Supplement of Atmos. Chem. Phys., 22, 12907–12922, 2022
https://doi.org/10.5194/acp-22-12907-2022-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

NH_3 spatiotemporal variability over Paris, Mexico City, and Toronto, and its link to $PM_{2.5}$ during pollution events

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Figure S1: Monthly means of NH_3 total columns (molecules/cm²) derived from 10 years (2008-2017) of IASI NH_3-retrieved columns over the so-called Europe domain. The blue cross indicates Paris location.

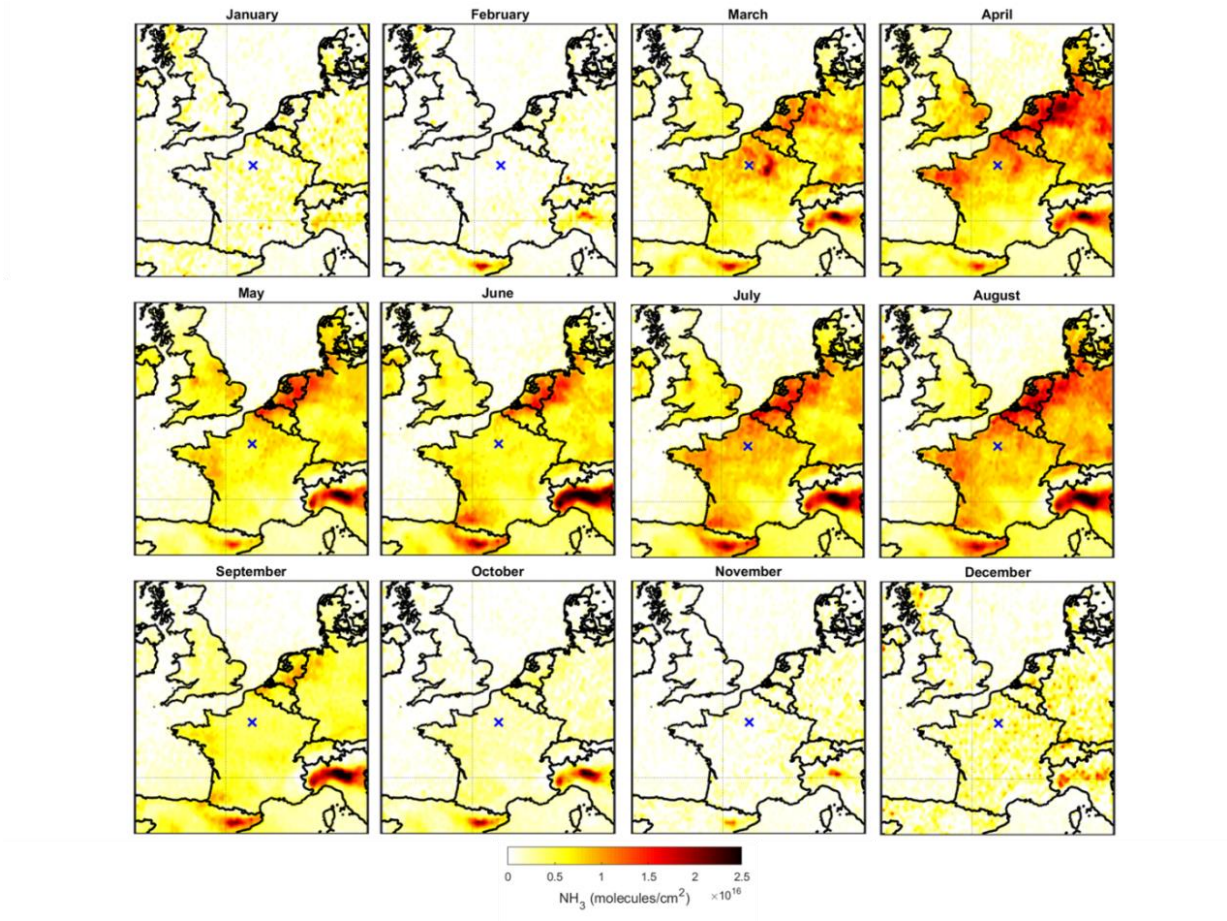


Figure S2: Same than figure S1 but for the North America region.

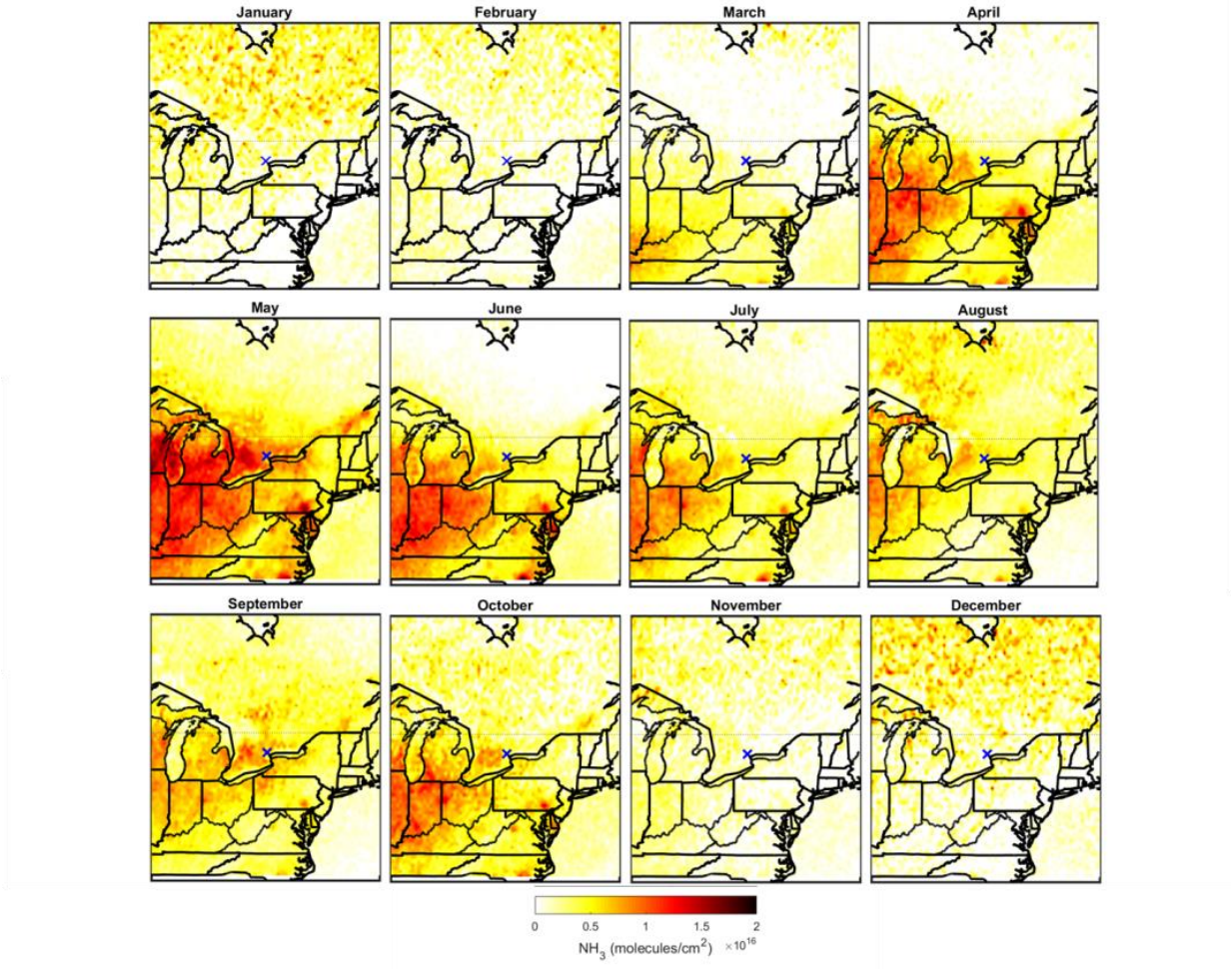


Figure S3: Same than figure S1 but for the southern North America region.

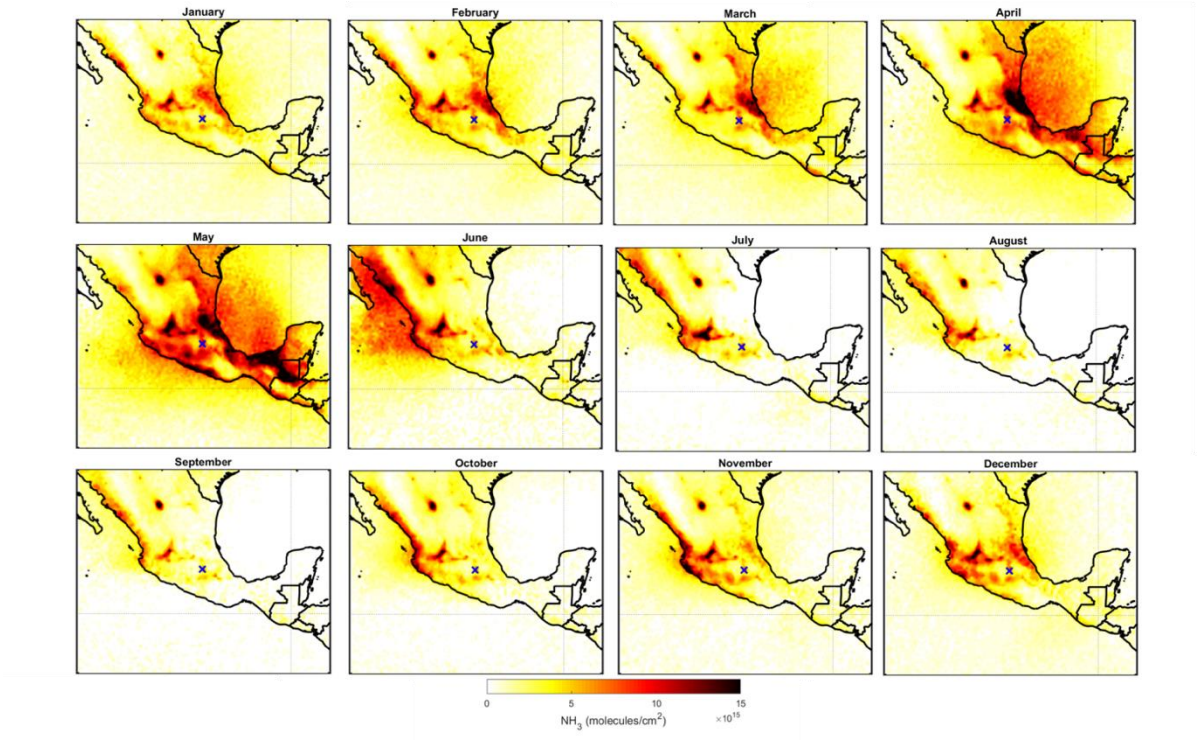
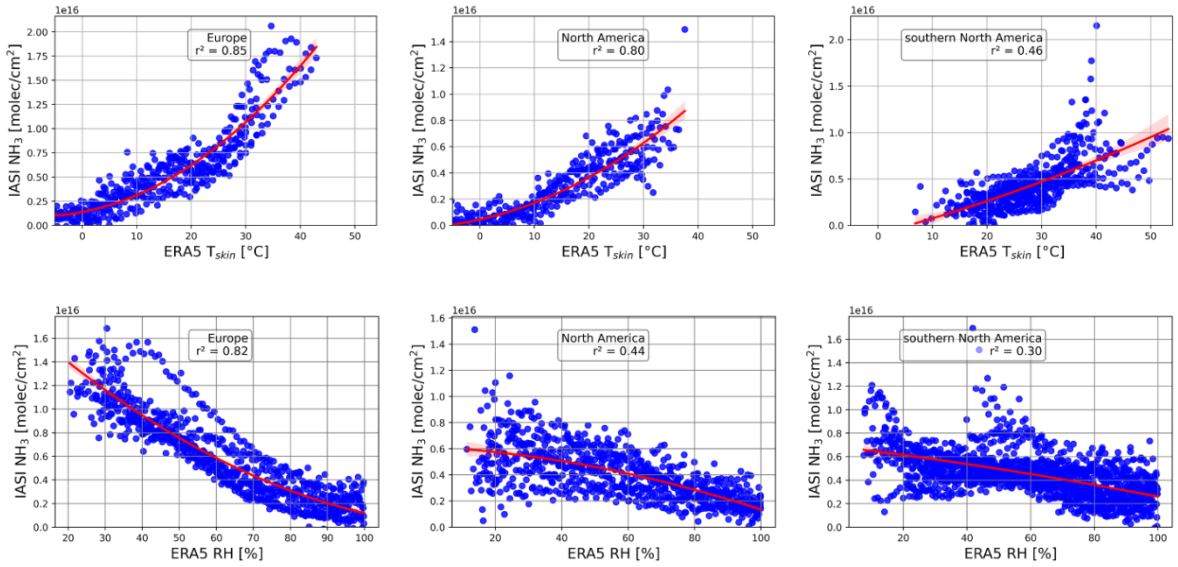


Figure S4: Evolution of NH_3 with respect to land surface temperature (upper panels) and relative humidity (lower panels) from ERA5 in the 3 study domains. The figure is done by averaging IASI NH_3 total columns per bins of ERA5 skin temperatures, with an interval of 1°C. Blue dots: Yearly IASI NH_3 total columns (molecules/cm²) averaged per bins of ERA5 skin temperature in the upper panel (relative humidity in the lower panel) with an interval of 1°C (1% of RH) between each consecutive bin. We do not consider bins that contain less than 5% of the maximum number of measurements per bin; hence, the averages with not enough measurements per bins are excluded. The regions considered here are the regions presented in Table 1 above Europe (left panel), North America (middle panel), and southern North America (right panel). The red line is a polynomial fit of second order, and the relevant r^2 is shown on each panel.



The effect of relative humidity on NH_3 concentrations is different in each of the study domains, as we can see in the lower panels of Figure 4. The highest correlation factor we observe is over Europe, and it accounts to $r^2 = 0.82$ (lower left panel). A study by Reynolds and Wolf (1987) concluded that the relative humidity of the air does not play a major role in NH_3 volatilization unless the soil is dry. In fact, soil is drier in Europe than both of the other regions, with North America being the most humid area. This can explain why we see a good correlation in Europe and lower one in North America. In southern North America, however, throughout the year we observe high temperatures and high humidity, which can explain the low correlation factor $r^2 = 0.30$ (lower right panel).

Despite the differences in the correlations, we can still see a decreasing trend of ammonia as the relative humidity of the air increases. We looked at the times during which the NH_3 concentrations where detected and we summarize them below:

- RH = 0 – 40%: Most of the NH_3 detected in all regions is during the spring season when the concentrations are the highest. One note that when $0 \leq RH \leq 25\%$, April dominates in Europe and May dominates in North America and southern North America.
- RH = 40 – 60%: The NH_3 detected is during summer and spring, hence the lower average as the RH increases when the summer approaches.
- RH = 60 – 85%: The NH_3 is decreasing as RH increases and as the time approaches winter (when RH is highest). In southern North America, however, these ammonia measurements correspond to the spring season mostly.
- RH = 85 – 100%: Most of NH_3 detected are during winter in southern North America and Europe, and evenly distributed throughout the year in North America.

Figure S5: Yearly IASI NH_3 total columns (molecules/cm²) averaged per bins of ERA5 skin temperatures (°C), with an interval of 1°C between each consecutive bin. The red circles denote the growing seasons, at least 60% of the NH_3 are detected during March-May and Sept-Nov periods. The regions considered here are the regions presented in Table 1 in North America.

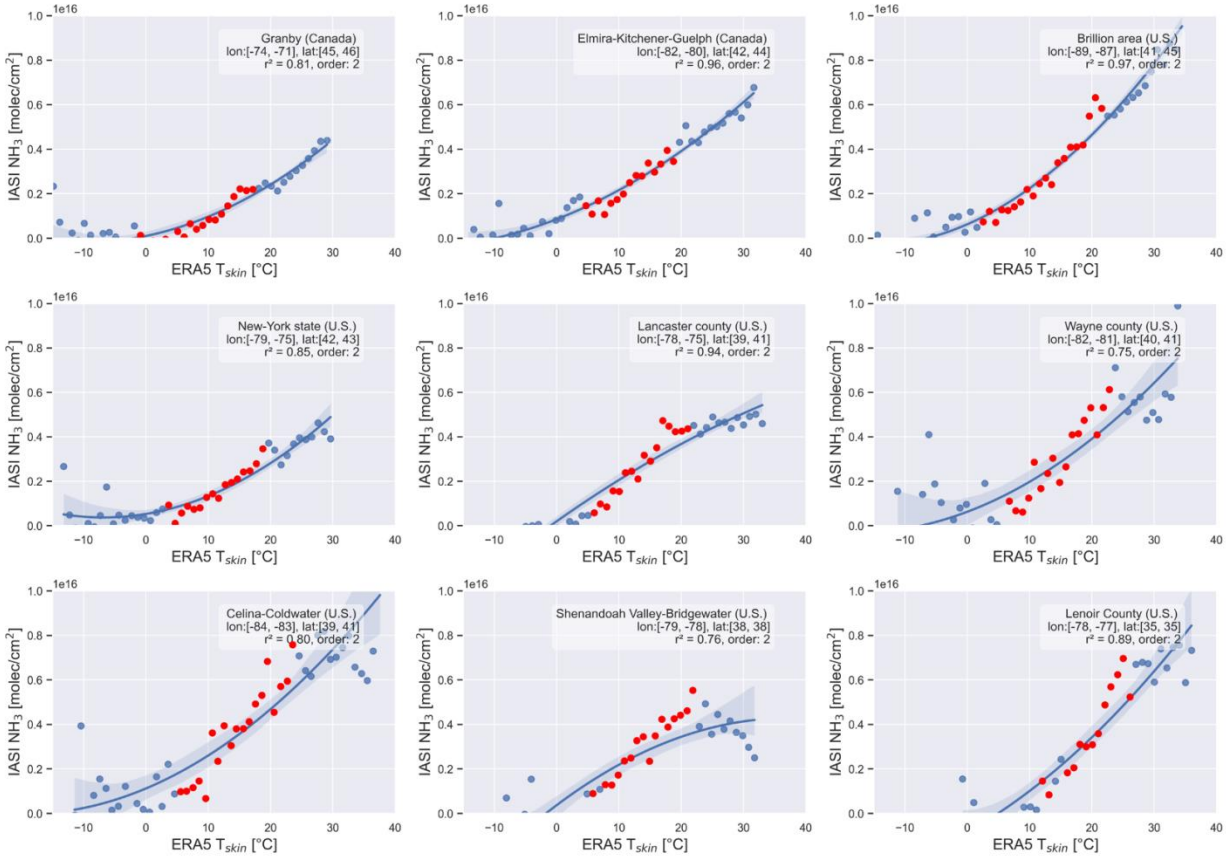
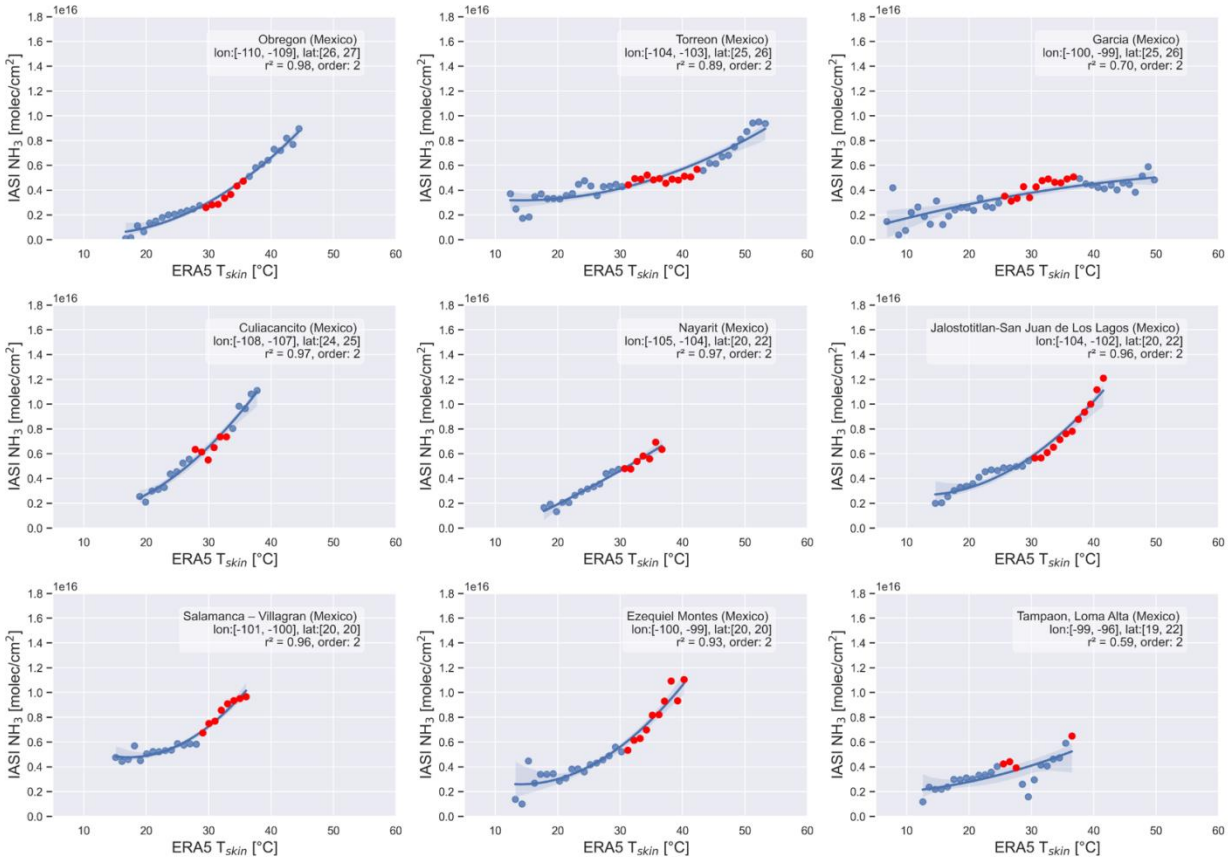


Figure S6: Yearly IASI NH_3 total columns (molecules/cm²) averaged per bins of ERA5 skin temperatures (°C), with an interval of 1°C between each consecutive bin. The red circles denote the growing seasons, at least 60% of the NH_3 are detected during March-May and Sept-Nov periods. The regions considered here are the regions presented in Table 1 above southern North America: (a) sub-regions A to I, (b) sub-regions J to S.

(a)



(b)

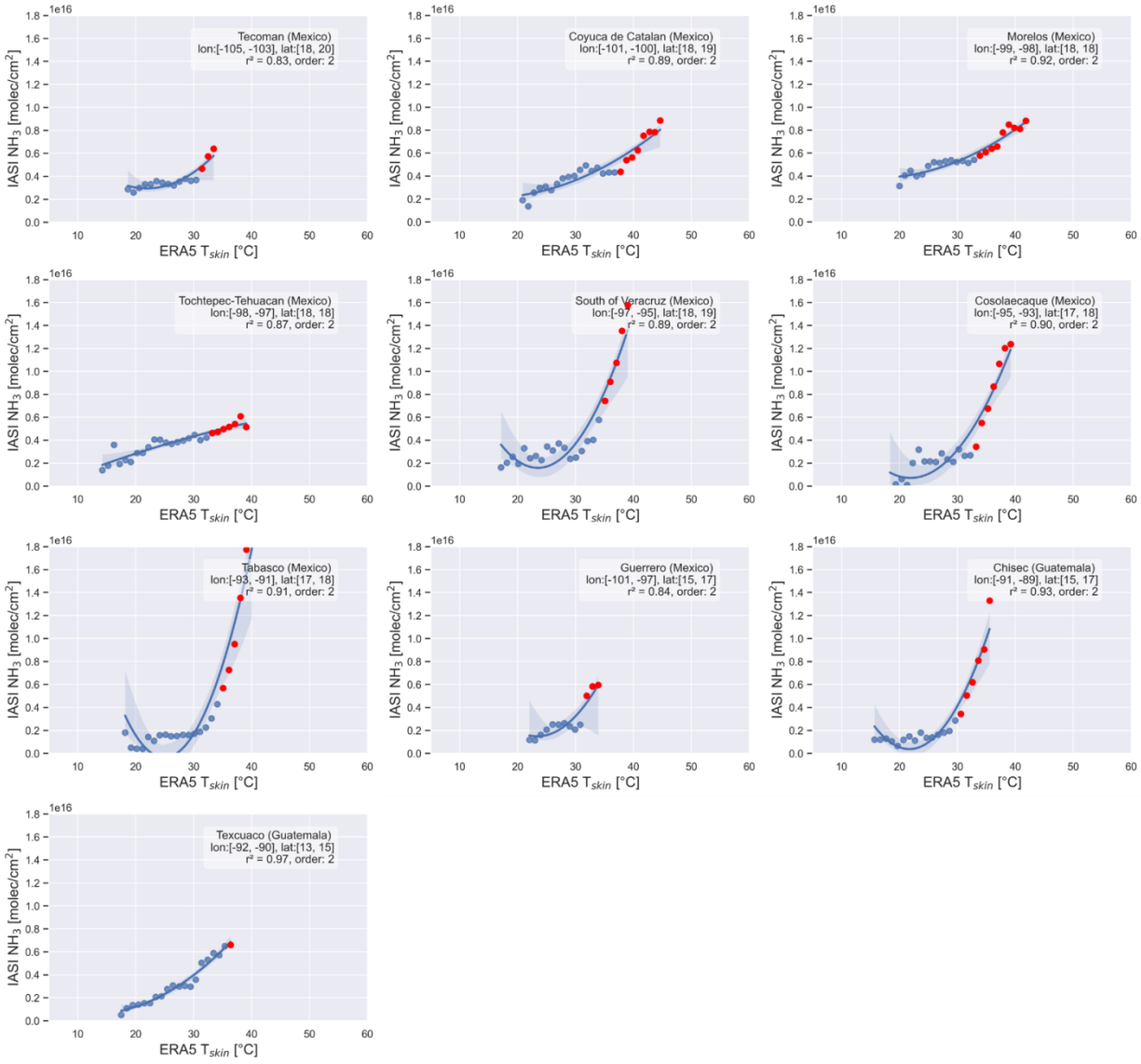
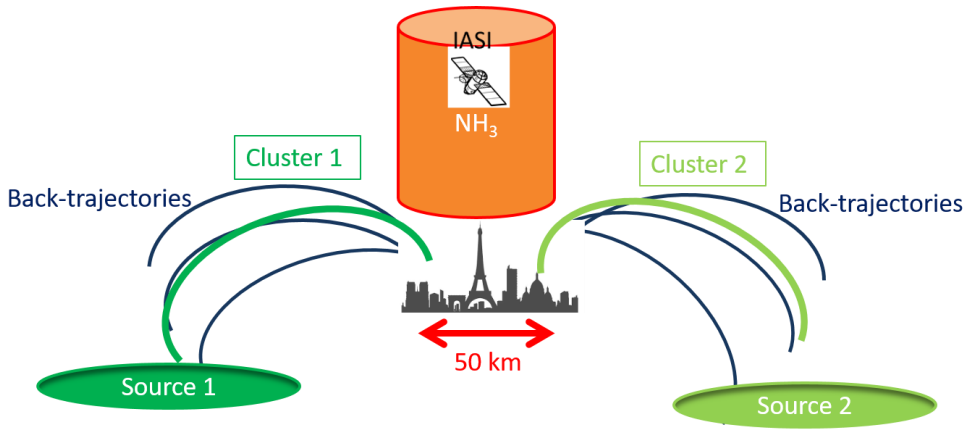

Tecoman (Mexico)
lon:[-105, -103], lat:[18, 20]
r² = 0.83, order: 2
Coyuca de Catalan (Mexico)
lon:[-101, -100], lat:[18, 19]
r² = 0.89, order: 2
Morelos (Mexico)
lon:[-99, -98], lat:[18, 18]
r² = 0.92, order: 2
Tochtepec-Tehuacan (Mexico)
lon:[-98, -97], lat:[18, 18]
r² = 0.87, order: 2
South of Veracruz (Mexico)
lon:[-97, -95], lat:[18, 19]
r² = 0.89, order: 2
Cosolaecaque (Mexico)
lon:[-95, -93], lat:[17, 18]
r² = 0.90, order: 2
Tabasco (Mexico)
lon:[-93, -91], lat:[17, 18]
r² = 0.91, order: 2
Guerrero (Mexico)
lon:[-101, -97], lat:[15, 17]
r² = 0.84, order: 2
Chisec (Guatemala)
lon:[-91, -89], lat:[15, 17]
r² = 0.93, order: 2
Texcuaco (Guatemala)
lon:[-92, -90], lat:[13, 15]
r² = 0.97, order: 2
IASI NH3 [molec/cm2]
ERA5 Tskin [°C]
1e16

Figure S7: Cluster analysis - method to analyze the impact of long-range transport on NH_3 concentrations measured over the cities.



1) For each day, we have run HYSPLIT back-trajectories ending in the cities at the overpass time of the IASI satellite (blue lines in Figure S7).

2) For each day, we have calculated the amount of NH_3 derived from IASI observations within a circle of 50km radius around the cities (orange cylinder in Figure S7).

3) We have run the cluster analysis to merge trajectories that are near each other (green lines in Figure S7). The cluster analysis computes the spatial variance and minimize differences between trajectories within a cluster while differences between clusters are maximized [Abdalmogith et al., 2005; https://www.ready.noaa.gov/documents/Tutorial/html/traj_cluseqn.html]. NH_3 mean concentrations measured inside the cities by IASI have been allocated to the different mean cluster trajectories according to the corresponding back-trajectories.

Abdalmogith, S. S. and Harrison, R. M.: The use of trajectory cluster analysis to examine the long-range transport of secondary inorganic aerosol in the UK, Atmos. Environ., 39(35), 6686–6695, doi:https://doi.org/10.1016/j.atmosenv.2005.07.059, 2005.