

Public justification (visible to the public if the article is accepted and published):

I understand challenges around uncertainties; good on you for added Table S6 and for new section 3.5. But, back to my original question. If SO₂ comparisons (EDGAR-HTAP) carry variability per sector ranging from 5 to 80%, and if - in most cases - those variabilities prove much larger for 2018 emissions compared to 2000 emissions, with what confidence can one conclude a temporal trend (decrease, 2000 to 2018) for SO₂ of global 100 vs 73? Likewise but perhaps worse (lower confidence) for global NO_x increase? Authors can decide what, if anything, they want to do, at proof stage. They have certainly not convinced this reader; other readers more familiar with pollutant emissions data may take different views.

We acknowledge the additional comment of the Reviewer and we take the opportunity to further clarify how to interpret the emission trends both rephrasing some sentences in the text and expanding the uncertainty section of the paper.

We added the following sentence to Section 3.1 just before the description of the trends by pollutant:

‘In the following paragraphs we shortly present global and regional air pollutant emissions and their trends over the 2000-2018 period as provided by the HTAP_v3 data. Emissions are not presented with a confidence level since no comprehensive bottom-up uncertainty analysis has been performed in the context of the mosaic compilation, however see discussion in section 3.5.’

Moreover we further developed section 3.5 on emission uncertainty to clarify the doubts of the Editor and we believe that thanks to these changes and explanations the paper has strongly improved.

‘3.5 Emission Uncertainties

3.5.1 Overview on uncertainties

Unlike greenhouse gas inventories, uncertainty is not routinely estimated for air pollutant emissions by country inventory systems. In part this is due to the different and often disparate processes used to generate air pollution data at the country level (Smith et al., 2022), making it more difficult to conduct uncertainty analysis. While combinations of observational and modelling techniques can be used to evaluate air pollutant emissions, these are inherently site specific and can be difficult to generalize.

The potential level of uncertainty in any emission estimate depends on how much emission factors vary for a particular activity. We note that the emission species with the lowest uncertainty is carbon dioxide from fossil fuel combustion. This is because CO₂ emission factors are closely tied to fuel energy content, which is a quantity that is tracked and reported by both government and commercial reporting systems. Similar considerations apply to SO₂ emissions, where emissions can be reliably estimated if the sulphur content of fuels and the operational characteristics of emission control devices are known. A key aspect here is that uncertainty in fuel sulphur content is largely uncorrelated across regions, which means that global uncertainty is relatively low, while regional uncertainty is often much higher (Smith et al., 2011). On the opposite end of the spectrum, the emission rates for particulate matter depend sensitively on combustion conditions and the operation of any emission control devices and can vary over

several orders of magnitude. While this is not an indication of the uncertainty in inventory estimates, this indicates the difficulty of constructing quantitative uncertainty estimates. The type of emission process also influences uncertainty, with fugitive emissions and emissions associated with biological processes generally having higher uncertainty levels.

We note also that uncertainty in the overall magnitude of emissions does not necessarily imply a similar level of uncertainty in relative emission trends. Even with uncertainties, the widespread use of emission control devices has resulted in reductions in air pollutant emissions in North America and Europe (Liu et al., 2018; Jamali et al., 2020), as verified by observational and modelling studies.

The emissions in the HTAP_v3 mosaic emissions originate from a variety of sources which has some implications for relative uncertainty. Emissions for some regions, such as North America and Europe, were generated by country inventory systems which have been developed and refined over the last several decades. It is reasonable to assume these emissions are robust, however even in these regions detailed studies have indicated that actual emissions in some cases appear to be lower than inventory values (Anderson et al., 2014; Hassler et al., 2016; Travis et al., 2016). Where EDGAR emission estimates were used in the mosaic uncertainties are likely be higher overall given that inventory information developed in those countries was not available for these regions (Solazzo et al., 2021).

Some information on the robustness of the HTAP_v3 mosaic can be gained by comparing different inventory estimates, which is shown in supplement section S2. In many cases, the agreement between estimates (for example in North America and Europe) simply indicates common data sources and assumptions, although this does indicate that the different inventory groups did conclude that these values were plausible. The larger differences in other regions, however, does point to larger uncertainty there.

3.5.2 Qualitative assessment of the uncertainty of a global emission mosaic

Assessing the uncertainty of a global emission mosaic is challenging since it consists of several 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 which is documented, where available, by the corresponding literature describing each dataset (see Table 2 and section 2.3). However, the mosaic compilation process may also introduce additional uncertainties compared to the input datasets. In order to limit these additional uncertainties, we made the following considerations:

-for each emission inventory both the national totals and gridded data by sector were gathered. This process allows the mosaic compilers not to introduce additional uncertainty compared to the original input regional datasets. While additional uncertainties may arise from the 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), this is not the case in the current dataset. Therefore, when regional trends are described by region and pollutant (see section 3), no additional source of uncertainty has to be considered from the mosaic compilation approach.

-the sector definition and mapping has been developed following the IPCC categories and when no data was available for a certain combination of sector and pollutant a gapfilling procedure is applied using the EDGAR database. Therefore, the datasets are comparable in terms of sectoral coverage, which reduces uncertainties in this aspect.

- since each inventory provided monthly resolution emission gridmaps and time series there is no additional uncertainty introduced by temporal disaggregation as part of the construction of the HTAP_v3 mosaic.

In this work we also provide a qualitative indication of the emission variability by HTAP sector and pollutant at the global level. Table S6 summarises the variability of global HTAP_v3 emissions by sector for the boundary years of this mosaic (2000 and 2018) compared to the global EDGARv6.1 data. EDGAR emissions are considered as the reference global emission 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 difference of the estimates of the two inventories, i.e. $(\text{EDGARv6.1} - \text{HTAP_v3}) / \text{HTAP_v3}$. 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 EMEP/EEA Guidebook (2019) information. The largest variability is found domestic shipping 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 variability can be used as proxy of structural uncertainty, keeping in mind that variability could 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 updates.'