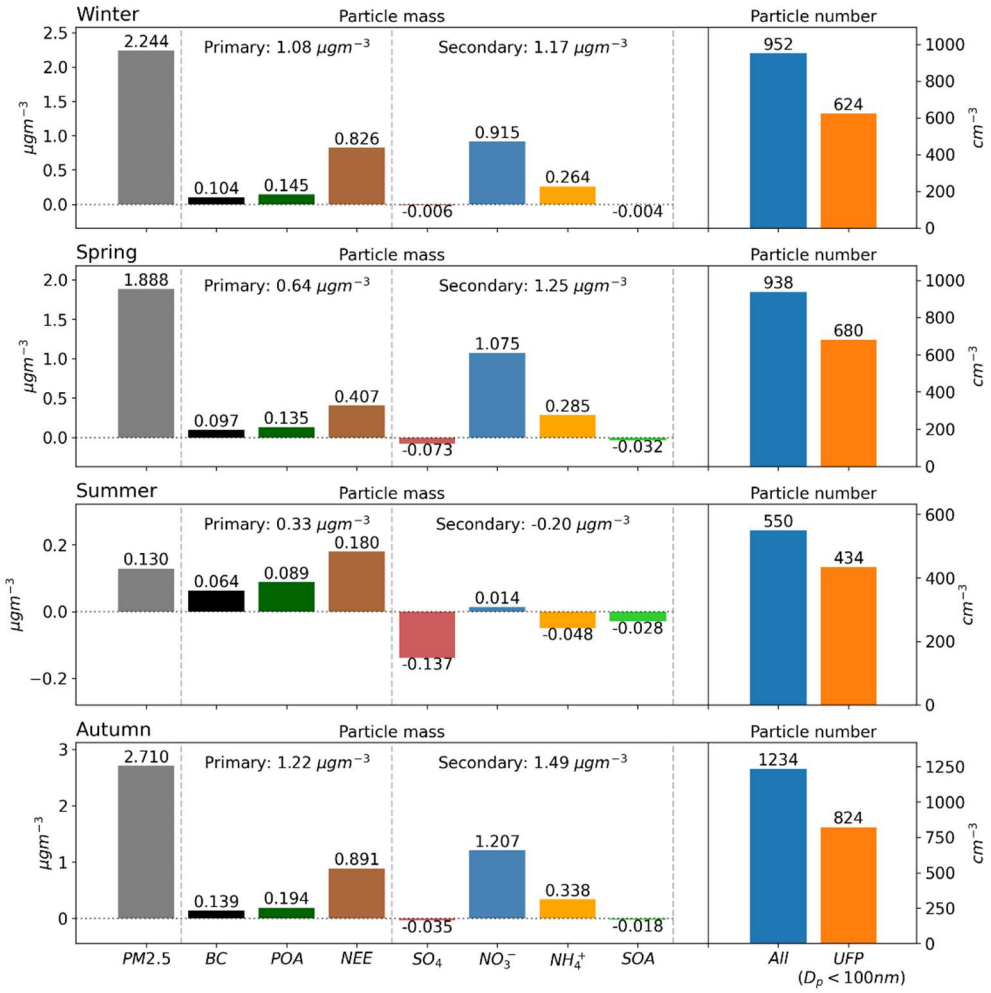

Figure 5: The equivalent total particle mass (left) and number (right) from traffic sector in Malmö Sweden. The rows show the seasons. The particle mass is represented as $PM_{2.5}$ and for select components, particle number for all and ultra fine sizes are showed.

3.2.2 Equivalent total particles from traffic under different conditions

Now, we investigate how different conditions (model assumptions) affect the traffic emissions equivalent total particle mass and number in summer by changing one condition at the time. Table 1 presents the traffic primary and secondary $PM_{2.5}$ and UFP. Using fixed O_3 concentration profiles have very little effect on the traffic contribution, only a slight increase in the secondary nitrate mass. If we change the stability to neutral or even stable boundary layer conditions, both the traffic primary particle mass, UFP concentrations and total number concentrations increase predictably. Notably, the neutral condition results in a positive contribution from traffic to secondary mass, due to much higher nitrate particle mass formation under these conditions. In stable conditions, traffic again reduce the secondary mass.

Turning of nucleation scheme from (Svenhag et al., 2024) to the standard method in TM5-MP does not affect the particle mass, but UFP number is slightly increased. In contrast, removing the primary UFP

emissions from traffic predictably drastically reduces the UFP concentrations. Finally, using a longer model timestep results in slightly less traffic primary mass and a smaller reduction in secondary mass, while UFP are reduced compared to the base case. With a shorter timestep the traffic contribution to the particle mass and number concentrations are similar to the base case.

Table 1: Equivalent total particle mass (presented as primary and secondary $PM_{2.5}$) and UFP number concentrations from traffic sector for several sensitivity tests. The values represent the average difference between TM5-ePMI with and without traffic sector.

Experiment	PM2.5 (ug/m3) Primary	Secondary	PN (cm-3) UFP
Base case	0.36	-0.20	434
Fixed O3 chem	0.35	-0.18	423
Neutral stability	0.52	0.97	609
Stable	1.73	-1.2	1485
No ammonia-sulfuric acid nucleation	0.35	-0.19	534
No SOA from aromatics*	0.35	-0.16	418
No UFP from CO2	0.35	-0.19	61
1-hour timestep	0.33	-0.16	354
15-min timestep	0.37	-0.2	486
5-min timestep	0.39	-0.21	413

*Total SOA (from all sources) decreased by 0.7 ug/m3 without SOA from aromatics, but the traffic SOA contribution is nearly unchanged

3.3 Access to code repository

The code repository can be accessed at https://zenodo.org/records/19233774. The repository contains following directories and contents:

- /bin: the compiled binary. Since the current version is not statically linked, it is unlikely that it is possible to run the binary without compiling it on the user machine.
- /src: the Fortran source code, which is a mix of heavily modified TM5-MP source code and custom ePMI code. It is not possible to compile TM5-MP with this code alone.
- /input: contains input files and data for the photolysis with aerosol feedback (Aan de Brugh et al., 2011; Williams et al., 2012), ammonia – sulfuric acid nucleation lookup table (Svenhag et al., 2024) and the two input files with seasonal background concentrations and meteorology on a 1°x1° grid in Europe.
- /preprocess: a python script with a helper function to get emissions based on an emission inventory, and an example use case with CAMS-GLOB-ANT v5.3 (Soulie et al., 2024) and HTAPv3 (Crippa et al., 2023). Needs to be modified by the user.
- /postprocess: two example python scripts that plot various parts of the .nc output. Needs to be modified by the user.
- README.md: contains a shorter description of the model, a user guide for compiling and running the model with examples and some best practices.
- test.rc: an example of model settings
- build_epmi.sh: a short bash script to automate the compilation of the model

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