1	The HTAP_v3 emission mosaic: merging regional and global monthly emissions (2000-	Formatted: Font: 12 pt, Font color: Auto
2	2018) to support air quality modelling and policies	Formatted: Font: 12 pt
3		Formatted: Font: 12 pt
4 5 6	Monica Crippa <sup>1</sup> , Diego Guizzardi <sup>1</sup> , Tim Butler <sup>2</sup> , Terry Keating <sup>3</sup> , Rosa Wu <sup>16</sup> , Jacek Kaminski <sup>4</sup> , Jeroen Kuenen <sup>5</sup> , Junichi Kurokawa <sup>6</sup> , Satoru Chatani <sup>15</sup> , Tazuko Morikawa <sup>7</sup> , George Pouliot <sup>17</sup> , Jacinthe Racine <sup>8</sup> , Michael D. Moran <sup>16</sup> , Zbigniew Klimont <sup>9</sup> , Patrick M. Manseau <sup>8</sup> , Rabab	
7	Mashayekhi <sup>8</sup> , Barron H. Henderson <sup>17</sup> , Steven J. Smith <sup>10</sup> , Harrison Suchyta <sup>10</sup> , Marilena	Formatted: Font: Times New Roman
8 9	Muntean <sup>1</sup> , Efisio Solazzo <sup>11</sup> , Manjola Banja <sup>1</sup> , Edwin Schaaf <sup>1</sup> , Federico Pagani <sup>12</sup> , Jung-Hun Woo <sup>13,14</sup> , Jinseok Kim <sup>14</sup> , Fabio Monforti-Ferrario <sup>1</sup> , Enrico Pisoni <sup>1</sup> , Junhua Zhang <sup>16</sup> , David	
10	Niemi <sup>8</sup> , Mourad Sassi <sup>8</sup> , Tabish Ansari <sup>2</sup> , Kristen Foley <sup>17</sup>	Formatted: Font color: Auto
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12	<sup>1</sup> European Commission, Joint Research Centre (JRC), Ispra, Italy <sup>2</sup> Institute for Advanced Sustainability Studies (IASS), Potsdam, 14467, Germany	Formatted: Font: 12 pt, Font color: Auto
13 14	<sup>3</sup> U.S. Environmental Protection Agency, Washington DC 20460, USA	Formatted: Font: 12 pt
15	<sup>4</sup> Institute of Environmental Protection, National Research Institute, Poland	
16	<sup>5</sup> Department of Climate, Air and Sustainability, TNO, Utrecht, The Netherlands	
17 18	<sup>6</sup> Asia Center for Air Pollution Research (ACAP), 1182 Sowa, Nishi-ku, Niigata-shi 950-2144, Japan	
19	<sup>7</sup> Japan Automobile Research Institute (JARI)	
20 21 22	<sup>8</sup> Air Quality Policy-Issue Response Section, Canadian Centre for Meteorological and Environmental Prediction, Environment and Climate Change Canada (ECCC), Dorval, Quebec, Canada	
23	<sup>9</sup> International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria	
24 25	<sup>10</sup> Joint Global Change Research Institute, Pacific Northwest National Lab, College Park, MD, USA	
26	<sup>11</sup> UniSystems Company, Milan (Italy)	
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28	<sup>13</sup> Department of Civil and Environmental Engineering, Konuk University, Seoul, South Korea	
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31 32	<sup>16</sup> Air Quality Research Division, Environment and Climate Change Canada, Toronto, Ontario, Canada	
33	<sup>17</sup> U.S. Environmental Protection Agency, North Carolina, USA	Formatted: Font: 12 pt, Font color: Auto
34	Correspondence to: Marilena.muntean@ec.europa.eu, monica.crippa@ext.ec.europa.eu	Formatted: Font: 12 pt, Font color: Auto
35	Abstract. This study, performed under the umbrella of the Task Force on Hemispheric	Formatted: Font: 12 pt
36	Transport of Air Pollution (TF-HTAP), responds to the need of the global and regional	Formatted: Font: 12 pt, Font color: Auto
37 38	atmospheric modelling community of having a mosaic emission inventory of air pollutants that conforms to specific requirements: global coverage, long time series, spatially distributed	Formatted: Font: 12 pt, Font color: Auto
30	contornis to specific requirements, global coverage, long time series, spatially distributed	Formatted: Font: 12 pt

1 emissions with high time resolution, and a high sectoral resolution. The mosaic approach of integrating official regional emission inventories based on locally reported data, with a global 2 inventory based on a globally consistent methodology, allows modellers to perform simulations 3 of a high scientific quality while also ensuring that the results remain relevant to policymakers. 4

5 HTAP v3, an ad-hoc global mosaic of anthropogenic inventories, has been developed by-

integrating official inventories over specific areas (North America, Europe, Asia including 6 7 Japan and Korea) with the independent Emissions Database for Global Atmospheric Research

8 (EDGAR) inventory for the remaining world regions. The results are spatially and temporally

9 distributed emissions of SO2, NOx, CO, NMVOC, NH3, PM10, PM2.5, Black Carbon (BC), and

10 Organic Carbon (OC), with a spatial resolution of  $0.1 \times 0.1$  degree and time intervals of months

11 and years covering the period 2000-2018 (DOI-https://doi.org/10.5281/zenodo.7516361,

12 https://edgar.jrc.ec.europa.eu/dataset htap v3). The emissions are further disaggregated to 16

13 anthropogenic emitting sectors. This paper describes the methodology applied to develop such

14 an emission mosaic, reports on source allocation, differences among existing inventories, and

best practices for the mosaic compilation. One of the key strengths of the HTAP v3 emission 15

mosaic is its temporal coverage, enabling the analysis of emission trends over the past two 16

decades. The development of a global emission mosaic over such long time series represents a 17

unique product for global air quality modelling and for better-informed policy making, 18

reflecting the community effort expended by the TF-HTAP to disentangle the complexity of 19

transboundary transport of air pollution. 20

#### 21 **1** Introduction

47

22 Common international efforts have procured an agreement to reduce global air pollutant 23 emissions. For this purpose, the United Nations Economic Commission for Europe (UNECE) Convention on Long Range Transboundary Air Pollution (CLRTAP) and the Task Force on 24 Hemispheric Transport of Air Pollution (TF-HTAP) have been instrumental in developing the 25 26 understanding of intercontinental transport of air pollution and thus contributing to the reduction of key pollutants in Europe and North America. 27

28 The success of CLRTAP is based on meeting strict reduction targets for pollutant releases. 29 Therefore, evaluating the resulting implications of these reductions requires an ongoing 30 improvement of global emission inventories in terms of emission updating and of 31 methodological refinements. These aspects are instrumental to gain understanding of 32 transboundary air pollution processes and drivers and to measure the effectiveness of emissions reduction and air quality mitigation policies. New guidance is available to achieve further 33 emission reductions across all emitting sectors. For example, the 2019 establishment of the 34 35 Task Force for International Cooperation on Air Pollution, which is intended to promote 36 international collaboration for preventing and reducing air pollution and improving air quality 37 globally (UNECE, 2021). As part of the ongoing effort by CLRTAP to reduce emissions and 38 to set out more effective and accountable mitigation measures, the 2005 Gothenburg Protocol 39 (UNECE, 2012) has been revised, including the review of the obligations in relation to 40 emission reductions and mitigation measures (e.g., black carbon and ammonia) and the review 41 of the progress towards achieving the environmental and health objectives of the Protocol. 42

The Task Force on Hemispheric Transport of Air Pollution (TF-HTAP) of the Convention has 43 44 a mandate to promote the scientific understanding of the intercontinental transport of air pollution to and from the UNECE area (https://unece.org/geographical-scope), to quantify its 45 46 impacts on human health, vegetation and climate, and to identify emission mitigation options that will shape future global policies.

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1 This paper describes and discusses a consistent global emission inventory of air pollutants 2 emitted by anthropogenic activities. This important database has been developed to assess the 3 contribution of anthropogenic air pollution emission sources within and outside the UNECE-4 area through atmospheric modelling. This inventory has been compiled based on officially 5 reported emissions, and an independent global inventory where officially reported emissions 6 are not used. This harmonised emissions "mosaic" dataset, hereafter referred to as the 7 HTAP v3, contains annual and monthly:

- emission time series (from 2000 to 2018) of SO<sub>2</sub>, NOx (expressed as NO<sub>2</sub> mass unit), CO, NMVOC, NH<sub>3</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC by emitting sector and country, and

8

9

10

- spatially distributed emissions on a global grid with spatial spacing of 0.1x0.1 degree.

11 The creation of a global emission mosaic requires the harmonisation of several data sources. 12 detailed analysis of contributing sectors for the different input inventories, development of data quality control procedures, and a robust and consistent gap-filling methodology when lacking 13 information. The development of HTAP v3 builds upon the previous experience of the 14 15 HTAPv1 (Janssens-Maenhout et al., 2012), and HTAPv2.2 (Janssens-Maenhout et al., 2015), 16 global inventories. HTAP\_v3, as requested by the TF-HTAP modelling community, provides 17 a more refined sectoral disaggregation compared to the previous HTAP emission mosaics. It 18 also includes tools (https://edgar.jrc.ec.europa.eu/htap\_tool/) that allow the extraction of emission data over selected domains (detailed later in section 4). 19

This paper describes the development the HTAP v3 database as a global anthropogenic air 20 pollutant emissions inventory mosaic for the period 2000-2018. The HTAP v3 mosaic has 21 been composed by integrating official, spatially distributed emissions data from CAMS-REG-22 23 v5.1 (Kuenen et al., 2022), US EPA (U.S. Environmental Protection Agency, 2021b, a), 24 Environment and Climate Change Canada (ECCC) (NPRI, 2017), REAS, CAPSS-KU, and 25 JAPAN (https://www.env.go.jp/air/osen/pm/info.html) (Kurokawa and Ohara, 2020; Chatani 26 et al., 2018; Chatani et al., 2020) inventories. As the information gathered from the official 27 reporting covers only part of the globe, HTAP\_v3 has been completed using emissions from 28 the Emissions Database for Global Atmospheric Research (EDGAR) version 6.1 29 (https://edgar.jrc.ec.europa.eu/dataset ap61).

30 One of the key strengths of the HTAP v3 emission mosaic is the temporal coverage of the emissions, spanning the 2000-2018 period, enabling the analysis of emission trends over the 31 past two decades. The development of a global emission mosaic over such long time series 32 33 represents a unique product for air quality modelling and for better-informed policy making, reflecting the effort of the TF-HTAP community to improve understanding of the 34 35 transboundary transport of air pollution. The year 2000 was chosen as the start-year, year since it often represents the year from which complete datasets of annual air pollutant emissions can 36 be generated. It also represents a turning point for several emerging economies (e.g., China) 37 and the strengthening of mitigation measures in historically developed regions (e.g., EU, USA, 38 39 etc.).

40 The two previous HTAP emission mosaics had limited temporal coverage. HTAPv1 covered 41 the period 2000-20005 with annual resolution (https://edgar.jrc.ec.europa.eu/dataset\_htap\_v1, 42 (Janssens-Maenhout et al., 2012)), while HTAPv2.2 covered two recent years (2008 and 2010), al., 2015) 43 but with monthly resolution (Janssens-Maenhout et (https://edgar.jrc.ec.europa.eu/dataset htap v2). However, the needs of the TF-HTAP 44 45 modelling community are continuously evolving to both foster forward-looking air quality science and produce more fit-for-purpose analyses in support of efficient policy making, 46 47 HTAP v3 therefore not only covers the time period of the previous HTAP phases, but also

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extends it forward by almost a decade, to provide the most up-to-date picture of global air
 pollutant emission trends. Another distinguishing feature of the HTAPv3 mosaic is a
 considerably higher sectoral resolution than previous iterations of the HTAP mosaic
 inventories (section 2.2), enabling more policy-relevant use of the inventory.

5 The methodology and data sources for the HTAP\_v3 emission mosaic are described in section 6 2. The long-time coverage of two decades, allows comprehensive trend analysis (see section 7 3), the HTAP\_v3 data format and data-set access are presented in section 4 and conclusions 8 are provided in section 5.

9

### 10 2 HTAP\_v3 emission mosaic overview: data sources, coverage, and methodology

#### 11 2.1 Data input

12 The HTAP v3 mosaic is a database of monthly- and sector-specific global air pollutant emission gridmaps developed by integrating spatially explicit regional information from recent 13 officially-reported national or regional emission inventories. Data from six main regional 14 15 inventories were integrated into HTAP v3, which covered only North America, Europe, and a portion of Asia (including Japan, China, India, and South Korea) (Fig.1). The geographical 16 domain covered by each of these inventories is depicted in Fig. 1, while further details on each 17 18 contributing inventory are presented in section 2.3. The emissions for all other countries, 19 international shipping and aviation (international and domestic) have been retrieved from the 20 Emissions Database for Global Atmospheric Research (EDGARv6.1. https://edgar.jrc.ec.europa.eu/dataset\_ap61) as represented by the grey areas in Fig.1. 21 22 Depending on the pollutant, more than half of global emissions are provided by region-specific 23 inventories, while the remaining contribution is derived from the EDGAR global inventory as reported in the bar graph of Fig.1, where the share of each individual inventory to global 24 emissions is represented. For all pollutants, the Asian domain is contributing most to global 25 emissions, hence the importance of having accurate emission inventories for this region. 26

27 Recent literature studies (Puliafito et al., 2021; Huneeus et al., 2020; Álamos et al., 2022; Keita et al., 2021; MEIC, 2022) document additional regional/local inventories which may contribute 28 29 to future updates of HTAP v3, in particular extending the mosaic compilation to regions in the Southern Hemisphere. Considering relative hemispheric emission levels as well as the 30 atmospheric dynamics happening in the Northern Hemisphere and regulating the 31 transboundary transport of air pollution, the current HTAP v3 mosaic should still satisfy the 32 needs of the atmospheric modelling community, although improvements using latest available 33 inventories for Africa and South America may also be considered for future updates, 34

Table 1 provides an overview of all data providers, in terms of geographical and temporal coverage, data format, and sectoral and pollutant data availability. Table 2 defines the HTAP\_v3 sectors and corresponding IPCC codes. Table 3 further details the sector-pollutant data availability for each inventory and the gap-filling approach required for some sectors and pollutants.

#### 40 2.2 Pollutant, spatial, temporal and sectoral coverage

The HTAP\_v3 emission mosaic helps to address the transboundary role of air pollutants by providing a key input for atmospheric modellers and supporting the evaluation of environmental impact analyses for poor air quality. For this reason, HTAP\_v3 provides global 0.1 x 0.1 degree emission gridmaps for all air pollutants and specifically for acidifying and Formatted: Font: 12 pt

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1 eutrophying gases (such as SO<sub>2</sub>, NH<sub>3</sub>, NOx), ozone precursors (NMVOC, CO, NOx), and primary particulate matter (PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC). 2

Emissions from each officially-reported inventory were submitted to HTAP on 0.1 x 0.1 degree 3

regional gridmaps. Spatial allocation was performed to these gridmaps for each sector by each 4

5 inventory group using the best available set of subsector spatial surrogate fields used by each group (e.g., https://www.cmascenter.org/sa-tools). EDGARv6.1 global gridmaps are also on a

6 7

0.1 x 0.1 degree grid.

Compared to the two previous HTAP emission mosaics, HTAP v3 input emission gridmaps 8 9 were provided with monthly time distributions to better reflect the regional seasonality of sector 10 specific emissions (e.g., household, power generation, and agricultural activities). Information on emission peaks over certain months of the year is also a useful information for the 11 development of territorial policies to mitigate localised emission sources in space and time 12

13 (e.g., emissions from residential heating over winter months, agricultural residue burning, etc.).

14 The HTAP v3 mosaic provides emissions for gaseous and particulate matter air pollutants 15 arising from all anthropogenic emitting sectors except for wildfires and savannah burning, which represent major sources of particulate matter and CO emissions. Wildfires and savannah 16 burning are not included in the current mosaic since community efforts are ongoing to tackle 17 these sources specifically. Modellers can find these additional sources on several publicly 18 available global wildfire emission datasets compiled based on the best available scientific 19 20 knowledge. such as the Global Fire Emission Database (GFED. https://www.globalfiredata.org/) or the Global Wildfire Information System (GWIS, 21 https://gwis.jrc.ec.europa.eu/). When using satellite retrieved emissions from fires, they should 22 be treated with caution to avoid double counting the emissions released by e.g. agricultural 23 24 crop residue burning activities.

25 HTAP\_v3 provides emissions at higher sectoral disaggregation than previous HTAP experiments<sup>1</sup><sub>AA</sub> to better understand drivers of emission trends and the effectiveness of sector-26 27 specific policy implementation. Emissions from 16 sectors are provided by the HTAP v3 28 mosaic, namely: International Shipping; Domestic Shipping; Domestic Aviation; International Aviation; Energy; Industry; Fugitives; Solvent Use; Road Transport; Brake and Tyre Wear; 29 30 Other Ground Transport; Residential; Waste; Agricultural Waste Burning; Livestock; and 31 Agricultural Crops. Further details on the sector definitions as well as their correspondence with the IPCC codes (IPCC, 1996, 2006) are provided in Table 2. The selection of the number 32 of sectors was constrained by the sectoral disaggregation of the input inventories (see Table 33 34 S1). Table 3 provides the complete overview of the emission data provided by each inventory group indicating the pollutants covered for each sector and eventual gap-filling information 35 included using the EDGARv6.1 data. Table 4 reports a summary of the main features of the 3 36 HTAP emission mosaics, showing the advancements achieved with this work. The high sector 37 disaggregation available within the HTAP v3 mosaic gives needed flexibility to modellers to 38 include or exclude emission sub-sectors in their simulations, in particular when integrating the 39 anthropogenic emissions provided by HTAP v3 with other components (e.g. natural 40 41 emissions, forest fires, etc.). However, we recommend particular caution when using a natural

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<sup>&</sup>lt;sup>1</sup>HTAPv1 covered 10 broad emission sectors (Aircraft, Ships, Energy, Industry Processes, Ground Transport, Residential, Solvents, Agriculture, Agriculture Waste Burning, and Waste), while even broader sectoral emissions were provided in HTAPv2.2 (Air, Ships, Energy, Industry, Transport, Residential (including waste), and Agriculture (only for NH<sub>3</sub>)).

 emissions model such as MEGAN (Model of Emissions of Gases and Aerosols from Nature, https://www2.acom.ucar.edu/modeling/model-emissions-gases-and-aerosols-nature-megan),
 which includes the estimation of NMVOC emissions from crops and soil NOx emissions

4 (including agricultural soils) that are also provided by the HTAP\_v3 mosaic.

#### 6 **2.3 Inventory overviews**

7 In the following sub-sections, details are provided on each officially-reported inventory used
 8 to construct the HTAP\_v3 emission mosaic.

### 9 2.3.1 CAMS-REG-v5.1 inventory

The CAMS-REG-v5.1 emission inventory was developed to support air pollutant and 10 greenhouse gas modelling activities at the European scale. The inventory builds largely on the 11 official reported data to the UN Framework Convention on Climate Change (UNFCCC) for 12 greenhouse gases (for CO<sub>2</sub> and CH<sub>4</sub>), and the Convention on Long-Range Transboundary Air 13 Pollution (CLRTAP) for air pollutants. For the latter, data are collected for NOx, SO<sub>2</sub>, CO, 14 NMVOC, NH<sub>3</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>, including all major air pollutants. For each of these pollutants, 15 the emission data are collected at the sector level at which these are reported for the time series 16 2000-2018 for each year and country. The CAMS-REG inventory covers UNECE-Europe, 17 18 extending eastward until 60°E, therefore including the European part of Russia. For some non-EU countries, the reported data are found to be partially available or not available at all. In 19 20 other cases, the quality of the reported data is found to be insufficient, i.e. with important data gaps or following different formats or methods. In this case, emission data from the IIASA 21 GAINS model instead (IIASA, 2018), are used. This model is the main tool used to underpin 22 23 pan-European and EU level air quality policies such as the UNECE Convention on Long Range Transboundary Air Pollution (UNECE, 2012), and the EU National Emission reduction 24 25 Commitments Directive (European Commission, 2016), After collecting all the emission data from reporting and GAINS, the source sectors are 26 27 harmonised, distinguishing around 250 different subsectors. For each detailed sector, a speciation is applied to the  $PM_{2.5}$  and  $PM_{10}$  emissions, distinguishing elemental carbon 28 (representing BC in the HTAP\_v3 inventory), organic carbon and other non-carbonaceous 29 emissions for both the coarse (2.5-10  $\mu$ m) and fine (<2.5  $\mu$ m) mode. 30

31 A consistent spatial resolution is applied across the entire domain, where a specific proxy is selected for each subsector to spatially distribute emissions, including for instance the use of 32 point source emissions, e.g., from the European Pollutant Release and Transfer Register (E-33 34 PRTR), complemented with additional data from the reporting of EU Large Combustion Plants (European Commission, 2001) and the Platts/WEPP commercial database for power plants 35 36 (Platts, 2017), Road transport emissions are spatially disaggregated using information from 37 OSM (Open Street Map, 2017), combined with information on traffic intensity in specific road segments from OTM (OpenTransportMap, 2017), Agricultural livestock emissions are 38 39 spatially distributed using global gridded livestock numbers (FAO, 2010), Furthermore, 40 CORINE land cover (Copernicus Land Monitoring Service, 2016) and population density are 41 other key spatial distribution proxies.

After having spatially distributed the data, the ~ 250 different source categories are aggregated
to fit with the HTAP\_v3 sector classification (Table S1). CAMS-REG-v5.1 is an update of an
earlier version, CAMS-REG-v4.2 and based on the 2020 submissions to cover the years 20002018. A detailed description of the CAMS-REG-v4.2 inventory is provided in Kuenen et al.
(2022).

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1 2 3 4 5	The data are provided as gridded annual totals at a resolution of 0.05°x0.1° (lat-lon). Along with the grids, additional information is available including height profiles as well as temporal profiles to break down the annual emissions into hourly data (monthly profiles, day-of-the-week profiles and hourly profiles for each day). Furthermore, the CAMS-REG inventory provides dedicated speciation profiles for NMVOC per year, country and sector.			
6	2.3.2 US EPA inventory			
7 8 9 10 11 12 13 14 15	Emissions estimates for the United States were based primarily on estimates produced for the EPA's Air QUAlity TimE Series Project (EQUATES), which generated a consistent set of modelled emissions, meteorology, air quality, and pollutant deposition for the United States spanning the years 2002 through 2017 ( <u>https://www.epa.gov/cmaq/equates</u> ). For each sector, a consistent methodology was used to estimate emissions for each year in the 16-year period, in contrast to the evolving methodologies applied in the triennial U.S. National Emissions Inventories (NEIs) produced over that span. The HTAPv3 time series was extended back one year to 2001 and forward one year to 2018 using country, sector, and pollutant specific trends from EDGARv6.1.	F	formatted: Font: 12 pt, Font color: Auto formatted: Font: 12 pt, Font color: Auto formatted: Font: 12 pt	
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17 18	Emissions estimates were calculated for more than 8000 Source Classification Codes grouped into 101 sectors and then aggregated to the 16 HTAP_v3 emission sectors. The 2017 NEI (U.S.		ormatted: Font: 12 pt	$\Box$
19 20	Environmental Protection Agency, 2021b) served as the base year for the time series. For each sector, emissions estimates were generated for previous years using one of four methods: 1)	F	ormatted: Font: 12 pt	
21	applying new methods to create consistent emissions for all years, 2) scaling the 2017 NEI			
22	estimates using annual sector-specific activity data and technology information at the county			
23	level, 3) using annual emissions calculated consistently in previous NEIs and interpolating to			
24 25	fill missing years, and 4) assuming emissions were constant at 2017 levels. The assumption of constant emissions was applied to a very limited number of sources. Foley et al. (2023)			
26	provides a detailed explanation of the assumptions used for each sector.	F	formatted: Font: 12 pt	
27 28 29	Emissions from electric generating units were estimated for individual facilities, combining available hourly emissions data for units with continuous emissions monitors (CEMs) and applying regional fuel-specific profiles to units without CEMS. On-road transport and non-			
30	road mobile emissions were estimated using emission factors from the MOVES v3 model (U.S.	F	formatted: Font: 12 pt	
31 32	Environmental Protection Agency, 2021a), A complete MOVES simulation was completed only for the NEI years with national adjustment factors applied for years plus or minus one	F	formatted: Font: 12 pt	
33	from the NEI years. For California, emission factors for all on-road sources for all years were			
34	based on the California Air Resources Board Emission Factor Model (EMFAC)			
35	(https://ww2.arb.ca.gov/our-work/programs/mobile-source-emissions-inventory/). New non-	F	ormatted: Font: 12 pt, Font color: Auto	
36	road emissions estimates for Texas were provided by the Texas Commission on Environmental	F	ormatted: Font: 12 pt, Font color: Auto	
37 38	Quality. Emissions from oil and gas exploration and production were calculated using point source specific data and the EPA Oil and Gas Tool (U.S. Environmental Protection Agency,	F	formatted: Font: 12 pt	
39	2021b), incorporating year-specific spatial, temporal, and speciation profiles. Residential wood	[F	ormatted: Font: 12 pt	
40	combustion estimates were developed with an updated methodology incorporated into the 2017	F	formatted: Font: 12 pt	
41	NEI and scaled backward to previous years using a national activity as a scaling factor. Solvent			
42	emissions were estimated using the Volatile Chemical Product (VCPy) framework of Seltzer	C		
43 44	et al. (2021), Emissions from livestock waste were calculated with revised annual animal counts to address missing data and methodological changes over the period. Emissions for	$\sim$ $\geq$	ormatted: Font: 12 pt	$\exists$
44	counts to address missing data and memodological changes over the period. Emissions for	F	formatted: Font: 12 pt	

agricultural burning were developed using a new suite of activity data with the same
 methodology and input data sets from 2002 onwards. County-level estimates were only
 available for 2002 because activity data based on satellite information was not yet available.
 Emissions for forest wildfires, prescribed burns, grass and rangeland fires were also calculated
 in EQUATES but not included in the HTAP\_v3 data. For EQUATES, fugitive dust emissions

6 (e.g., unpaved road dust, coal pile dust, dust from agricultural tilling) were reduced to account

7 for precipitation and snow cover by grid cell. For use in HTAP\_v3, however, no

8 meteorological adjustments (which decrease annual PM<sub>10</sub> emissions by about 75% on average)
9 were applied to fugitive dust emissions. Wind-blown fugitive dust emissions are not included

10 in the estimates for other regions in the  $HTAP_v3$  mosaic

Non-point source emissions were allocated spatially based on a suite of activity surrogates (e.g. population, total road miles, housing, etc.), many of which are sector specific. The spatial allocation factors were calculated for the 0.1 degree grid used by EDGARv6.1 with no intermediate re-gridding. The spatial allocation factors for all sectors were held constant for the entire time series except for oil and gas sectors which were year-specific. Depending on the sector, either 2017-based or 2014-based surrogates were developed for the same sectors as in the EQUATES.

18 Emissions from the US EPA inventory were provided from 2002-2017 (Table 1). Emissions 19 for the year 2018 were estimated applying country, sector and pollutant specific trends from

for the year 2018 were estimated applying country, sector and pollutant specific trends from EDGAR, as well as for years 2000 and 2001 to complete the entire time series. Table S1

provides an overview about the US EPA inventory sector mapping to the HTAP v3 sectors.

## 22 2.3.3 Environment and Climate Change Canada (ECCC) inventory

23 The Canadian emissions inventory data were obtained from 2018-released edition of Canada's Air Pollutant Emissions Inventory (APEI) originally compiled by the Pollutant Inventories and 24 Reporting Division (PIRD) of Environment and Climate Change Canada (ECCC) (APEI, 25 26 2018). This inventory contains a comprehensive and detailed estimate of annual emissions of seven criteria air pollutants (SO2, NOx, CO, NMVOC, NH3, PM10, PM2.5) at the national and 27 provincial/territorial level for each year for the period from 1990 to 2016. The APEI inventory 28 was developed based on a bottom-up approach for facility-level data reported to the National 29 30 Pollutant Release Inventory (NPRI) (NPRI, 2017), as well as an in-house top-down emission estimates based on source-specific activity data and emissions factors. In general, 31 methodologies used to estimate Canadian emissions are consistent with those developed by the 32 U.S. EPA (EPA, 2009) or those recommended in the European emission inventory guidebook 33 (EMEP/EEA, 2013). These methods are often further adjusted by PIRD to reflect the Canadian 34 35 climate, fuels, technologies and practices.

To prepare emissions in the desired HTAP classification, the APEI sector emissions were first mapped to the United Nations Economic Commission for Europe (UNECE) Nomenclature for Reporting (NFR) categories, which involved dividing the sector emissions into their combustion and process components. The NFR categories were then mapped to the HTAP 16 sector categories provided in the sector disaggregation scheme guide. Table S1 provides an overview of ECCC sector mapping to the HTAP\_v3 sectors.

The HTAP-grouped APEI inventory emissions files were further processed by the Air Quality
Policy-Issue Response (REQA) Section of ECCC to prepare the air-quality-modelling version
of inventory files in the standard format (i.e., FF10 format) supported by the U.S EPA

45 emissions processing framework. To process emissions into gridded, speciated and total 46 monthly values, a widely-used emissions processing system called the Sparse Matrix Operator Formatted: Font: 12 pt, Font color: Auto

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Kernel Emissions (SMOKE) model, version 4.7 (UNC, 2019), was used. As part of the 1 preparation for SMOKE processing, a gridded latitude-longitude North American domain at 2 0.1 x 0.1 degree resolution was defined with 920 columns and 450 rows covering an area of -3 142W to -50W and 40N to 85N. The point-source emissions in the APEI include latitude and 4 longitude information so those sources were accurately situated in the appropriate grid cell in 5 the Canadian HTAP gridded domain. However, to allocate provincial-level non-point source 6 emissions into this domain, a set of gridded spatial surrogate fields was generated for each 7 8 province from statistical proxies, such as population, road network, dwellings, crop 9 distributions, etc. Over 80 different surrogate ratio files were created using the 2011 Canadian census data obtained from Statistics Canada website (https://www12.statcan.gc.ca/census-10 11 recensement/2011/index-eng.cfm) and other datasets, such as the Canadian National Road Network (https://open.canada.ca/data/en/dataset/3d282116-e556-400c-9306-ca1a3cada77f). 12

To map the original APEI inventory species to the HTAP's desired list of species, PM speciation profiles from the SPECIATE version 4.5 database (EPA, 2016) were used to calculate source-type-specific EC and OC emissions. As a final step in SMOKE processing, the monthly emissions values were estimated using a set of sector-specific temporal profiles developed and recommended by the U.S. EPA (Sassi, 2021). For the point sources the NPRI annually reported monthly emissions proportions were applied. Emissions for the years 2017 and 2018 were calculated by applying sector- and pollutant-specific trends from EDGAR.

### 20 2.3.4 REASv3.2.1 inventory

The Regional Emission inventory in ASia (REAS) series have been developed for providing historical trends of emissions in the Asian region including East, Southeast, and South Asia. REASv3.2.1, the version used in HTAP\_v3, runs from 1950 to 2015. REASv3.2.1 includes emissions of SO<sub>2</sub>, NO<sub>x</sub>, CO, NMVOCs, NH<sub>3</sub>, CO<sub>2</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>, BC, and OC from major anthropogenic sources: fuel combustion in power plant, industry, transport, and domestic sectors; industrial processes; agricultural activities; evaporation; and others.

27 Emissions from stationary fuel combustion and non-combustion sources are traditionally calculated using activity data and emission factors, including the effects of control 28 technologies. For fuel consumption, the amount of energy consumption for each fuel type and 29 sector was obtained from the International Energy Agency World Energy Balances for most 30 countries and province-level tables in the China Energy Statistical Yearbook were used for 31 China. Other activity data such as the amount of emissions produced from industrial processes 32 33 were obtained from related international and national statistics. For emission factors, those without effects of abatement measures were set and then, effects of control measures were 34 considered based on temporal variations of their introduction rates. Default emission factors 35 and settings of country- and region-specific emission factors and removal efficiencies were 36 obtained from scientific literature studies as described in Kurokawa and Ohara (2020), and 37 38 references therein.

39 Emissions from road transport were calculated using vehicle numbers, annual distance 40 travelled, and emission factors for each vehicle type. The number of registered vehicles were 41 obtained from national statistics in each country and the World Road Statistics. For emission factors, year-to-year variation were considered by following procedures: (1) Emission factors 42 of each vehicle type in a base year were estimated; (2) Trends of the emission factors for each 43 vehicle type were estimated considering the timing of road vehicle regulations in each country 44 45 and the ratios of vehicle production years; (3) Emission factors of each vehicle type during the target period were calculated using those of base years and the corresponding trends. 46

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In REASv3.2.1, only large power plants were treated as point sources. For emissions from
 cement, iron, and steel plants, grid allocation factors were developed based on positions,
 production capacities, and start and retire years for large plants. Gridded emission data of
 EDGARv4.3.2 were used for grid allocation factors for the road transport sector. Rural, urban,
 and total population data were used to allocation emissions from the residential sector. For
 other sources, total population were used for proxy data.

For temporal distribution, if data for monthly generated power and production amounts of
industrial products were available, monthly emissions were estimated by allocating annual
emissions to each month using the monthly data as proxy. For the residential sector, monthly
variation of emissions was estimated using surface temperature in each grid cell. If there is no
appropriate proxy data, annual emissions were distributed to each month based on number of

12 dates in each month.

Monthly gridded emission data sets at 0.25° ★0x0,25° resolution for major sectors and emission
table data for major sectors and fuel types in each country and region during 1950-2015 are
available in text format from a data download site of REAS (<u>https://www.nies.go.jp/REAS/</u>).
Table S1 provides an overview about the REASv3.2.1 sector mapping to the HTAP\_v3 sectors.

More details of the methodology of REASv3.2.1 are available in Kurokawa and Ohara (2020) and its supplement. (Note that REASv3.2.1 is the version after error corrections of REASv3.2

of Kurokawa and Ohara (2020)). Details of the error corrections are described in the data

20 download site of REAS.) Table S1 provides an overview about the REASv3.2.1 sector mapping

21 to the HTAP v3 sectors.

The MEIC inventory (<u>http://meicmodel.org</u>, 2021) is not currently included in the HTAP\_v3 mosaic. Since the REAS inventory only includes emissions until 2015, the REAS-based HTAP\_v3 mosaic is only complete until this year. Emissions beyond 2015 were extrapolated using trends derived from a combination of MEIC and EDGAR. To extend the Chinese

26 emission estimates to most recent years, MEIC data were used to adjust sector and pollutant 27 specific trend for China for the years 2016 and 2017 (refer to Table S2 for the mapping sectors

of MEIC and HTAP\_v3). Then, the 2018 data were calculated based on the 2015-2017 trend.

29 For all the other countries belonging to the REAS domain, the emissions were extended beyond

30 2015 applying the sector-, country-, and pollutant-specific trends from EDGAR.

### 31 2.3.5 CAPSS-KU inventory

In the Republic of Korea, the National Air Emission Inventory and Research Center (NAIR) 32 estimates annual emissions of the air pollutants CO, NOx, SOx, TSP, PM10, PM2.5, BC, VOCs, 33 and NH3 via the Clean Air Policy Support System (CAPSS). The CAPSS inventory is divided 34 35 into four source-sector levels (high, medium, low and detailed) based on the European 36 Environment Agency's (EEA) CORe Inventory of AIR emissions (EMEP/CORINAIR). For activity data, various national- and regional-level statistical data collected from 150 domestic 37 institutions are used. For large point sources, emissions are estimated directly using real-time 38 stack measurements. For small point, area and mobile sources, indirect calculation methods 39 40 using activity data, emission factors, and control efficiency are used.

Even though CAPSS (Clean Air Policy Support System) has been estimating annual emissions since 1999, some inconsistencies exist in the time series because of the data and methodological changes over the period. For example, emissions of  $PM_{2.5}$  were initiated from the year 2011 and not from 1999. Therefore, in the CAPSS emission inventory,  $PM_{2.5}$  emissions were calculated from 2011, and post-2011 the  $PM_{10}$  to  $PM_{2.5}$  emission ratio was used to calculate the emissions from 2000 to 2010. These limitations make it difficult to compare and analyse

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emissions inter-annually. To overcome these limitations, re-analysis of the annual emissions
 of pollutants was conducted using upgrades of the CAPSS inventory, such as missing source
 addition and emission factor updates.

The biomass combustion and fugitive dust sector emissions from 2000 to 2014 were estimated and added in the inventory, which are newly calculated emission sources from 2015. As for the on-road mobile sector, new emission factors using 2016 driving conditions were applied from the year 2000 to 2015. Since the emissions from the combustion of imported anthracite coal were calculated only from 2007, the coal use statistics of imported anthracite from 2000 to 2006 were collected to estimate emissions for those years.

10 After all the adjustments, a historically re-constructed emissions inventory using the latest 11 emission estimation method and data was developed. Table S1 provides an overview about the

12 CAPSS sector mapping to the HTAP v3 sectors.

### 13 2.3.6 JAPAN inventory (PM2.5EI and J-STREAM)

14 The Japanese emission inventory contributing to the HTAP\_v3 mosaic is jointly developed by 15 the Ministry of the Environment, Japan (MOEJ) for emissions arising from mobile sources and 16 by the National Institute of Environmental Studies (NIES) for estimating emissions from fixed 17 sources.

The mobile source emissions data for the HTAP\_5.1, 5.2, and 5.4 sectors are based on the air 18 19 pollutant emission inventory named "PM2.5 Emission Inventory (PM2.5ED). https://www.env.go.jp/air/osen/pm/info.html). PM2.5EI has been developed for the years 2012 20 and 2015, while for 2018 is currently under development. Almost all anthropogenic sources 21 22 are covered, but emissions from vehicles are estimated in particular detail based on JATOP 23 (Shibata, 2021). The emission factor of automobiles is constructed by MOEJ as a function of 24 the average vehicle speed over several kilometres in a driving cycle that simulates driving on 25 a real road. Emission factors are organized by 7 types of vehicles, 2 fuel types, 5 air pollutants, 26 and regulation years, and have been implemented since 1997 as a project of MOEJ. By using 27 these emission factors and giving the average vehicle speed on the road to be estimated, it is 28 possible to estimate the air pollutant emissions per kilometre per vehicle. The hourly average vehicle speed of trunk roads, which account for 70% of Japan's traffic volume, is obtained at 29 30 intervals of several kilometres nationwide every five years, so the latest data for the target year is used. For narrow roads, the average vehicle speed by prefecture measured by probe 31 information is applied. It is 20 km/h in Tokyo, but slightly faster in other prefectures. Starting 32 emission is defined as the difference between the exhaust amount in the completely cold state 33 and the warm state in the same driving cycle and is estimated by the times the engine started 34 in a day. Chassis dynamometer tests are performed in a well-prepared environment, so for more 35 realistic emissions estimates, temperature correction factor, humidity correction factor, 36 37 deterioration factor, DPF regeneration factor, and soak time correction factor are used. In 38 addition to running and starting emissions, evaporative emissions from gasoline vehicles and 39 non-exhaust particles such as road dust (including brake wear particles) and tire wear particles are combined to provide a vehicle emissions database with a spatial resolution of approximately 40 1 km  $\times$  1 km (30° latitude, 45° longitude), and a temporal resolution of an hour by month, 41

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42 including weekdays and holidays.

Off-road vehicle emissions are estimated separately for 17 types of construction machinery,
 industrial machinery (forklifts), and 5 types of agricultural machinery. In all cases, emission
 factors by type and regulatory year per workload are used, as researched by the MOEJ.
 Although not as precise as automobiles, the off-road database is provided with the same

5 temporal and spatial resolution as the automobile database.

Emissions from stationary sources in Japan are derived from the emission inventory developed 6 7 in the Japan's Study for Reference Air Quality Modelling (J-STREAM) model intercomparison 8 project (Chatani et al., 2018; Chatani et al., 2020). In this emission inventory, emissions from 9 stationary combustion sources are estimated by multiplying emission factors and activities 10 including energy consumption, which is available in the comprehensive energy statistics. Large 11 stationary sources specified by the air pollution control law need to report emissions to the government every three years. The emission factors and their annual variations were derived 12 from the emissions reported by over 100,000 sources (Chatani et al., 2020). For fugitive VOC 13 14 emissions, MOEJ maintains a special emission inventory to check progress on regulations and 15 voluntary actions targeting 30% reduction of fugitive VOC emissions starting from 2000. VOC emissions estimated in this emission inventory are used. Emissions from agricultural sources 16 are consistent with the emissions estimated in the national greenhouse gas emission inventory 17 (Center for Global Environmental Research et al., 2022), Emissions of all the stationary sources 18 are divided into prefecture, city, and grid (approximately 1 x 1 km) levels based on spatial 19 proxies specific to each source. Emissions for the year 2018 were estimated applying sector-20 21 and pollutant-specific trends from EDGAR. Table S1 provides an overview about the Japanese 22 inventory sector mapping to the HTAP v3 sectors.

### 23 2.4 Gap-filling methodology with EDGARv6.1

EDGAR is a globally consistent emission inventory of air pollutant and greenhouse gases 24 developed and maintained by the Joint Research Centre of the European Commission 25 (https://edgar.jrc.ec.europa.eu/). The EDGAR methodology used to compute GHG and air 26 27 pollutant emissions has been described in detail in several publications (Janssens-Maenhout et al., 2019; Crippa et al., 2018) and summarised here after. In EDGAR, air pollutant emissions 28 29 are computed making use of international statistics as activity data (e.g., International Energy 30 Balance data, Food and Agriculture Organisation statistics, US Commodity statistics, etc.), region- and/or country-specific emission factors by pollutant/sector and technology and 31 32 abatement measures, following Eq. 1:

$$EM_{i(C,t,x)} = \sum_{k,j,k} AD_{i(C,t)} * TECH_{i,j(C,t)} * EOP_{i,j,k(C,t)} * EF_{i(C,t,x)} * (1 - RED)_{i,j,k(C,t,x)}$$

34 (Eq. 1)

33

where EM are the emissions from a given sector i in a country C accumulated during a year t 35 for a chemical compound x, AD the country-specific activity data quantifying the human 36 activity for sector i, TECH the mix of j technologies (varying between 0 and 1), EOP the mix 37 38 of k (end-of-pipe) abatement measures (varying between 0 and 1) installed with a share k for each technology j, and EF the uncontrolled emission factor for each sector i and technology j 39 with relative reduction (RED) by abatement measure k. Emission factors are typically derived 40 from the EMEP/EEA Guidebooks (EMEP/EEA, 2013, 2019, 2016), the AP-42(EPA, 2009) 41 inventory and scientific literature. 42

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Annual country and sector specific air pollutant emissions are then disaggregated into monthly
 values (Crippa et al., 2020) and subsequently spatially distributed making use of detailed proxy
 data (Janssens-Maenhout et al., 2019; Crippa et al., 2021).

As the most comprehensive and globally consistent emission database, the latest update of the 4 5 EDGAR pollutant emissions inventory, EDGARv6.1 air 6 (https://edgar.jrc.ec.europa.eu/dataset ap61), is used in the HTAP v3 mosaic to complete missing information from the officially reported inventories, as reported in Table 3. 7 EDGARv6.1 includes important updates to estimate air pollutant emissions such as the 8 9 improvement of road transport emission estimates for many world regions, the inclusion of agricultural NMVOC emissions, revised monthly emission profiles (Crippa et al., 2020) and 10 11 updated spatial proxies to distribute national emissions by sector over the globe (Crippa et al., 2021), EDGARv6.1 also includes new international shipping proxies and their monthly 12 distribution based on the STEAM model (Jalkanen et al., 2012; Johansson et al., 2017), In the 13 14 Supplementary Material (section S2), the assessment of EDGAR emission data is reported in 15 comparison with global and regional inventories.

#### 16 3 Results

#### 17 3.1 Annual time series analysis: trends and regional and sectoral contributions

Having a consistent set of global annual emission inventories for a two-decade period allows the investigation of global emissions trends for the inventory pollutants and regional and sectoral contributions. Figure 2 presents annual time series (2000-2018) of the global emissions of the nine air pollutants included in the HTAP\_v3 mosaic separated into the actual contributions of 12 regions. Figure 3 shows the corresponding relative contributions of (a) 16 sectors and (b) 12 regions to the 2018 global emissions of these same pollutants. We can then discuss each pollutant in turn.

25 Global SO2 emissions declined from 99.4 to 72.9 Mt over the past two decades. This decreasing pattern is found for several world regions with the fastest decline in Eastern Asia, where after 26 27 the year 2005 SO<sub>2</sub> emissions began to decrease steadily. This is consistent with the use of cleaner fuels with lower sulphur content and the implementation of desulphurisation techniques 28 29 in power plants and industrial facilities in China in accordance with the 11th Five-Year Plan 30 (FYP, 2006–2010 (Planning Commission, 2008)) and the 12th Five-Year Plan (FYP, 2011– 2015 (Hu, 2016)) (Sun et al., 2018), Similarly, industrialised regions, such as North America 31 and Europe, are characterised by a continuous decreasing trend in SO<sub>2</sub> emission, which had 32 33 started well before the year 2000 due to the implementation of environmental and air quality legislation (EEA, 2022), Increasing SO2 emissions, on the other hand, are found for Southern 34 Asia (+112% compared to 2000), South-East Asia and developing Pacific (+62%), and Africa 35 36 (+40%). These increases mostly arise from the energy, industry, and (partly) residential 37 sectors, and reflect the need for emerging and developing economies to mitigate these emissions. Emissions estimated using satellite retrievals and model inversions confirm the 38 39 trends provided by the HTAP v3 mosaic (Liu et al., 2018). SO<sub>2</sub> is mostly emitted by power 40 generation and industrial activities, which in 2018 represent 42% and 26%, respectively, of the global total. Despite measures in some specific sea areas to mitigate sulphur emissions, globally 41 they have been rising steadily with increasing activity. International shipping represents 13.8% 42 of global SO<sub>2</sub> emissions in 2018, and it is 41% higher compared to the 2000 levels (Fig. 3). 43

<u>Global NOx emissions</u> increased from 110.4 Mt in 2000 to 117.4 Mt in 2018 as a result of the
 increase in energy- and industry-related activities for most of the world regions (in particular
 over the Asian domain). The strongest decreases are found for North America (-63%), Europe
 (-42%), Asia-Pacific Developed (-32%) and to a lower extent for Eurasia (-6%). Comparable

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1 2	spatio-temporal patterns are found by satellite OMI data and ground based measurements of NO2 concentrations (Jamali et al., 2020). NOx is mainly produced at high combustion		Formatted: Font: 12 pt
3 4	temperatures (e.g., power and industrial activities, 38% of the global total), but also by transportation (27% of the global total) and international shipping (14% of the global total).		Formatted: Font: 12 pt
5 6	<u>CO</u> is mostly emitted by incomplete combustion processes from residential combustion, transportation and the burning of agricultural residues. Globally, CO emissions showed little		
7 8	change over the past two decades (502.7 Mt in 2000 vs. 499.8 in 2018), but different regional trends are present. Historically industrialised regions have reduced their emissions over the		
9	years (-42% in Europe and -62% in North America), while CO emissions increased in Africa		
10 11	by 45% and in Southern Asia by 49%. Road transport CO emissions halved over the past two decades (-55%), while the emissions from all other sectors increased. These results are		
12	consistent with MOPITT satellite retrievals, which mostly show the same trends over the		
13	different regional domains over the past decades (Yin et al., 2015).	<	Formatted: Font: 12 pt
14 15	<u>NMVOC emissions</u> increased from 115.2 Mt in 2000 to 146 Mt in 2018. These emissions are mostly associated with the use of solvents (25% of the 2018 global total), fugitive emissions		Formatted: Font: 12 pt
15 16	(23%), road transportation (including both combustion and evaporative emissions, 15%) and		
17 18	small-scale combustion activities (19%). The most prominent increases in the emissions at the global level are found for the solvents sector (+81%). In 2018, NMVOC emissions from		
19	solvents were 3.7 and 3.5 times higher than in 2000 in China and India, respectively, while a		
20	rather stable trend in found for US and Europe.		
21 22	<u>Global NH<sub>3</sub> emissions</u> increased from 47.4 Mt in 2000 to 58.9 Mt in 2018 due to enhanced emissions from agricultural activities. In particular, NH3 emissions strongly increased in Africa		
23	(+60.5%), South-East Asia and developing Pacific (48.3%), Southern Asia (+38.7%), and Latin		
24	America and Caribbean (+41.1%).		
25 26	<u>Particulate matter emissions</u> showed little change over the past two decades at the global level, whereas regional emission increases are found for Africa (e.g., +47.0% for PM <sub>10</sub> ), Latin		
27	America and Caribbean (+39.1%), Middle East (48.3%), and Southern Asia (+56%), mostly		
28 29	associated with increases in agricultural waste burning and the livestock, energy, and waste sectors. By contrast, Eastern Asia (-39.5%), Europe (-24.3%), and Asia-Pacific Developed (-		
30	36.8%) significantly decreased their PM10 emissions over the past two decades due to the		
31 32	continuous implementation of reduction and abatement measures for the energy, industry, road transport and residential sectors ((Crippa et al., 2016), As shown in Fig. 3, the relative		Formatted: Font: 12 pt
33	contribution of North America to global PM10 is quite high compared to other substances due	<	Formatted: Font: 12 pt
34 35	to fugitive dust emissions (e.g., unpaved road dust, coal pile dust, dust from agricultural tilling) which have not been adjusted for meteorological conditions (e.g., rain, snow) and near-source		
36 27	settling and mitigation (e.g., tree wind breaks) because these removal mechanisms are better		
37 38	addressed by the chemical transport models. Additional uncertainty may be therefore introduced for these emissions, depending on the modelling assumptions of each official		
39 40	inventory. Similarly, particulate matter speciation into its carbonaceous components is often challenging and subjected to higher level of uncertainty, for instance because different		
40 41	definitions are used for PM in inventories, including condensable emissions or not (Denier van		Formatted: Font: 12 pt
42 43	der Gon et al., 2015), Attempts to improve the accuracy of such emissions (e.g. BC and OC emissions over the European domain) are ongoing.		Formatted: Font: 12 pt
44	Figure 3a shows more than 50% difference at the global level between $PM_{2.5}$ emissions and the		Formatted Foot: 12 pt Foot color: Auto
44 45	sum of its carbonaceous components (BC and OC), which however varies depending on the		Formatted: Font: 12 pt, Font color: Auto
46	region and sector. The largest difference between $PM_{2.5}$ and the sum of BC and OC is generally		

 $\label{eq:sector} 47 \qquad \text{found for the energy and industrial sectors, where due to the high temperatures BC and OC are}$ 

1 largely burned. Within this sector, the non carbonaceous fraction of PM2.5 represents around 75% in Europe, 78% in the USA and up to more than 95% over Asian countries (e.g. China 2 and India). This PM fraction is represented by other minerals, ash (mostly when burning coal) 3 4 and sulphate. Road transport is also a sector showing large differences between PM2.5 and the 5 sum of BC and OC, with around 40% difference for Europe, around 90% difference for USA and lower values for India and China (around 15%). This component may be associated with 6 other minerals. For the residential sector, this difference is generally lower and around 25% 7 (for Europe and Asian countries), while around 37% in the USA and is possibly associated 8 9 with other minerals and ash due to coal combustion. Shipping is also a sector where a large component of PM2.5 (around 70%) is not associated with carbonaceous fractions but to 10 11 sulphate. In particular, regions within the Sulphur Emission Control Area (SECA) show lower 12 contributions from sulphates (e.g. Europe and USA) with an overall contribution of 5-10%. Another source of uncertainty which may contribute to enhancing the difference between PM<sub>2.5</sub> 13 and the sum of BC and OC is associated on how different inventories consider condensable 14 15 particulate matter.

### 16 **3.2 Emission maps**

17 Spatially distributed emission data describe where emissions take place, as input for local, 18 regional and global air quality modelling. As noted in section 2.2, nationally aggregated air pollutant emissions are spatially distributed over the corresponding national territory using 19 spatial proxy data which are believed to provide a relatively good representation of where 20 emissions takes place. Depending on the emitting sector, air pollutants can be associated with 21 22 the spatial distributions of point sources (e.g., in the case of power plant or industrial activities), 23 road networks (e.g., for transportation related emissions), settlement areas (e.g., for small-scale combustion emissions), crop and livestock distribution maps, ship tracks etc. Using reliable 24 and up-to-date spatial information to distribute national emissions is therefore relevant, 25 although challenging. Multiple assumptions are often made by inventory compilers when 26 developing their inventories, which may result in differences when analysing spatially 27 28 distributed emissions provided by different inventory compilers over the same geographical 29 domain.

One key goal of the HTAP v3 mosaic is to collate in one inventory the most accurate spatially-30 distributed emissions for all air pollutants at the global level, based on the best available local 31 information. Point sources related with emissions from power plant and industrial facilities 32 represent one the most critical spatial information to be retrieved, and their misallocation can 33 34 significantly affect the characterisation of local air quality. This challenge is also present in the 35 HTAP v3 mosaic. For example, the REASv3.2.1 inventory is still using limited information to distribute emissions from these two sectors especially for industrial plants. Depending on 36 37 the region, point source information could be limited compared to datasets used in inventories of North America and Europe. To overcome this issue, the participation of national emission 38 39 inventory developers not only from China, but also India and other Asian countries is recommended. The impact can be seen in Fig. 4, which shows the global map of SO<sub>2</sub> emissions 40 41 in 2018 based on the HTAP\_v3 mosaic compilation, where information about the magnitude and the type of emission sources for the different regions can be retrieved. The energy and 42 industry sectors contribute a large fraction of SO<sub>2</sub> emissions (Fig. 3a), but the spatial 43 44 distribution of these emissions is qualitatively different in North America and Europe than in 45 Asia (i.e., more "spotty", less smooth and widely distributed). Ship tracks cover the entire geographical marine domain, consistent with emissions from the STEAM model (Jalkanen et 46 al., 2012; Johansson et al., 2017), included in the EDGARv6.1 database, although showing 47 marked emissions over the Mediterranean Sea, Asian domain, Middle East and North 48

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American coasts. Furthermore, emissions from power plant and industrial activities, as well as
 small-scale combustion are prominent over the Asian domain, Eastern Europe, and some
 African regions.

Sector-specific case studies are presented in the maps of Figs. 5-8. Figure 5 shows the 4 5 comparison of annual NOx emissions for the year 2000 and 2018. The road transport sector is 6 a key source of NOx emissions (cf. Fig. 3a), and this contribution is reflected in the visible presence of road networks in the maps. Decreasing emissions are found for industrialised 7 regions (USA, Europe, Japan) thanks to the introduction of increasingly restrictive legislation 8 9 on vehicle emissions since the 1990s, whereas a steep increase is found for emerging 10 economies and in particular India, China, and the Asian domain. Figure 6 shows the different 11 spatial allocation of  $PM_{10}$  emissions from the residential sector during the month of January 2018, with higher emission intensities evident in the Northern Hemisphere (cold season) and 12 the lower values in the Southern Hemisphere (warm season). Figures 7 and 8 show the spatio-13 temporal allocation of agriculture-related emissions, and specifically, PM<sub>10</sub> emissions from 14 15 agricultural waste burning and NH3 emissions from agricultural soil activities.

### 16 **3.3 Monthly temporal distribution**

#### 17 3.3.1 Monthly variability by region

18 The magnitude of air pollutant emissions varies by month because of the seasonality of 19 different anthropogenic activities and their geographical location (e.g., Northern vs. Southern 20 Hemisphere regions). Figures 9 and 10 (and S3.1, S3.2 and S3.3) show the monthly distribution of regional emissions for those pollutants and sectors for which higher variability is expected. 21 The year 2015 was chosen since it is the last year for which all of the official data providers 22 23 have data. Figure 9 shows monthly NH<sub>3</sub> emissions by region from three agricultural activities 24 (agricultural waste burning, livestock, and crops). These sectors display the largest variability by month, reflecting the seasonal cycle and the region-specific agricultural practices, such as 25 26 fertilisation, crop residue burning, manure and pasture management, animal population 27 changes, etc. In Figure 10, NOx emissions from residential activities show a particular monthly 28 distribution, with the highest emissions occurring during the cold months shifted for the Northern and Southern Hemispheres. By contrast, regions in the equatorial zone do not show 29 a marked monthly profile even for residential activities. The energy sector also follows 30 monthly-seasonal cycles related to the demand for power generation, which is also correlated 31 with ambient temperature and local day length. Transport-related emissions do not show a large 32 33 variation by month, whereas daily and weekly cycles for transport-related emissions, which are 34 typically more relevant, are beyond the temporal resolution of this work.

35 Although a spatio-temporal variability of the HTAP v3 emissions is found in these figures, a 36 more in-depth analysis reveals that with the exception of few regions and sectors (e.g., Canada, 37 USA and regions gap-filled with EDGAR), no inter-annual variability of the monthly profiles is present, meaning that the majority of official inventories assume the same monthly 38 distribution of the emissions for the past two decades (refer to Figs. S3.4-S3.9). This is different 39 40 from the approach used for example by EDGAR (Crippa et al., 2020), ECCC for Canada, and U.S. EPA for the USA, where year-dependent monthly profiles are used for specific sectors, in 41 particular for residential, power generation, and agricultural activities. Further analysis has 42 shown that for the European domain regional rather than country-specific monthly profiles are 43 applied. Therefore, for Europe new state-of-the-art profiles have been made available under 44 45 the CAMS programme by Guevara et al. (2021),

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#### 1 3.3.2 Spatially-distributed monthly emissions

2 An important added value of HTAP\_v3 comes from the availability of monthly gridmaps that 3 reflect the seasonality of the emissions for different world regions. Access to spatially 4 distributed monthly emissions is essential to design effective mitigation actions, providing 5 information on hot spots of emissions and critical periods of the year when emissions are 6 highest.

Figure 11 shows mid-season PM<sub>2.5</sub> monthly emissions arising from the residential sector in 7 2018. The global map shows higher emissions in the Northern Hemisphere during January, 8 while the opposite pattern is found for the Southern Hemisphere in July. Agriculture is an 9 important activity characterised by strong seasonal patterns, as shown in Figs. 12 and 13. Figure 10 12 shows PM<sub>10</sub> monthly emission maps from agricultural residue burning in 2018 from 11 12 HTAP v3, highlighting higher emissions over certain months of the year related with specific burning practices of agricultural residues for different world regions. For example, during the 13 month of April, intense burning of crop residues is found in Africa (Nigeria, Ethiopia, Sudan, 14 15 South Africa, etc.), South America (Brazil, Argentina, Colombia, etc.), Northern India, and South-Eastern Asia (e.g., Vietnam, Thailand, Indonesia, Philippines, etc.). Figure 13 represents 16 17 the yearly variability of NH<sub>3</sub> emissions from agricultural soils activities, mostly related with 18 fertilisation. During the month of March and April, intense agricultural soils activities are found over Europe and North America compared to other months, while during the month of October 19 the highest emissions are for this sector are found in China, India, several countries of the Asian 20 21 domain, but also in USA, Australia, and Latin America. These results are consistent with satellite based observations performed using Cross-track Infrared Sounder (Shephard et al., 22 23 2020).

### 24 **3.4 Vertical distribution of the emissions**

### 25 3.4.1 Aircraft emissions

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In EDGAR6.1 the emissions are provided at three effective altitude levels (landing/take-off, 26 27 ascent/descent, and cruising). The spatial proxy for the aviation sector is derived from International Civil Aviation Organization (ICAO, 2015), which specifies a typical flight pattern 28 29 with landing/take-off cycle within few km of the airport, followed by climb-out/descending 30 phase during the first 100 km and the last 100km of a flight and finally the remaining part from 101 km until the last 101 km as the cruise phase. Routes and airport locations are taken from 31 32 the Airline Route Mapper of ICAO (2015), In HTAP\_v3, aircraft emissions are provided as 33 domestic and international, but with no information about altitude ranges. We recommend corresponding 34 modellers to use the EDGARv6.1 data 35 (https://edgar.jrc.ec.europa.eu/dataset ap61) including the vertical distribution of the 36 emissions.

#### 38 3.4.2 Speciation of NMVOC emissions

39 For emission data to be useful for modellers, total NMVOC emissions must be decomposed into emissions of individual NMVOC species. As the chemical mechanisms used by models 40 can differ with respect to the NMVOC species they include, it is not practical to provide an 41 42 NMVOC speciation which is usable by all models. Instead, a speciation is provided here for 43 the set of 25 NMVOCs defined by Huang et al. (2017), and the corresponding data are made available on the HTAP v3 website. The absolute values of 25-category speciated NMVOC 44 emissions were obtained for all countries for the 28 EDGAR sectors from here: 45 46 https://edgar.jrc.ec.europa.eu/dataset\_ap432\_VOC\_spec\_ The absolute NMVOC emissions of each species from each sector in this dataset were remapped to the HTAP\_v3 sectors following 47

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1 the mapping from Table 2, then converted to a speciation by dividing by the total emissions of

each individual species for the four world regions defined by Huang et al. (2017): Asia; Europe; 2

North America; and Other. The resulting NMVOC speciation is provided in the supplementary 3

material to this paper for the 25 NMVOC species, 4 world regions, and 15 emitting NMVOC 4 5

sectors<sup>2</sup> following the HTAP\_v3 sector classification (including 13 sectors defined over the 4

6 world regions, and the two international sectors: international shipping and international aviation). The list of countries comprising each region is also provided in the supplement. 7

3.5 Qualitative assessment of the uncertainty of a global emission mosaic 8

9 Assessing the uncertainty of a global emission mosaic is challenging since it consists of several

10 bottom-up inventories and by definition it prevents a consistent global uncertainty calculation.

Each emission inventory feeding the HTAP v3 mosaic is characterized by its own uncertainty 11

which is documented by the corresponding literature describing each dataset (see Table 2 and 12

section 2.3) and which should be cited by the users of the mosaic for a quantitative assessment 13

of regional uncertainties. However, the mosaic compilation process may also introduce 14

15 additional uncertainties compared to the input datasets. In order to limit these additional

uncertainties, we made the following considerations: 16

17 -for each emission inventory both the national totals and gridded data by sector were gathered.

18 This process allows the mosaic compilers not to introduce additional uncertainty compared to

19 the original input regional datasets. In fact, additional uncertainties may arise from the 20

extraction of the national totals from spatially distributed data (e.g. country border issues which were one limitation of previous editions of the HTAP mosaics). Therefore, when regional 21

22 trends are described by region and pollutant (see section 3), no additional source of uncertainty

has to be considered from the mosaic compilation approach. 23

#### -the sector definition and mapping has been accurately developed following the IPCC 24 25 categories and when no data was available for a certain combination of sector and pollutant a gapfilling procedure is applied using the EDGAR database. Also in this case no additional 26 uncertainty should be considered compared to the input datasets. 27

-any additional uncertainty introduced by the temporal disaggregation can be deemed as 28 29 negligible since each inventory already provided monthly resolution emission gridmaps and 30 time series.

In this work we also provide a qualitative indication of the emission variability by HTAP sector 31 32 and pollutant at the global level. Table S6 summarises the variability of global HTAP\_v3 33 emissions by sector for the boundary years of this mosaic (2000 and 2018) compared to the 34 global EDGARv6.1 data. EDGAR emissions are considered as the reference global emission 35 inventory against which comparing the HTAP\_v3 estimates although these two global products are not fully independent. The variability of the global emissions is calculated as the relative 36 37 difference of the estimates of the two inventories, i.e. (EDGARv6.1-HTAP v3)/HTAP v3). 38 Emission variabilities are also classified as low (L, L<15%), low medium (LM, 15%<LM<50%), upper medium (UM, 50%<UM<100%), high (H, H>100%), based on the 39 40 EMEP/EEA Guidebook (2019) information. The largest variability is found domestic shipping 41 emissions (CO and NMVOC), energy (OC, BC), agricultural crops (PM), road transport (PM, NMVOC) and industry (NH3, NMVOC). In absence of a full uncertainty assessment the 42

<sup>2</sup> No speciation profile is provided for the 'tyre and brake wear sector' not being a source of NMVOC emissions.

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1 variability can be used as proxy of structural uncertainty, keeping in mind that variability could 2 be biased towards overconfidence, thus underestimating the uncertainty. Furthermore, the uncertainty of the spatial proxies has not been assessed and maybe subject of future activity 3 4 updates. 5 4 Data availability 6 7 The HTAP v3 emission mosaic data can be freely accessed and cited using DOI+ https://doi.org/10.5281/zenodo.7516361, All data can be also accessed through the EDGAR 8 website at the following link: https://edgar.jrc.ec.europa.eu/dataset htap v3. 9

10 Data are made available in the following formats:

- Monthly gridmaps of emissions (in Mg/month) at 0.1x0.1degree resolution: there is one
   NetCDF file per year and substance that includes the emissions for each sector for the
   12 months.
- Monthly gridmaps of emission fluxes (in kg/m2/s) at 0.1x0.1degree resolution: there is one .NetCDF file per year and substance that includes the emission fluxes for each sector the emission fluxes for the 12 months.
  - Annual gridmaps of emissions (in Mg/year at 0.1x0.1degree resolution: there is one .NetCDF file per year and substance that includes the emissions for each sector.
    - Annual gridmaps of emission fluxes (in kg/m2/s) at 0.1x0.1degree resolution: there is one .NetCDF file per year and substance that includes the emission fluxes for each sector.

The full set of HTAP v3 data is quite large, requiring substantial network bandwidth and time 22 23 for download, and substantial storage space. To make it easier for users to query and use the data, additional products are available. For global modellers who may not require such high 24 25 spatial resolution, gridmaps at 0.5x0.5 degree resolution are made available following the abovementioned specifications of the higher spatial resolution data. Furthermore, to allow 26 regional modellers to download only the data for the regions they need, the JRC EDGAR group 27 28 has also developed an interface to allow the users of the HTAP v3 mosaic to extract emission 29 data over arbitrarily specified geographical domains. The HTAP tool is accessible after creation of an ECAS account (https://webgate.ec.europa.eu/cas/login) and it is available at: 30 31 https://edgar.jrc.ec.europa.eu/htap\_tool/,

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## 33 5 Conclusions

34 The global air pollution mosaic inventory HTAP\_v3 presented and discussed in this paper is a 35 state-of-the-art database for addressing the present status and the recent evolution of a set of policy-relevant air pollutants. The inventory is made by the harmonization and blending of six 36 37 regional inventories, gapfilled using the most recent release of EDGAR (EDGARv6.1). By 38 directly incorporating the best available local information, including the spatial distribution of emissions, the HTAP v3 mosaic inventory can be used for policy-relevant studies at both 39 regional and global levels. As such, the HTAP\_v3 mosaic inventory provides a complement to 40 41 globally consistent emission inventories such as EDGAR. The global and regional trends of air pollutant emissions in the HTAP v3 mosaic are comparable with other commonly available 42

43 global emission datasets.

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1	By providing consistent times series for almost two decades, HTAP v3 allows an evaluation-		Formatted: Justified
2 3 4 5	of the impact and success of the pollution control measures deployed across various regions of the world since 2000. Similarly, its finer sectoral resolution is suitable for understanding how- and where technological changes have resulted in emissions reductions, suggesting possible pathways for strengthening appropriate policy actions.		Formatted: Font: 12 pt, Font color: Auto
6 7 8 9 10	All these features make HTAP_v3 a database of interest for policy makers active in the air quality regulatory efforts. HTAP_v3 provides a picture of a world where most pollutant emissions are following a steady or decreasing path. However, several areas of the world show an increasing emission trend, with wide portions of the world remaining subjected to unsatisfactory levels of ambient air quality.		Formatted: Font: 12 pt
11 12 13	When using the HTAP_v3 emission mosaic, users should consider the following limitations, for example when combining the HTAP_v3 data with other emission input needed to run atmospheric models:	_	Formatted: Font: 12 pt, Font color: Auto
14 15	- agricultural waste burning emissions should be treated with caution to avoid double-counting when combined with existing biomass burning emission inventories;		
16 17 18	- NMVOC and NOx emissions from agricultural soils should be treated with caution to avoid double-counting when combining the HTAP_v3 data with a natural emissions model such as MEGAN (Model of Emissions of Gases and Aerosols from Nature);		
19 20 21 22 23 24 25 26	- the speciation of NOx emissions into its components (NO, NO2, HONO) is not provided by the global HTAP_v3 mosaic and it is beyond the scope of the current work since the regional inventories report total NOx with no speciation. Standard practice in global models is to emit all anthropogenic NOx as NO, while we expect that regional modelling groups will have access to appropriate best practices for their particular regions. In particular for road transport, the partitioning of NOx emissions between NO, NO2, and HONO is highly region-dependent and it is based on the fleet composition (e.g., number of diesel vehicles relative to gasoline vehicles) and technology level (e.g., the level of exhaust after treatment).		
27 28 29 30 31 32 33	Thanks to the continuous improvement of local and regional emission inventories, recent literature shows new datasets that report regional information over areas of the world not covered by local inventories in the current HTAP_v3 mosaic (e.g. Argentina (Puliafito et al. 2021), Africa (Keita et al., 2021) and the MEIC inventory (http://meicmodel.org.cn/?page_id=1772⟨=en)). Future updates to this mosaic may also integrate reliable and up to data information over South America or Africa as time and resources permit.		Formatted: Font: 12 pt Formatted: Font: 12 pt, Font color: Auto
34 35 36 37 38	Similar to its predecessor (e.g. HTAP_v2.2 mosaic inventory), we expect that this new HTAP_v3 mosaic inventory will be used as a basis for global assessments of long-range, transboundary transport of air pollution under the Task Force on Hemispheric Transport of Air Pollution, while also providing a convenient and useful information for regional modellers seeking the best available regional emissions with a consistent gap-filling methodology.		Formatted: Font: 12 pt

# 40 Author contributions.

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MC and DG developed the mosaic gathering input from all data providers. The co-chairs of the
 TF-HTAP (TK, TB, RW and JaKa) fostered the dialogue with international institutions

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1 2 3 4 5 6 7	contributing to this work with their data. PM, RM, JR, JZ, DN, MS, MDM, RW provided data for Canada, JuKu, SC, TM provided data for Japan, JeKu provided data for Europe, J-HW, JK provided data for Korea, TK, GP provided data for USA, JiKi provided data for Asia. The JRC EDGAR group (MC, ES, DG, EP, MM, FM, ES, MB, FP) lead the drafting of the publication with input from colleagues contributing to the HTAP_v3 mosaic, SJS and HS performed detailed data comparison among available emission inventories. TA calculated and provided the NMVOC speciation fractions for all the sectors for the four regions.		Formatted: Font: Times New Roman, 12 pt Formatted: Font: 12 pt Formatted: Font: Times New Roman, 12 pt
8	·	(	Formatted: Font: 12 pt
9 10	<b>Competing interests.</b> The authors declare that they have no conflicts of interest nor competing interests.		
11	Acknowledgements.		
12 13 14	The authors would like to thank all HTAP_v3 data providers for the fruitful cooperation. The views expressed in this publication are those of the authors and do not necessarily reflect the views or policies of the European Commission.		
15 16 17 18 19 20	J-STREAM emission inventory for Japan was developed by Environment Research and Technology Development Fund (JPMEERF20165001 and JPMEERF20215005) of the Environmental Restoration and Conservation Agency Provided by the Ministry of Environment of Japan, and FRIEND (Fine Particle Research Initiative in East Asia Considering National Differences) Project through the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (2020M3G1A1114622).		
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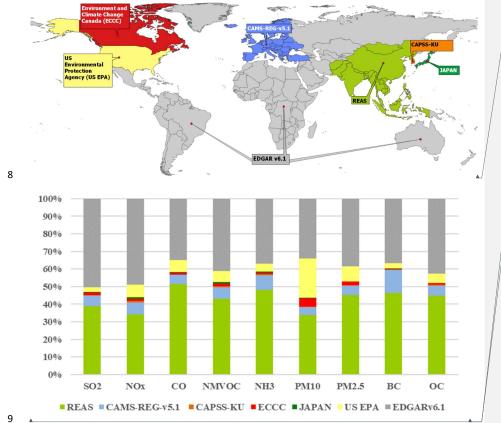
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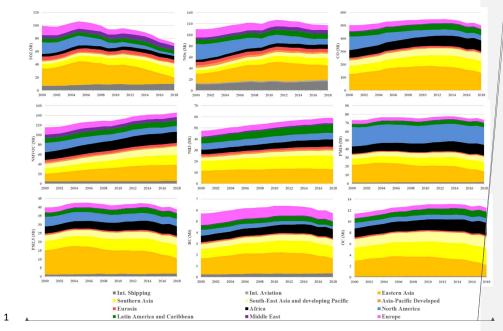
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Figure 1 – Overview of the HTAP\_v3 mosaic data providers. Data from officially
reported emission gridmaps were collected from the US Environmental Protection
Agency, Environment and Climate Change Canada, CAMS-REG-v5.1 for Europe,
REASv3.2.1 for most of the Asian domain, CAPSS-KU for Korea and JAPAN (PM2.5EI
and J-STREAM) for Japan. The share of the total emissions covered by each data
provider is reported in the bar chart at the bottom.

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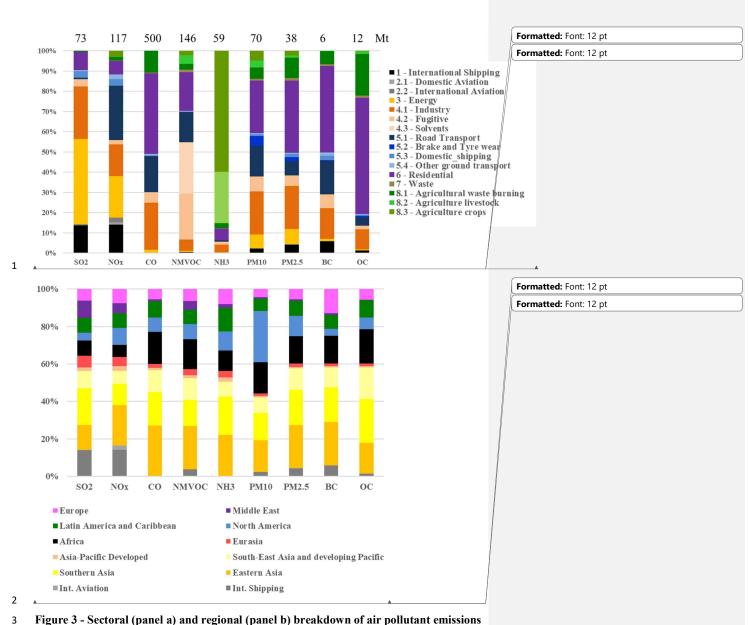
2 Figure 2 – Time series of gaseous and particulate matter pollutants from HTAP\_v3 by

3 aggregated regions. Regional grouping follows the Intergovernmental Panel on Climate

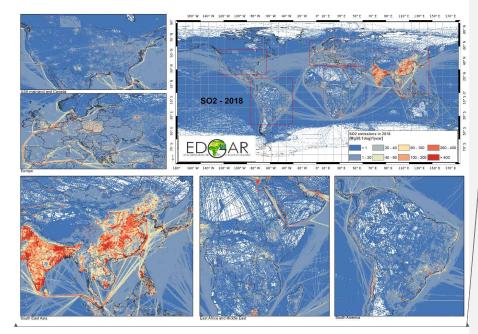
4 Change Sixth Assessment Report (IPCC AR6) definitions. Table S3 provides information

5 on the regional belonging of each country to the IPCC AR6 regions.

6 7



- Figure 3 Sectoral (panel a) and regional (panel b) breakdown of air pollutant emissions from HTAP\_v3 for the year 2018. At the top of each bar in panel a, total emissions for
- 4 from HTAP\_v3 for the year 2018. A
  5 each pollutant are reported (in Mt).



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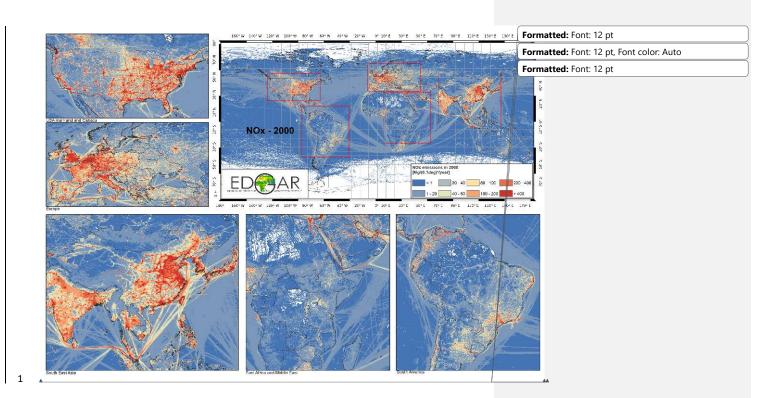
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Figure 4 – HTAP\_v3 mosaic: SO<sub>2</sub> emission gridmaps for the year 2018.

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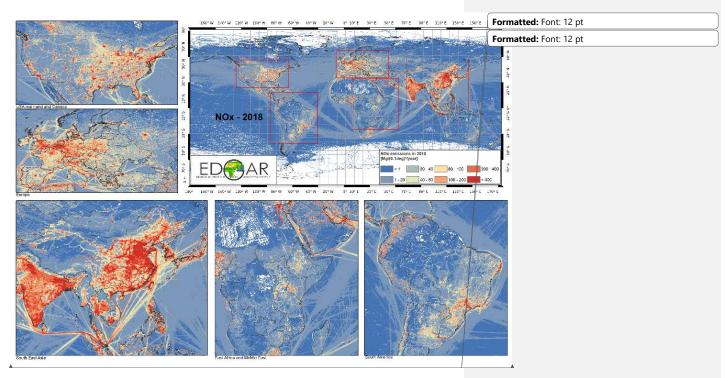
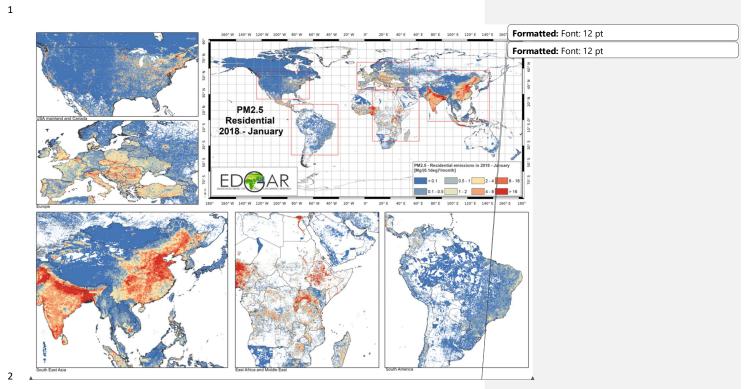
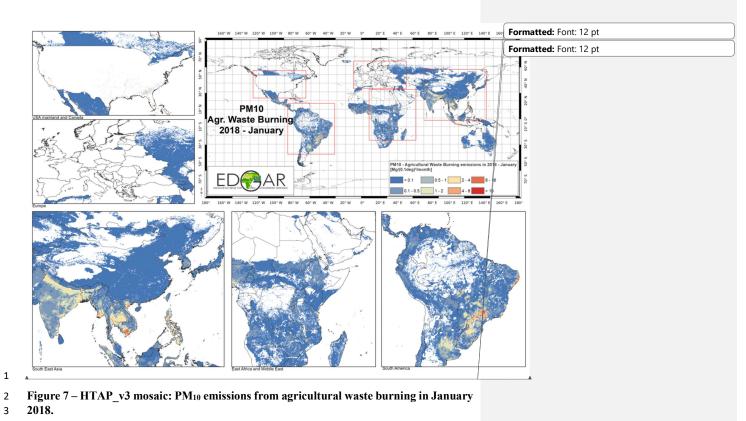


Figure 5 – HTAP\_v3 mosaic: NOx emission gridmaps in 2000 (top panel) and 2018 (bottom panel). 3



3 Figure 6 – HTAP\_v3 mosaic: PM<sub>2.5</sub> emissions from residential activities in January 2018.



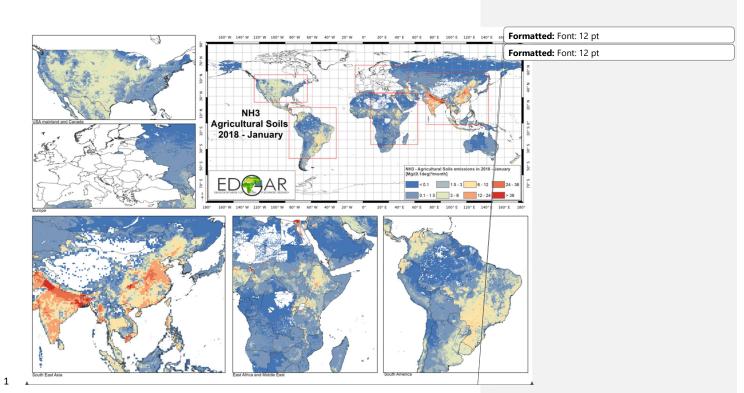
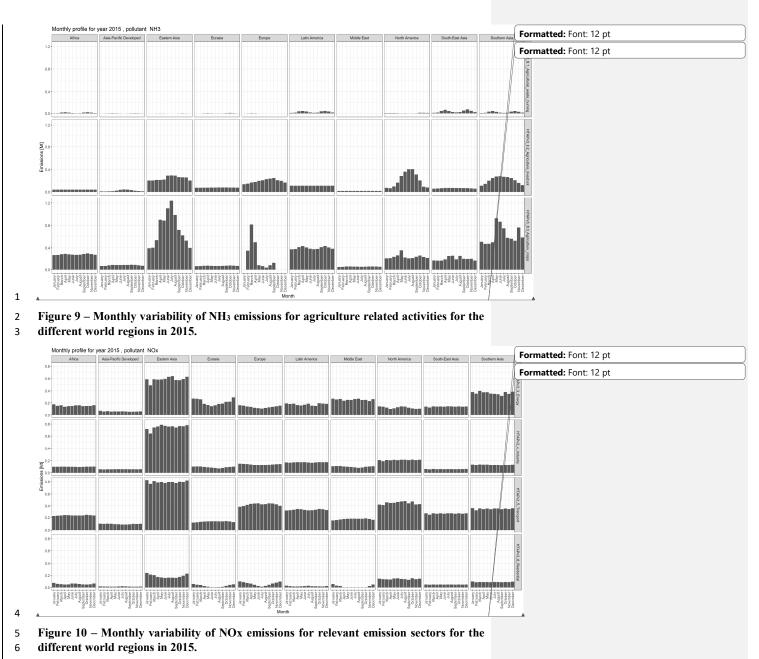
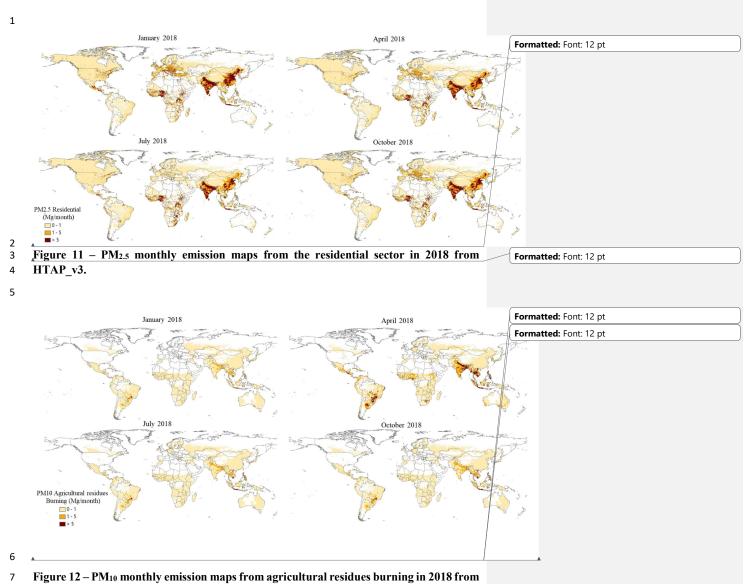
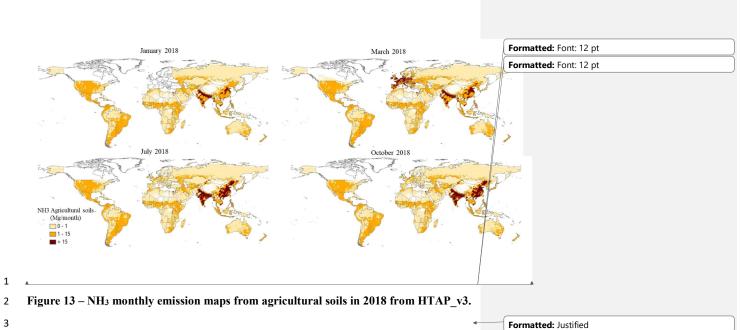


Figure 8 – HTAP\_v3 mosaic: NH<sub>3</sub> emissions from agricultural soils activities in January 2018.





8 HTAP\_v3.



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# Table 1 – Overview of data input to the HTAP\_v3 emission mosaic.

Data	CAMS-REG-	US EPA	ECCC	REASv3.2.1	 	Formatted Table
source	v5.1					
Type of	Country	Country inventory	Country inventory	Country		Formatted: Font: 12 pt
data	inventories as		as emission time	inventories as		
source	emission time		series by sector	emission		
	series by sector		and country and	gridmaps as text		
	and country and		emission	files.		
	emission gridmaps		gridmaps as			
	as .csv files.		.NetCDF files.			
Sectors	All sectors,	All sectors,	All sectors,	All sectors,		Formatted: Font: 12 pt
covera	excluding	excluding	excluding	excluding brake		
ge	international	international	agricultural waste	and tyre wear,		
	shipping and	shipping and	burning,	domestic		
	aviation	aviation	international	shipping, waste,		
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	domestic).	domestic).	aviation	burning,		
			(international and			
			domestic).	shipping and		
				aviation		
				(international and		
				domestic).		

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ral				from MEIC over		· · ·	
covera				China for 2016,			
ge				2017, 2018			
Tempo	Annual emission	Monthly emission	Monthly emission	Monthly emission		Formatted: Font: 12 pt	
ral resoluti	gridmaps + monthly profiles	gridmaps	gridmaps	gridmaps			
on Control	0.10.0.10	0.10.0.10	0.10.0.10	0.10.0.10 (TT)			
Spatial resoluti	0.1°x0.1°	0.1°x0.1°	0.1°x0.1°	0.1°x0.1° (The	$\sim$	Formatted: Font: 12 pt	
on				original spatial resolution of		Formatted: Justified	
				REASv3.2.1 is			
				0.25°x0.25°.			
				Assuming that			
				emissions are			
				equally distributed			
				in the $0.25^{\circ}$ cell,			
				REASv3.2.1 data			
				were converted to			
				0.1° cell and provided to			
				provided to HTAP v3)			
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nces	NMVOC, NH3,	NMVOC, NH3,	NMVOC, NH3,	NMVOC, NH3,			
	PM10, PM2.5,	PM10, PM2.5,	PM10, PM2.5,				
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Substanc	SO2, NOx, CO,	SO2, NOx, CO,				Formatted: Font: 12 pt
es	NMVOC, NH3, PM10, PM2.5, BC, OC	NMVOC, NH3, PM10, PM2.5, BC, OC	NH3, PM10, PM2.5, BC, OC			
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HTAP_v	HTAP_v3	Sector description	IPCC 1996	IPCC 2006	
3 main sectors	detailed sectors		codes	codes	
HTAP 1	HTAP 1:	International water-born	1C2	1.A.3.d.i	 Formatted: Font: 12 pt, Font color: Auto
: Internati onal Shipping	International Shipping	navigation.			
HTAP 2	HTAP_2.1:	Civil Aviation.	1A3aii	1.A.3.a.ii	 Formatted: Font: 12 pt, Font color: Auto
: Aviation	Domestic Aviation		1.4.2 .	1 4 2 .	
	HTAP_2.2: International Aviation	International Aviation.	1A3ai	1.A.3.a.i	
HTAP_3	HTAP_3:	Power generation.	1A1a	1.A.1.a	Formatted: Font: 12 pt, Font color: Auto
: Energy	Energy	_			·
HTAP_4	HTAP_4.1:	Industrial non-power large-	1A2 + 2 +	1A2 + 2	Formatted: Font: 12 pt, Font color: Auto
: Industry	Industry	scale combustion emissions and emissions of industrial processes. It includes: manufacturing, mining, metal, cement, chemical and fossil fuel fires.	5B	(excluding 2.D.3 + 2.E + 2.F + 2.G) + 7A	
	HTAP_4.2: Fugitive	It includes oil and gas exploration and production and transmission, including evaporative emissions (mainly NMVOC).	1B + 1A1b + 1A1ci + 1A1cii + 1A5biii	1.B + 1.A.1.b + 1.A.1.c.i + 1.A.1.c.i.i + 1.A.5.b.i.i.i	
	HTAP_4.3: Solvents	Solvents and product use.	3	2D3 + 2E + 2F + 2G	
HTAP 5	HTAP 5.1:	Road Transport, combustion	1A3b	1.A.3.b	 Formatted: Font: 12 pt, Font color: Auto
: Ground Transpo	Road Transport	and evaporative emissions only.	(excluding resuspensio n)	(excluding resuspension )	
rt	HTAP_5.2: Brake and Tyre wear	Re-suspended dust from pavements or tyre and brake wear from road transport.	1A3b (resuspensi on only)	1.A.3.b (resuspensio n only)	
	HTAP_5.3: Domestic shipping	Domestic shipping: inland waterways + domestic shipping.	1A3d2	1.A.3.d.ii	
	HTAP_5.4: Other ground transport	Ground transport by pipelines and other ground transport of mobile machinery.	1A3c + 1A3e +	1.A.3.c + 1.A.3.e.ii	
HTAP_6	HTAP_6:	Small-scale combustion,	1A4 + 1A5	1.A.4 +	 Formatted: Font: 12 pt, Font color: Auto
: Resident ial	Residential	including heating, cooling, lighting, cooking and auxiliary engines, to equip residential,		1.A.5	

		commercial buildings, service institutes, and agricultural facilities and fisheries.					
HTAP_7	HTAP_7:	Solid waste disposal and	6	4			Formatted: Font: 12 pt, Font color: Auto
: Waste	Waste	wastewater treatment.					
HTAP_8	HTAP_8.1:	Agricultural waste burning	4F	3.C.1.b			Formatted: Font: 12 pt, Font color: Auto
:	Agricultural	(excluding Savannah burning).					·
Agricult	waste burning						
ure	HTAP_8.2:	Livestock emissions, including	4B	3.A.2			
	Agriculture	manure management.					
	livestock	_					
	HTAP_8.3:	Emissions from crops,	4C + 4D	3.C.2	+		
	Agriculture	fertilisers, and all agricultural		3.C.3	+		
	crops	soils activities.		3.C.4	+		
	-			3.C.7			
					4	$\times$	Formatted: Font: 12 pt
							Formatted: Justified

4 Table 3 - Overview of pollutant and sector provided by each inventory in HTAP\_v3. Cells 5 with N/A indicate that the emissions for those sectors were not provided and/or used in HTAP\_v3 for a specific inventory, while gapfilled with the corresponding information 6 7 from EDGARv6.1. The other cells represent the data availability for each sector and inventory. The color codes used for the pollutants refer to the data source: black color 8 9 represents pollutant emissions provided by a specific inventory, red color indicates emissions gapfilled using EDGARv6.1 and violet color indicates combinations of sectors-10 pollutants available for specific regional inventories but not in EDGAR, which typically 11 12 represent minor sources of emissions included in officially reported inventories. These minor sources are included in the HTAP\_v3 mosaic. 13

## 14

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	REA	CAP				CAMS-		Ĩ	
	Sv3.2	SS-				REG-	EDGAR		
Data provider	.1	KU	JAPAN	ECCC	US EPA	v5.1	v6.1	•	Formatted: Font: 12 pt, Font color: Auto
HTAP_1:							All		Formatted: Justified
International							substanc		Formatted: Font: 12 pt, Font color: Auto
Shipping	N/A	N/A	N/A	N/A	N/A	N/A	es		Formatted: Font. 12 pt, Font color. Auto
HTAP_2.1:							All	•	Formatted: Justified
Domestic							substanc		Formatted: Font: 12 pt, Font color: Auto
Aviation	N/A	N/A	N/A	N/A	N/A	N/A	es		Formatted: Justified
HTAP_2.2:							All	*	
International							substanc		Formatted: Font: 12 pt, Font color: Auto
Aviation	N/A	N/A	N/A	N/A	N/A	N/A	es		Formatted: Justified
HTAD 2	All	All	BC, OC,	All	All	All	All		
<u>HTAP_3:</u>	subst	subst	NOx,	substanc	substanc	substanc	substanc	$\sim$	Formatted: Font: 12 pt, Font color: Auto
Energy	ances	ances	NH3,	es	es	es	es		Formatted: Justified

			CO, PM2.5, PM10, NMVO						
			C, SO2						
			BC, OC, NOx, NH3,						
<u>HTAP_4.1:</u>			CO,					*~	Formatted: Font: 12 pt, Font color: Auto
Industry	All subst	All subst	PM2.5, PM10, NMVO	All substanc	All substanc	All substanc	All substanc		Formatted: Justified
	ances	ances	C, SO2	es	es	es	es		
		BC, OC, NOx, NH3, CO,	BC, OC,						
<u>HTAP_4.2:</u>		PM2.	NOx,					≛~<	Formatted: Font: 12 pt, Font color: Auto
Fugitive		5, PM10	NH3, CO, PM2.5,						Formatted: Justified
	All subst	NMV OC,	PM10, NMVO	All substanc	All substanc	All substanc	All substanc		
	ances	so2	C, SO2	es	es	es	es		
	NMV OC,	NMV OC,			CO, NOx, OC,	NOx, NH3,			
<u>HTAP_4.3:</u>	NH3,	NH3,	NIMUO	NIMUO	NMVOC	CO,		$\sim$	Formatted: Font: 12 pt, Font color: Auto
Solvents	PM10	PM10	NMVO C, NH3,		, NH3, PM10,	PM2.5, PM10,	All		Formatted: Justified
	PM2. 5	PM2. 5	PM10, PM2.5	PM10, PM2.5	PM2.5, SO2	NMVO C, SO2	substanc		
	All	All	All	All	All	All	es All	1	
HTAP 5.1:	subst	subst	substanc	substanc	substanc	substanc	substanc	*><	Formatted: Font: 12 pt, Font color: Auto
Road Transport	ances	ances	es	es	es	es	es		Formatted: Justified
		BC,							
		OC,							
		NOx,							
		NH3,							
HTAP_5.2:		CO, DM2						•>	Formatted: Font: 12 pt, Font color: Auto
Brake and Tyre		PM2. 5,							Formatted: Justified
wear		PM10							
					BC,OC,				
		, NMV OC,	BC,OC, PM2.5,P	BC,OC, PM2.5,P	PM2.5,P M10,	BC,OC, PM2.5,P	BC,OC, PM2.5,P		
	N/A	SO2	M10	M10	NMVOC	M10	M10		

						BC, OC, NOx,			
НТАР 5.3:						NH3,		~	Formatted: Font: 12 pt, Font color: Auto
Domestic						CO,			Formatted: Justified
shipping		All		All	All	PM2.5, PM10,	All		Formatted. Justined
		subst		substanc	substanc	NMVO	substanc		
	N/A	ances	N/A	es	es	C, SO2	es		
	10/1	unces	BC, OC,	0.5	0.5	0,002	0.5		
			NOx,						
HTAP 5.4:			NH3,						
Other ground			СО,					$\geq$	Formatted: Font: 12 pt, Font color: Auto
transport			PM2.5,						Formatted: Justified
	All	All	PM10,	All	All	All	All		
	subst	subst	NMVO	substanc	substanc	substanc	substanc		
	ances	ances	C, SO2	es	es	es	es		
			BC, OC, NOx,						
			NH3,						
НТАР 6:			CO.					• _	Formatted: Font: 12 pt, Font color: Auto
Residential			PM2.5,						Formatted: Justified
	All	All	PM10,	All	All	All	All		Formatted: Justified
	subst	subst	NMVO	substanc	substanc	substanc	substanc		
	ances	ances	C, SO2	es	es	es	es		
		All	All	All	All	All	All		
HTAP_7: Waste	<b>N</b> T/ A	subst	substanc	substanc	substanc	substanc	substanc	$\sim$	Formatted: Font: 12 pt, Font color: Auto
UTAD 01.	N/A	ances All	es All	es	es All	es All	es All	_	Formatted: Justified
HTAP_8.1: Agricultural		subst	substanc		substanc	substanc	substanc		<b>Formatted:</b> Font: 12 pt, Font color: Auto
waste burning	N/A	ances	es	N/A	es	es	es		Formatted: Justified
haste saining	1011	NH3,		1 () 1 1					
		NMV							
	NH3,	OC,							
HTAP 8.2:	NMV	NOx,							Formatted: Font: 12 pt, Font color: Auto
Agriculture	OC,	PM10		NH3,	NH3,	NH3,		$\overline{}$	
livestock	NOx,	, ,	NH3,	NMVO	NMVOC	NMVO	NH3,		Formatted: Justified
	PM10	PM2.	NMVO	C, NOx,	, NOx,	C, NOx,	NMVO		
	, РМ2.	5, BC,	C, NOx, PM10,	PM10, PM2.5,	PM10, PM2.5,	PM10, PM2.5,	C, NOx, PM10,		
	PM2.	OC	PM10, PM2.5	BC, OC	BC, OC	OC	PM10, PM2.5		
	5	NH3,	1 1112.3	50,00	NH3,	NH3,	1 1112.3		
		NOx,		NH3,	NOx,	NOx,			
	NH3,	PM10		NOx,	PM10,	PM10,			
HTAP_8.3:	NOx,	,		PM10,	PM2.5,	PM2.5,		$\sim$	Formatted: Font: 12 pt, Font color: Auto
Agriculture_cro	PM10	PM2.	NH3,	PM2.5,	BC, OC,	SO2,	NH3,		Formatted: Justified
ps	,	5,	NOx,	BC,	CO,	CO, OC,	NOx,		
	PM2.	BC,	PM10,	NMVO	NMVOC	NMVO	PM10,		
			10110 5	C OC	000	C	10110 5		
	5	OC	PM2.5	C, OC	, SO2	C	PM2.5		Formatted: Font: 12 pt

#### Table 4 - Main features of the different HTAP mosaics. HTAP\_v2.2 HTAP\_v3 HTAP\_v1 Formatted: Font: 12 pt, Font color: Auto 2008 and 2010 Time coverage 2000-2005 2000-2018 Formatted: Justified yearly and Formatted: Font: 12 pt, Font color: Auto Time resolution yearly and monthly monhtly yearly Formatted: Justified CO, SO<sub>2</sub>, NOx, CO, $SO_2$ , NOx, CH<sub>4</sub>, NMVOC, NMVOC, NH<sub>3</sub> (only for NMVOC, NH<sub>3</sub>, CO. Formatted: Font: 12 pt, Font color: Auto SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, PM<sub>10</sub>, agriculture), PM<sub>10</sub>, PM10, PM<sub>2.5</sub>, Formatted: Justified PM<sub>2.5</sub>, BC, OC PM<sub>2.5</sub>, BC, OC BC, OC Substances Formatted: Font: 12 pt, Font color: Auto International Formatted: Justified Shipping, Domestic Shipping, Domestic Aviation, International Aviation, Energy, Industry, Fugitives, Use, Solvent Road Transport, Brake and Tyre Other Wear, Ground Transport, Residential, Aircraft, Ships, Energy, Waste, Industry Agricultural Processes, Energy, Ground Transport, Air, Ships, Waste Solvents, Transport, Residential, Industry, Burning, (including Livestock, and Agriculture, Agriculture Residential Waste Burning, waste), and Agriculture Agricultural and Sectors Waste (only for NH3) Crops Formatted: Font: 12 pt, Font color: Auto Geographical Formatted: Justified coverage Globe Globe Globe Formatted: Font: 12 pt, Font color: Auto 0.1°x0.1° Spatial resolution 0.1°x0.1° 0.1°x0.1° Formatted: Justified CAMS-REGv5.1. Formatted: Font: 12 pt, Font color: Auto REASv3.2.1, Formatted: Justified US EPA, US EPA, Environment ECCC, Canada, MICS, CAPSS-KU, TNO/EMEP JAPAN Europe

(MACC II), MICS Asia

REAS2.1,

(PM2.5EI and

J-STREAM),

EDGARv6.1

REAS,

III+

EDGARv4.3

UNFCCC,

EDGARv4.1

Input datasets

GAINS, EMEP, EPA,

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1	Reference	Janssens-Maenhout et al., 2012	Janssens-Maenhout al., 2015	et     This work	Formatted: Font: 12 pt, Font color: Auto Formatted: Justified
					Formatted: Font: 12 pt