

**ADDENDUM to Evaluation of 2002 CMAQ
Ammonia and Aerosol Concentration Predictions
for Use in Determining Background Pollutant
Concentrations in CALPUFF BART Analyses:**

**Impact of using CMAQ 2002 monthly averaged by
hour on 2001, 2002 and 2003 NO₃ predictions**

February 2006

Prepared for:

VISTAS

Prepared by:

Earth Tech, Inc.
196 Baker Avenue
Concord, MA 01742

In Section III-B of the main document called “Evaluation of 2002 CMAQ Ammonia and Aerosol Concentration Predictions for Use in Determining Background Pollutant Concentrations in CALPUFF BART Analyses” dated January 31, 2006, CMAQ 2002 monthly averaged were shown not to be adequate for being used as background pollutant for ALM application for all three years, 2001, 2002 and 2003. The present document is to complete this investigation of a possible use of the CMAQ 2002 output data as background for year 2001 and 2003. For this purpose, a diurnal cycle is included in the monthly averages. CMAQ 2002 concentrations outputs are monthly averaged by hour and used as input to CHEMEQ, the NO_3/HNO_3 equilibrium transformation of CALPUFF and POSTUTIL.

Figure 1 and Figure 2 display 2002 hourly time series of NO_3 ($\mu\text{g}/\text{m}^3$) and $\text{TNH}_3=\text{NH}_3(\text{g})+\text{NO}_3$ (ppb), output of CHEMEQ using as input CMAQ 2002 averaged in three different way: hourly (top), monthly (middle) and monthly by hour (bottom). Figure 1 plots are for Great Smoky Mountains National Park and Figure 2 for Shenandoah National Park. Note that only hours with valid CASTNet temperature and relative humidity are used for this exercise. The comparison of the three plots on each Figure underlines the difference between using hourly 2002 CMAQ data and monthly or monthly with diurnal cycle averaged CMAQ data. The NO_3 concentrations range between 0 and 11 $\mu\text{g}/\text{m}^3$ at Great Smoky Mountains NP and between 0 and 18 $\mu\text{g}/\text{m}^3$ at Shenandoah NP when hourly CMAQ are used while NO_3 concentration vary between 0 and 3 (0 and 4) at Great Smoky Mountains NP and between 0 and 4.5 (0 and 6) at Shenandoah for monthly (monthly with diurnal cycle). For these two sites, the addition of a diurnal cycle in the monthly average did not change much the results over using just monthly average.

Figure 3 shows that the monthly average by hour effectively represents a diurnal cycle by month for each pollutant at the two sites, Great Smoky Mountains NP and Shenandoah NP. Note that the hours are displayed in local time. The 2002 CMAQ run was processed in UTC time, so CMAQ output data were shifted by 5 hours. As expected, NO_3 concentrations decrease during the day and increase at night (inversely following the temperature diurnal cycle).

Figure 4 displays 2002 daily average time series of NO_3 concentrations and comparison with IMPROVE observations of the three tests (hourly CMAQ, monthly CMAQ and diurnally-varying monthly CMAQ), while Figure 5 displays 2002 weekly time series of NO_3 concentrations and comparison with CASTNet observations of the same three tests. The two figures show little differences between the run using CMAQ data monthly averaged and run using diurnally-varying monthly averages of CMAQ data. Figure 6 displays the Q-Q plot as function of observations of the time series plotted in Figures 4 and 5. It seems that using any of the three averaging of the CMAQ data at these two sites give NO_3 concentrations larger than observations (both CASTNet and IMPROVE) except for one point when compared to IMPROVE at

Shenandoah NP (bottom right plot). But these encouraging results (not too bad when monthly averages are used) are specific for these two sites with the year 2002. Indeed, plots for other sites and other years display worst results when monthly averages of monthly averages with a diurnal cycle are used instead of hourly averages of CMAQ data as it is demonstrated in the following.

In Addition, two Q-Q plots were generated using most of the CASTNet sites in the Southeast part of the US and IMPROVE sites collocated with CASTNet sites as shown in Figure 7. These two Q-Q plots present the same pattern when using CMAQ data monthly averaged and CMAQ data monthly averaged with a diurnal cycle. The largest NO₃ concentrations are much lower than in the observations, while NO₃ concentrations in the middle range are larger than the observations and also larger than when hourly CMAQ data are used. In general, the results for year 2002 using monthly averaged CMAQ data or diurnally-varying monthly average CMAQ data are worse than using hourly averaged CMAQ data.

Figures 8, 9 and 10 display similar plots processed for year 2001 and 2003 at the two sites, Great Smoky Mountains NP and Shenandoah NP and Figure 11 displays the quantile-quantile plots for all IMPROVE sites collocated with CASTNet sites for both year 2001 and 2003. Figures 8 and 9 are daily averaged time series at Great Smoky Mountains NP and Shenandoah NP, respectively. While Figure 10 summarizes the Q-Q plots for these two years at these two sites when NO₃ predictions are compared with IMPROVE NO₃ observations. For these two years, like for year 2002, using monthly averaged CMAQ data including a diurnal cycle or not gives approximately the same results. But unlike for year 2002 at these two specific sites, the largest NO₃ concentrations at Great Smoky Mountains NP and for a few NO₃ concentrations at Shenandoah NP are underpredicted, while the middle range NO₃ concentrations tends to be larger than the CMAQ hourly averaged. When all sites are put together like in Figure 11, similar conclusions can be drawn than when looking at individual sites for both year 2001 and 2003.

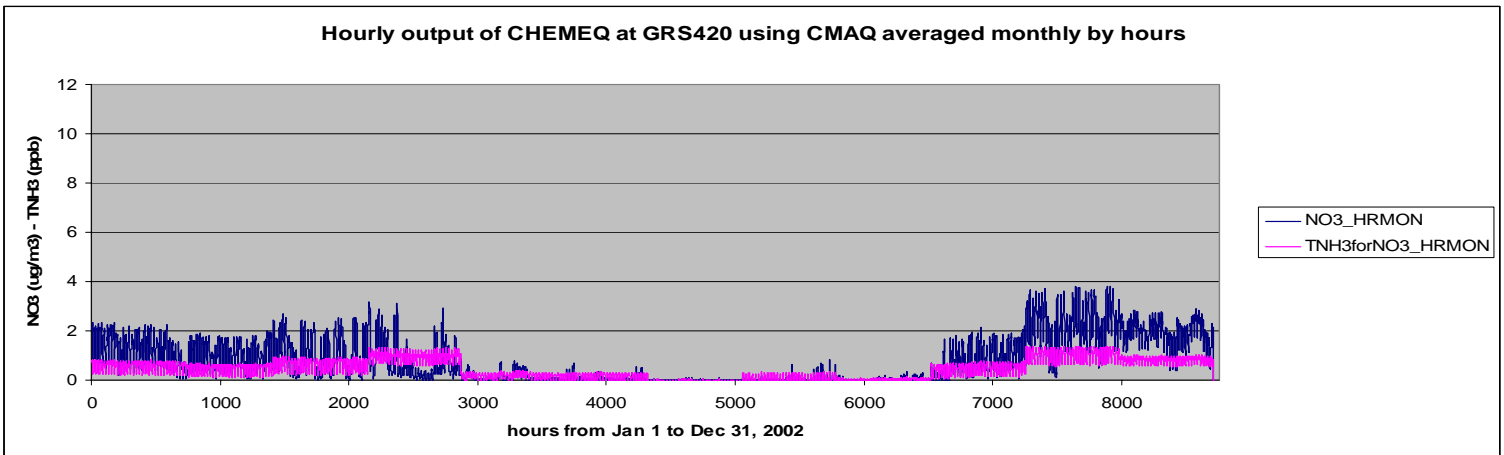
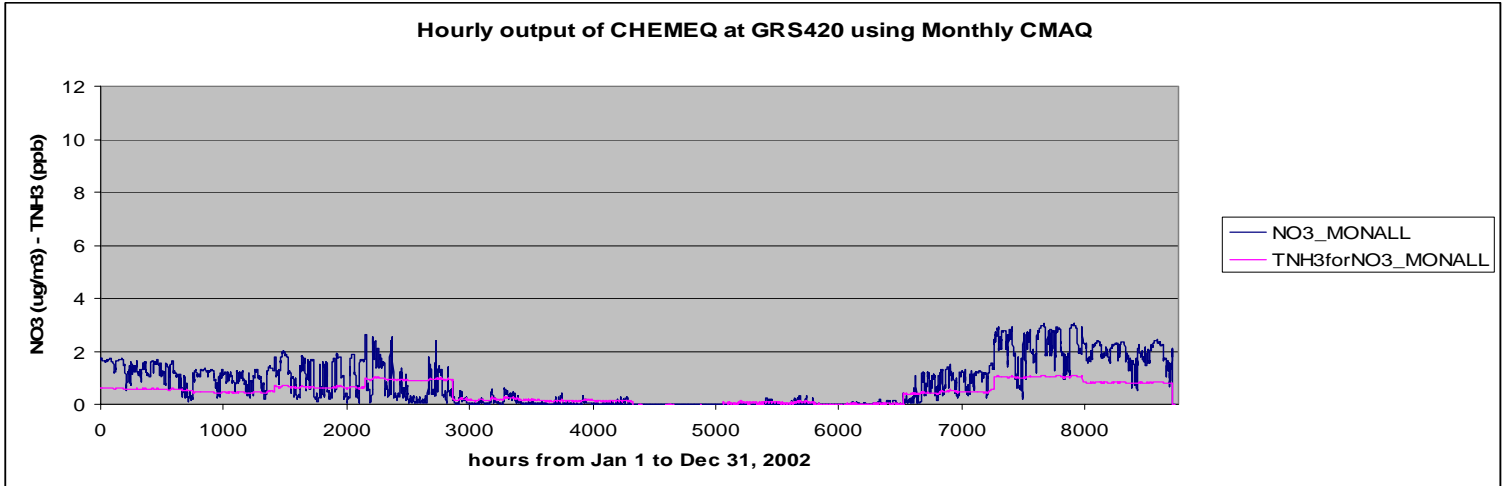
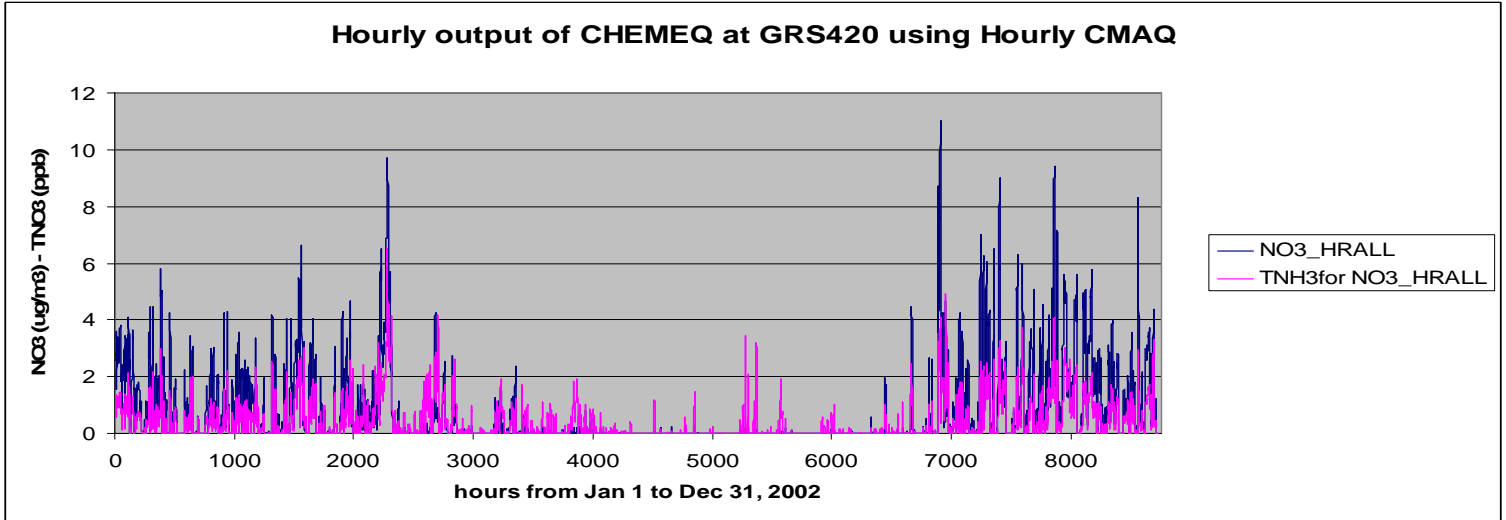


Figure 1. Hourly time series output of CHEMEQ NO_3 ($\mu\text{g}/\text{m}^3$) and TNH_3 (ppb) concentrations at Great Smoky Mountains National Park site using as input hourly CMAQ pollutants (top), CMAQ pollutant monthly averaged (middle) and CMAQ pollutants monthly averaged by hour (bottom). TNH_3 is equivalent to $\text{NH}_3(\text{g})+\text{NO}_3$ (in ppb).

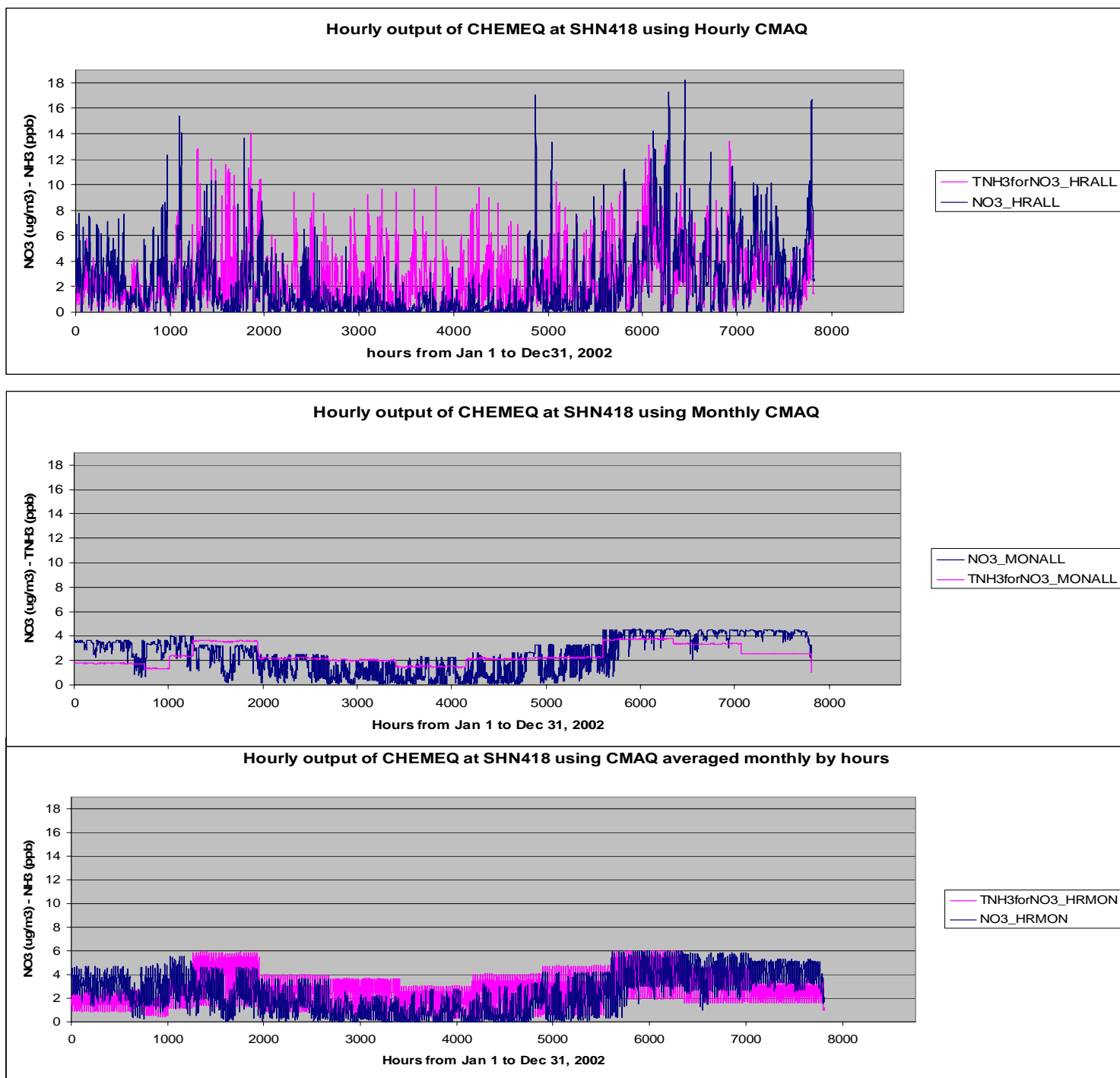
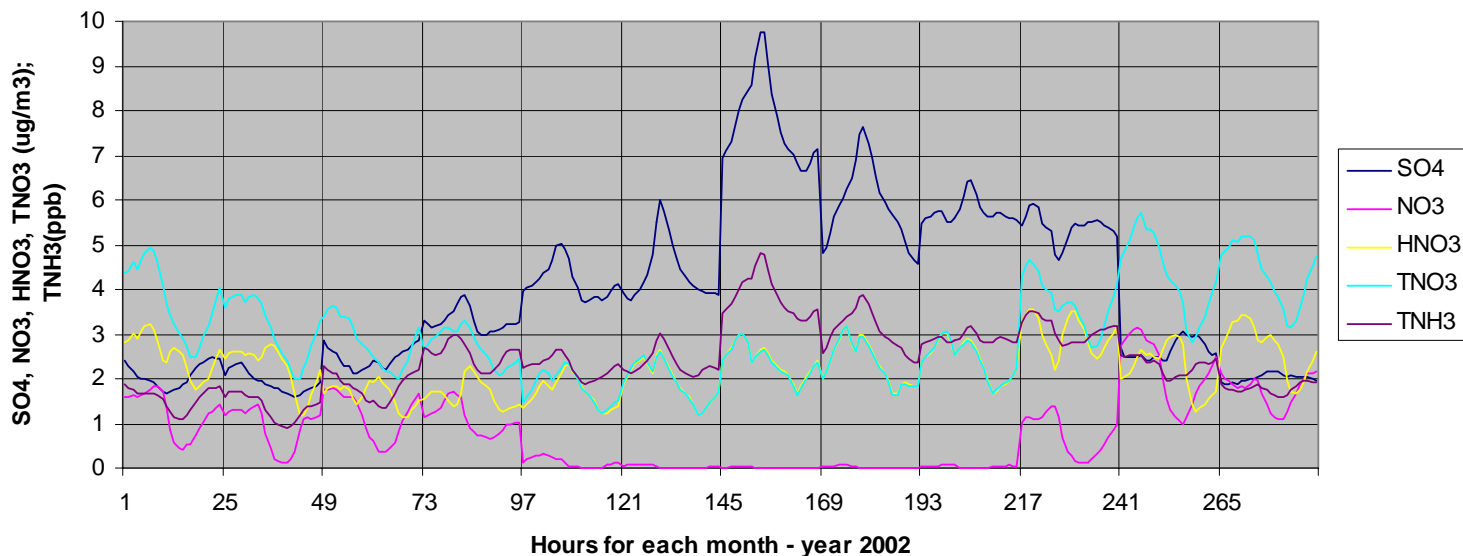


Figure 2. Hourly time series output of CHEMEQ NO₃ (μg/m³) and TNH₃ (ppb) concentrations at Shenandoah National Park site using as input hourly CMAQ pollutants (top), CMAQ pollutant monthly averaged (middle) and CMAQ pollutants monthly averaged by hour (bottom). TNH₃ is equivalent to NH₃(g)+NO₃ (in ppb).

CMAQ monthly averaged by hours at GRS420



CMAQ monthly averaged by hours at SHN418

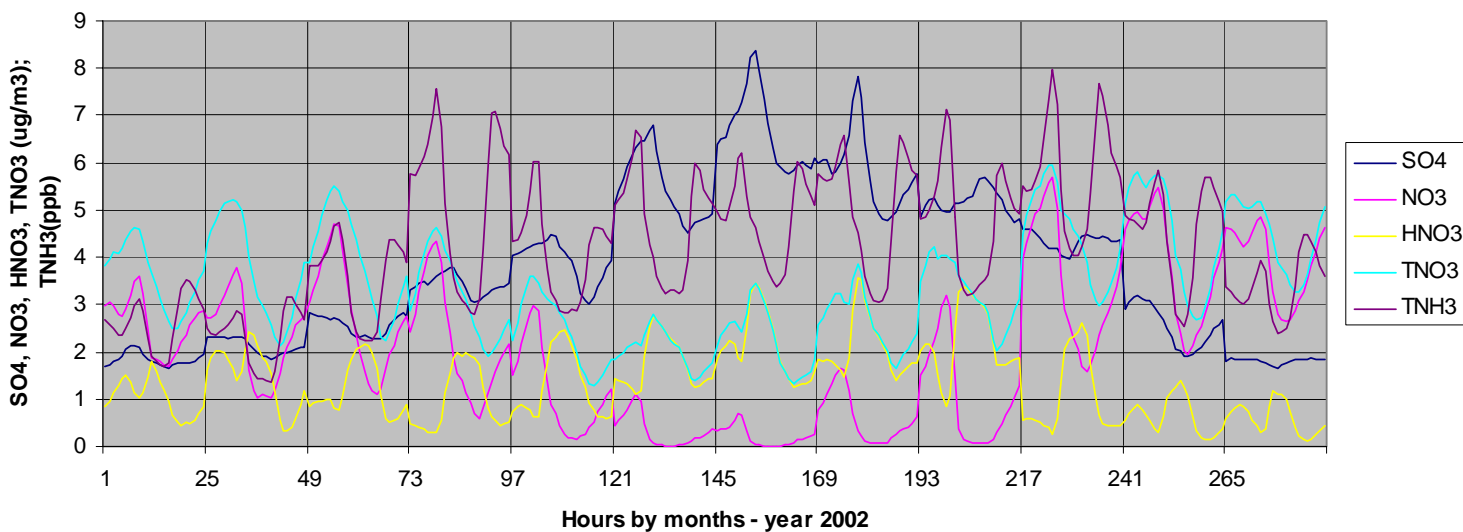


Figure 3. Time series of CMAQ output (2002) monthly averaged by hour plotted at Great Smoky Mountains NP (GRS420, top) and Shenandoah NP (SHN418, bottom). Between 1 and 25, hour 1 to hour 24 are plotted for January, between 25 and 49, hour 1 to hour 24 are plotted for February, etc. TNH_3 equivalent to $NH_3(g)+NO_3+2SO_4$, unlike in Figures 1 and 2 where $TNH_3=NH_3(g)+NO_3$

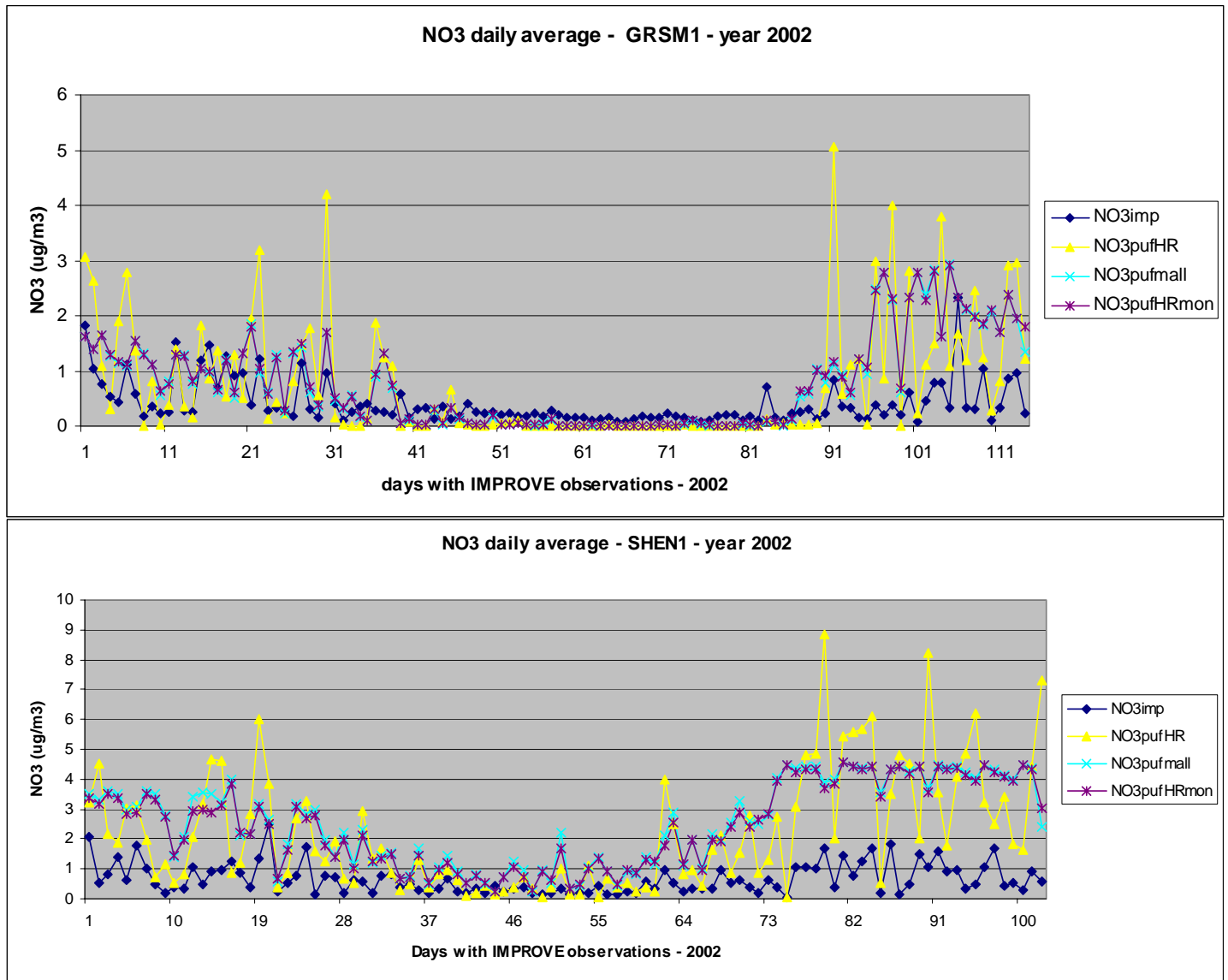


Figure 4. NO₃ concentration 2002 time series daily averaged at Great Smoky Mountains NP (top) and Shenandoah NP (bottom). “NO3imp” (Dark Blue) stands for IMPROVE Observations; “NO3pufHR” (Yellow) stands for Output of CHEMEQ using hourly CMAQ; “NO3pufmall” (Light Blue) stands for Output of CHEMEQ using CMAQ monthly average; and “NO3pufHRmon” (Purple) stands for Output of CHEMEQ using CMAQ monthly average by hour.

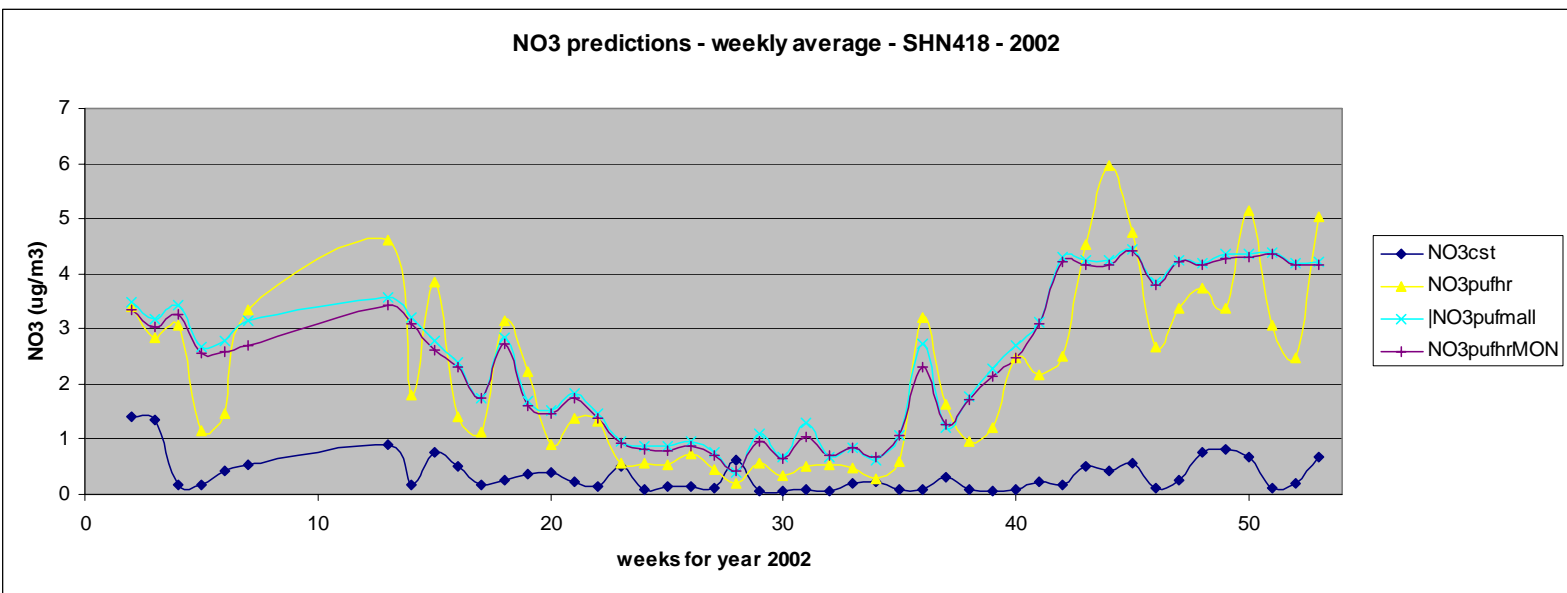
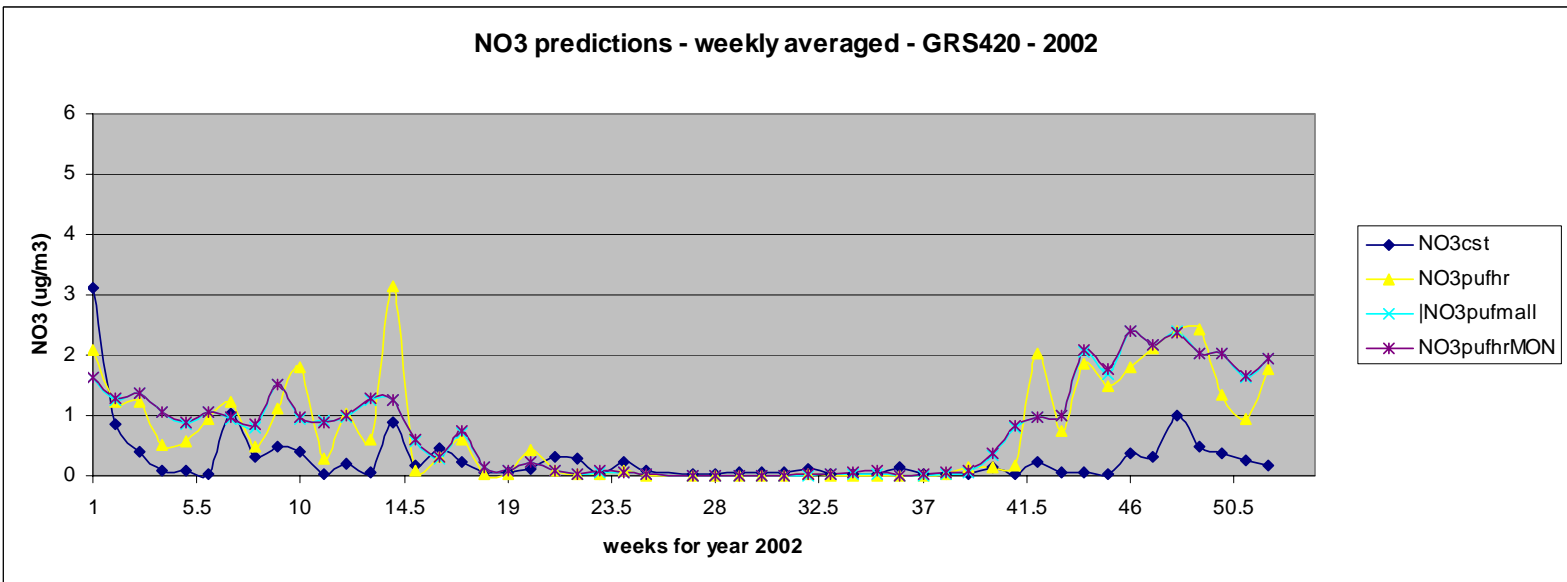


Figure 5. NO₃ concentration 2002 time series weekly averaged at Great Smoky Mountains NP (top) and Shenandoah NP (bottom). “NO3cst” (Dark Blue) stands for CASTNet Observations; “NO3pufHR” (Yellow) stands for Output of CHEMEQ using hourly CMAQ; “NO3pufmall” (Light Blue) stands for Output of CHEMEQ using CMAQ monthly average; and “NO3pufHRmon” (Purple) stands for Output of CHEMEQ using CMAQ monthly average by hour.

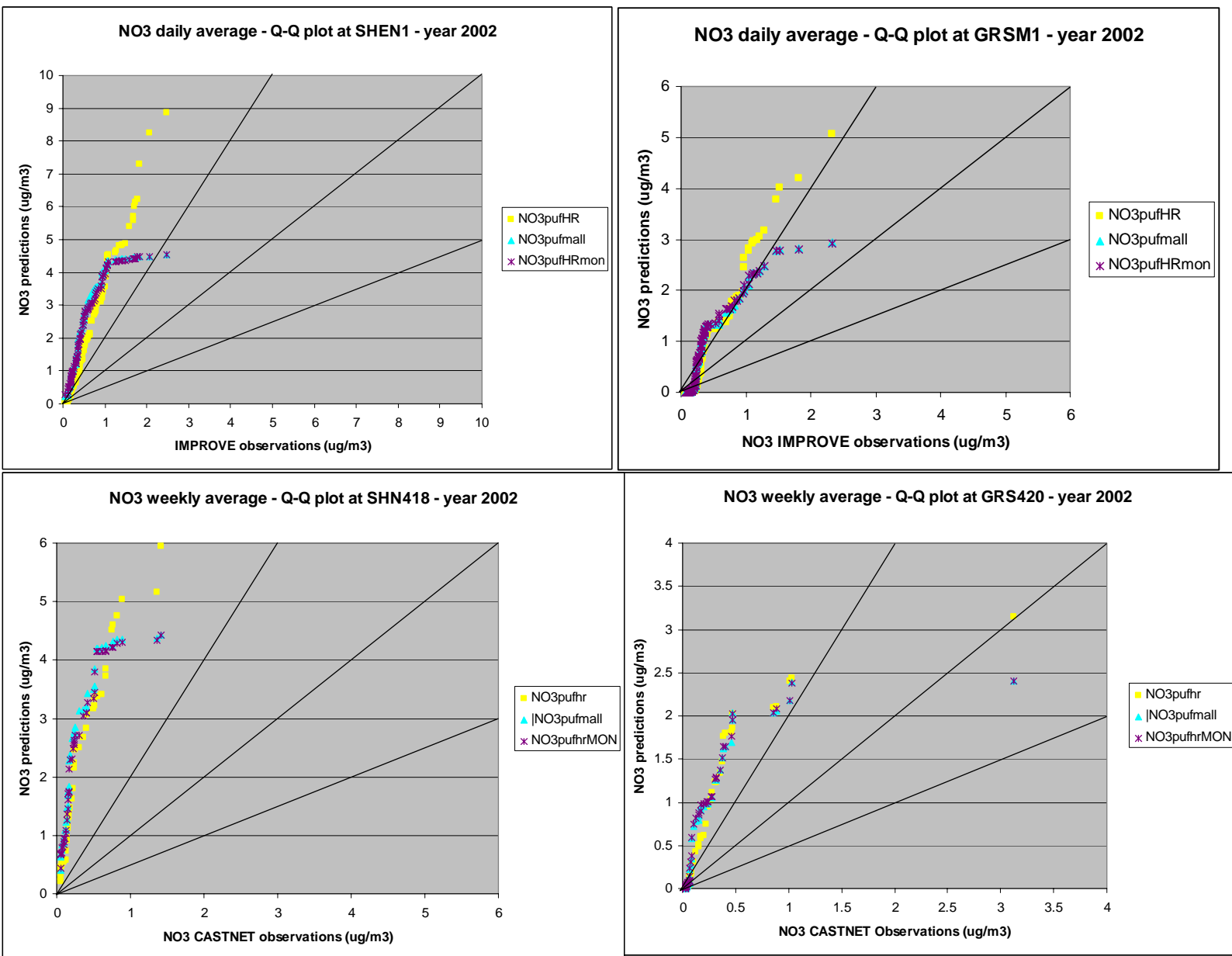


Figure 6. NO₃ concentrations for year 2002 at Great Smoky Mountains NP (right panels) and Shenandoah NP (left panels) plotted as daily averaged Q-Q as function of IMPROVE observations (top panels) and as weekly averages as function of CASTNet observations (bottom panels) “NO₃pufHR” (Yellow) stands for output of CHEMEQ using hourly CMAQ; “NO₃pufmall” (Light Blue) stands for output of CHEMEQ using CMAQ monthly average; and “NO₃pufHRmon” (Purple) stands for output of CHEMEQ using CMAQ monthly average by hour.

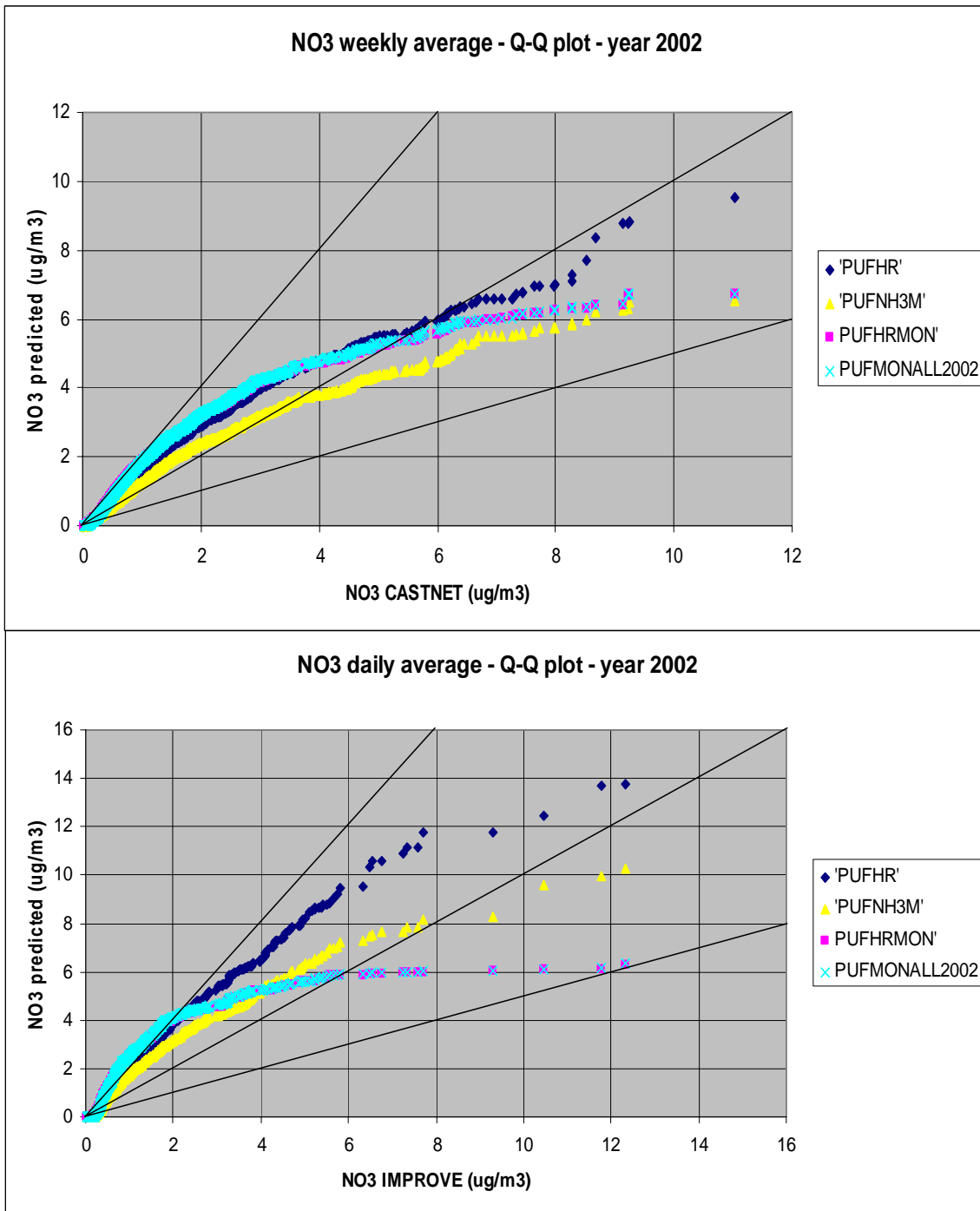


Figure 7. CMAQ monthly averaged by hours input into CHEMEQ comparison with observations (CASTNet and IMPROVE) – most sites included for CASTNet, 11 co-located sites for IMPROVE. Dark blue (PUFHR) stands for outputs of CHEMEQ using hourly CMAQ; Yellow (PUFNH3M) stands for output of CHEMEQ using hourly CMAQ + monthly NH₃; Pink (PUFHRMON) stands for output of CHEMEQ using CMAQ monthly average by hours; and Light blue (PUFMONALL2002) output of CHEMEQ using monthly CMAQ for all

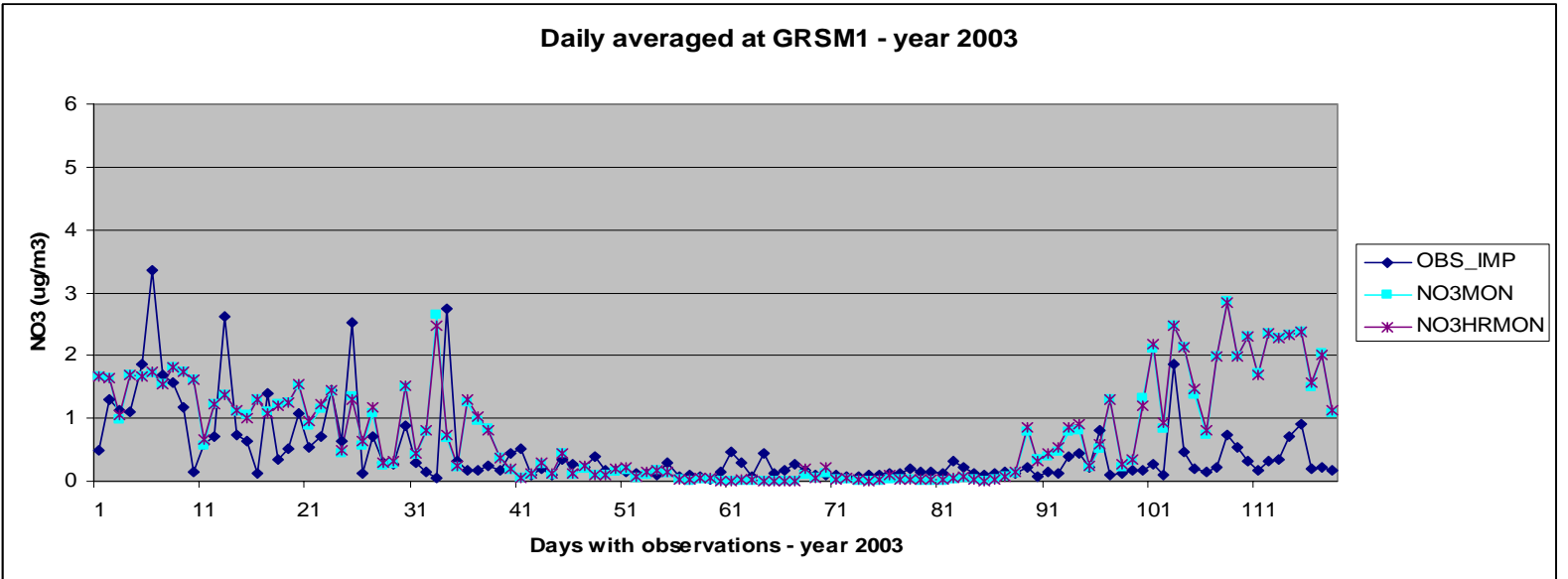
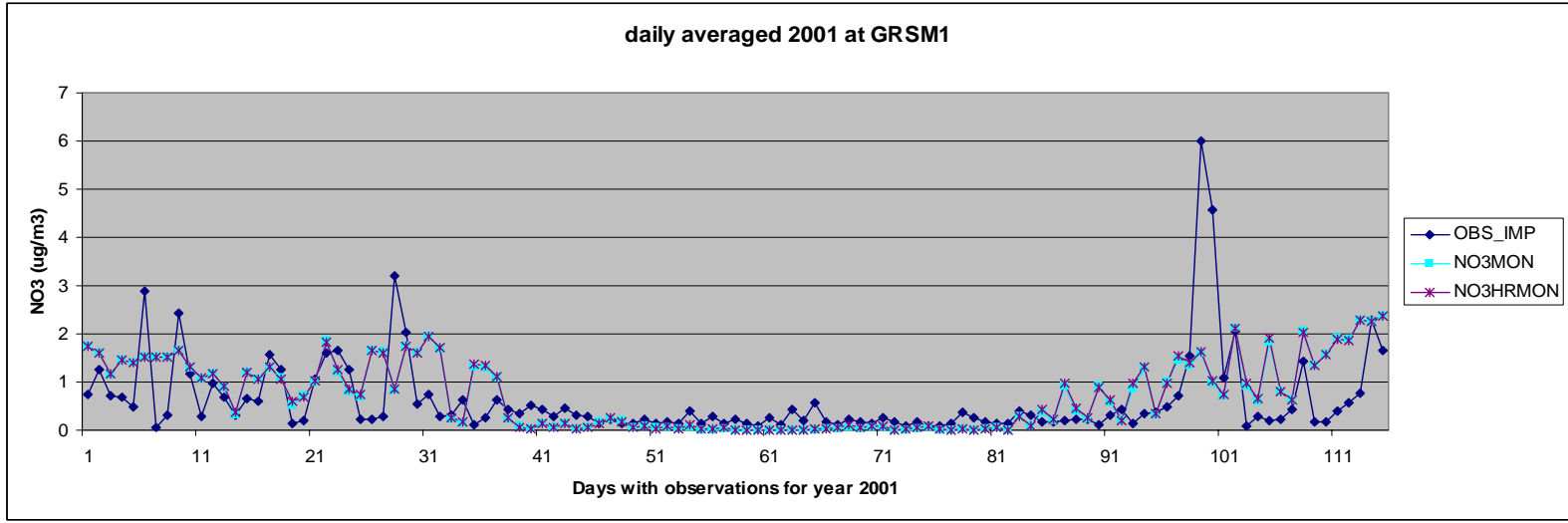


Figure 8. NO₃ concentration time series for year 2001 (top) and for year 2003 (bottom), daily averaged at Great Smoky Mountains NP (top), comparison with IMPROVE observations at the same sites. “OBS_IMP” (Dark Blue) stands for IMPROVE Observations; “NO3MON” (Light Blue) stands for Output of CHEMEQ using CMAQ monthly average; and “NO3HRMON” (Purple) stands for Output of CHEMEQ using CMAQ monthly average by hour.

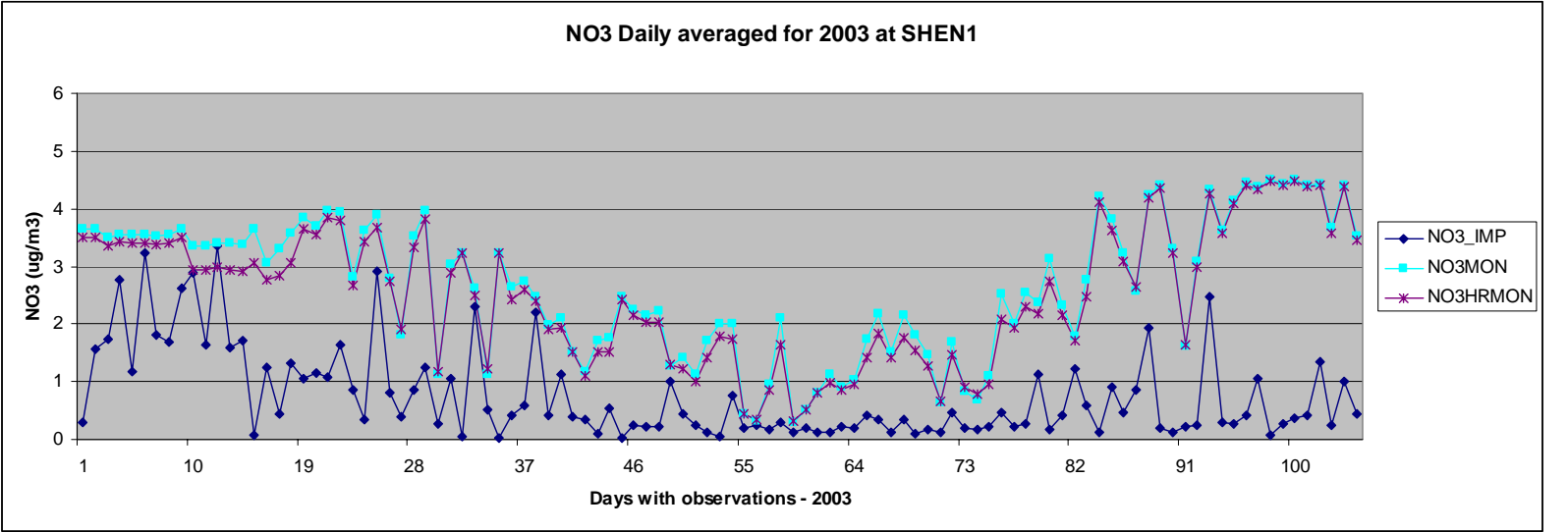
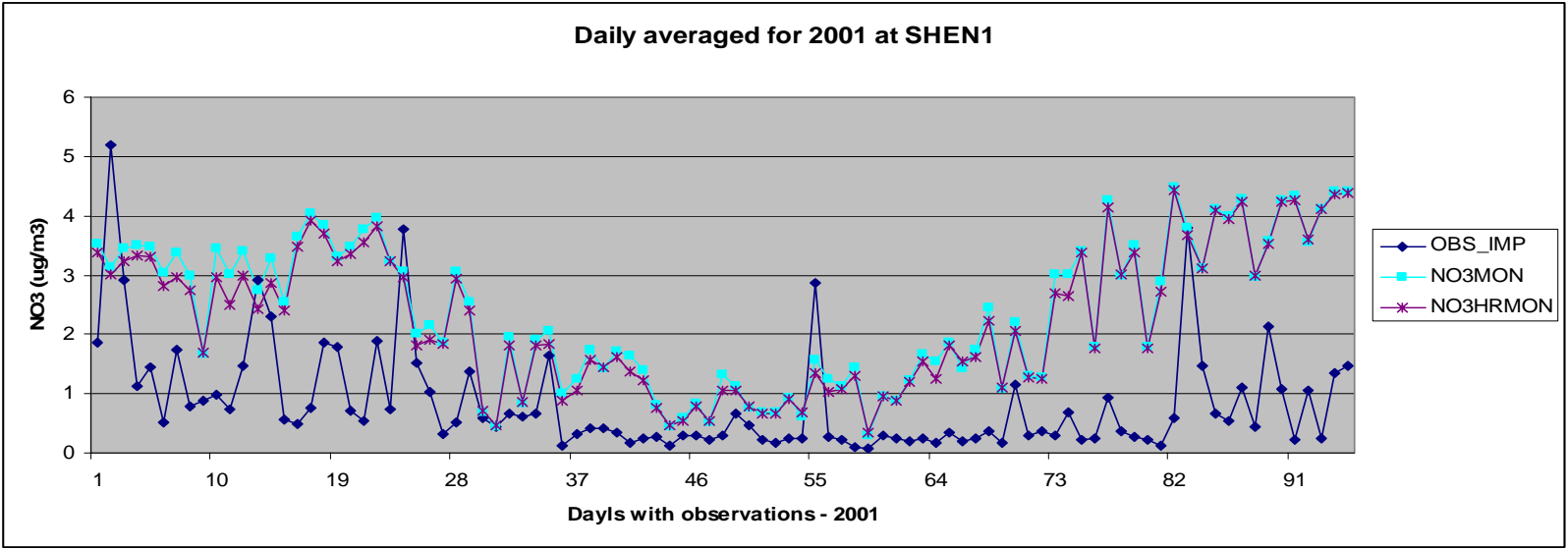


Figure 9. NO₃ concentration time series for year 2001 (top) and for year 2003 (bottom), daily averaged at Shenandoah NP (top), comparison with IMPROVE observations at the same sites. “OBS_IMP” (Dark Blue) stands for IMPROVE Observations; “NO3MON” (Light Blue) stands for Output of CHEMEQ using CMAQ monthly average; and “NO3HRMON” (Purple) stands for Output of CHEMEQ using CMAQ monthly average by hour.

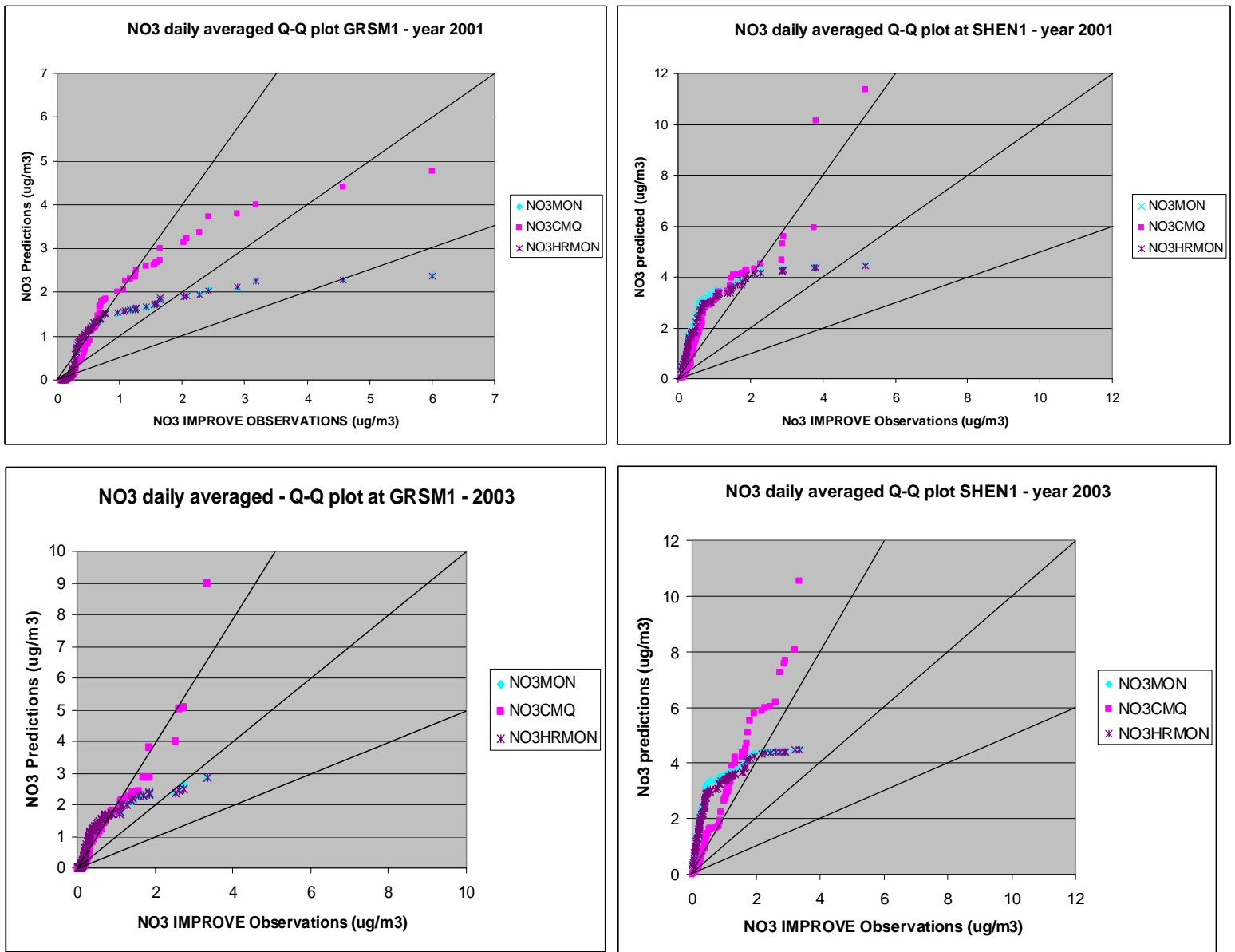


Figure 10. NO₃ concentration daily averaged Q-Q plots as function for IMPROVE observations for year 2001 (top) and for year 2003 (bottom) at Great Smoky Mountains NP (left panels) and at Shenandoah NP (right panels). “NO3CMAQ” (Pink) stands for IMPROVE CMAQ output; “NO3MON” (Light Blue) stands for Output of CHEMEQ using CMAQ monthly average; and “NO3HRMON” (Purple) stands for Output of CHEMEQ using CMAQ monthly average by hour.

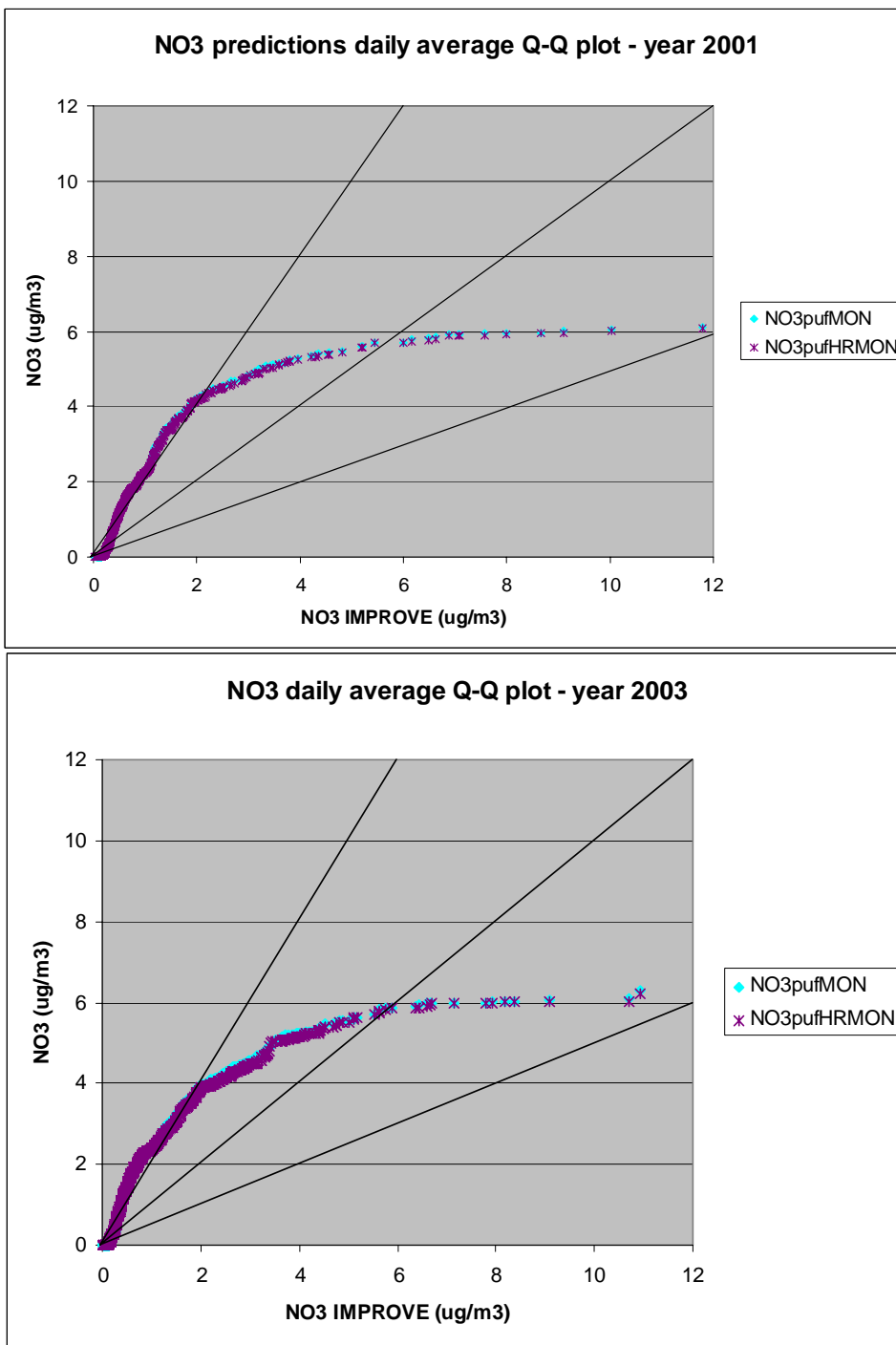


Figure 11. NO₃ concentration daily averaged Q-Q plots as function for IMPROVE observations for year 2001 (top) and for year 2003 (bottom) for all IMPROVE sites collocated with CASTNet sites. “NO3MON” (Light Blue) stands for Output of CHEMEQ using CMAQ monthly average; and “NO3HRMON” (Purple) stands for Output of CHEMEQ using CMAQ monthly average by hour.

SUMMARY AND CONCLUSION

In order to improve the poor results obtained by using 2002 CMAQ data monthly averaged for NO₃ concentrations predictions, a monthly average of the CMAQ pollutants by hour was performed. Despite an actual diurnal cycle showing for every pollutant (Figure 3), the NO₃ concentrations predicted are approximately the same as when monthly averages are used. The same conclusions can be drawn when the application is processed for all three years, 2002, 2001 or 2003. In summary, the use of the diurnally-varying monthly average of CMAQ data does not improve the results for years 2001 and 2003.

So, the final conclusions are the same as in the main document, *hourly* CMAQ data are recommended to be used in POSTUTIL for ALM application for all three years. The 2002 CMAQ hourly data can be used as background for year 2002 and it is recommended to gather hourly outputs of CMAQ model (or equivalent Eulerian model) run for year 2001 and year 2003. The quality of these new dataset needs to be proved to be as good as the 2002 CMAQ 36-km dataset or better. Earth Tech will perform a quality assessment of these datasets when they are available. If the quality is acceptable, a CALPUFF concentration files will be generated with the hourly background data of 2001 and 2003 and the ALM procedure described in Table 5-1 of the main document can be applied.

If it is not possible to get hourly Eulerian model output for year 2001 and 2003 or if the quality of the outputs are poor, the use of CASTNet observations (available on a weekly basis) can be interpolated spatially to cover all Class I areas and total NH₃ can be empirically computed by iteration from CASTNet observations for an alternative method to create variable background pollutants for ALM application for these two years.