

Multiscale observations of NH₃ around Toronto, Canada, Yamanouchi et al., AMT, 2020

General Description:

The authors intercompare NH₃ concentrations obtained as column densities from ground-based and space-based instruments, surface concentrations from a monitoring site, and 3D mixing ratios from the GEOS-Chem model in and around Toronto, Canada. These they use to determine long-term trends in NH₃ concentrations and assess consistency across the data platforms. The manuscript is in general well written and easy to follow, but requires additional details about the model setup and information about sulfate and nitrate in and around Toronto for interpretation of the NH₃ concentration trends. These and other comments are provided below.

General Comments:

Why was the nested version of GEOS-Chem over North America not used? It includes Toronto in the domain and is at finer resolution ($0.25^\circ \times 0.3125^\circ$) than the global domain.

There is quite a lot of information relevant to model representation of NH₃ that is missing in the model description section. These include the following: The inventories used in the model to represent US and Canadian SO₂ and NO_x sources that form sulfate and nitrate that influence NH₃ uptake to aerosols. The version of EDGAR and whether this is the inventory that represents anthropogenic NH₃ emissions over the domain of interest or whether it is a combination of EDGAR and GEIA (now quite outdated and only really used in the model to represent natural NH₃ emissions). The base year of each inventory. Whether annual scaling factors are applied to any of the emissions that would have declined due to emission regulations (typically NO_x and SO₂). Whether seasonal scaling factors are applied to NH₃ emissions in the model.

The model also seems to be underutilised to provide context for the study region. The inventories could, for example, be used to assess the relative proportion of vehicular, agricultural, and natural emissions to total NH₃ emissions and to determine the role of changes in sulfate and nitrate (due to emission regulations of SO₂ and NO_x sources) on observed trends in NH₃.

What is the fit that is applied to the data to obtain the trends? And what is the determination of significance? It is stated in the text that “The number of years of measurements needed for the trend to be statistically (2σ) significant was found to be 33.8 years and 29.3 years” (p. 6, lines 177-178), but it is not clear why this is the case given that the 2σ uncertainty is much less than the trend value. An explicit statement of what the authors use as a significance criterion might help avoid confusion.

The FTIR instrument and measurements are referred to in figures/tables/text as FTIR, TAO, or TAO FTIR. To avoid confusion, stick with one of these throughout.

Specific Comments:

p1, line 14: There is no context for the use of “resampling” in the abstract to be able to follow what this implies for the results obtained. What is being resampled? And why does it alter the correlation?

p2, line 38: Briefly elaborate on the link between NH_3 concentrations and SO_2 and NO_x emissions.

p2, line 39: “...as well as by reactions with acids in the atmosphere” sounds like it is happening in the gas phase. Make clear that this is a heterogeneous process.

p3, line 59: What is the NH_3 source from greenery? Application of fertiliser to gardens and public spaces?

p4, line 97: What is the shape of the a priori profile used for the retrieval? How does it compare to that from GEOS-Chem?

p4, line 121: Odd to express the swath like this. Standard is as 2200 km.

p5, line 137-138: Say what model years are sampled after the one year spin up.

p5, line 145-147: This approach is reasonable and widespread, but what if the spatial extent is less than the spatial resolution of IASI (at best 12 km at nadir), as seems to be the case in this work?

Figure 2: Does the seasonality differ if the median is calculated for each month?

Figure 2: Consider showing the y-axis as $1\text{e}16$ rather than $1\text{e}17$.

p7, line 189: Why is the seasonality solely attributed to emissions? What about partitioning of NH_3 to acidic aerosols? Is there any seasonality to this process?

Table 1: Is there a reason that this table is included if this information is already illustrated in Figure 2?

Table 2: The layout of the table is confusing, as the row labels correspond to specific time periods, but then the final column is labelled “during the same timeframe”. What is this timeframe then? Why is the FTIR TAO trend for this same timeframe not given?

Figure 4: The lines in (a) are not easy to see. Consider making these thicker.

p12, line 248: Tournadre et al. (2020) is not cited correctly.

p12, line 254: What is “simple linear regression”? Ordinary least squares?

p12, line 259-260: It’s not clear what this means: “Without temporal resampling, no significant correlation was found ($r \leq 0.27$) for any spatial coincidence criteria”. What is this temporal resampling and why does it impact the correlation?

Table 3: The information as presented in this table is okay, but would have been more visually interesting and easier to identify patterns in the data if each variable (r, slope etc.) was illustrated on 2D colored grids.

p13, line 267: What does this gridbox include other than Toronto that might dilute or increase NH_3 concentrations and affect the comparison?

Figure 7: It would be helpful to say in the caption or text what this is showing from Table 3.

Figure 8: It is not easy to discern the red and black points in panel (b).

Figure 9: Are units for GEOS-Chem in panel (b) correct?