

Review of Atmospheric Measurement Techniques Discussions Paper
“Ethylene Oxide Monitor with Part-per-Trillion Precision for In-Situ Measurements”

by Tara Yacovitch, Christoph Dyroff, Joseph Roscioli, Conner Daube, J. Barry
McManus, and Scott Herndon

This paper describes an instrument and associated measurement and data processing protocols for measurements of the important carcinogen trace gas ethylene oxide. As discussed, these measurements are extremely challenging due to the very low ambient concentrations at pptv levels and a number of potential spectroscopic interferences from higher concentration ambient constituents. This paper is highly relevant, the discussion is clearly written, and the measurements are carefully carried out. The ambient ethylene oxide enhancements downwind of two facilities known for operations involving this gas provide clear convincing evidence for the merits of this instrument as well as the instrument performance in real-world ambient conditions. This reviewer recommends final publication of this paper after the authors consider the following minor comments/suggestions to improve the paper quality further.

1. Would be informative to briefly describe the definition for the inhalation unit risk discussed in the Introduction.
2. Would also be informative to briefly describe in the Introduction that ethylene oxide (C_2H_4O , MW = 44.052 g/mole) is a cyclic three-membered ring structure with the O atom connected to both carbons.
3. The discussion of the simulations in Fig. 1 should be modified to indicate that you employed the Harrison et al. line parameters also for ethane here. The conditions of temperature and pressure should be included in figure caption. I am a little confused by the choice for the simulated concentrations included in Fig. 1. Shouldn't ambient levels of CH_4 around 2 ppm and H_2O levels of 1 to 4% be used in these simulations or do these simulations represent the residual concentrations after subtracting the humidified matched background spectra? This should be discussed here.
4. The meaning of normalized in Fig. S1 should be included in the figure caption just as you did in the Supplement text on line 46. Also the spelling of “Mcmanus” on line 27 in the Supplement should be corrected to “McManus”. Also maybe indicate why you get a normalized value up to 1.04 in Fig. S1. Is this due to noise or small inaccuracies in your polynomial baseline fitting here?
5. Indicate in the figure caption of Fig. S1 if the blue fit spectrum includes all the gases in the inset of Fig. 1?
6. The certified concentrations for EtO (1.075 ppm) and ethane (1.092 ppm) on line 76 needs to be reversed in accordance with the Analytical Results of Fig. S3. Also the X-axis labels in Fig. S2 in both cases needs to be corrected to 1.092 ppm in accordance with the

Analytical Results. Also, please explain in Fig. S2 why you label the left hand plot “Dry Calibration” as the H₂O values here are actually larger than the right hand plot.

7. On line 60 in the Supplement, you should consider either adding what is in the scrubber cartridge that removes EtO and not H₂O or indicate this is proprietary.
8. On line 87 in the discussion of dividing the subsequent sample spectra, it would be important to indicate if you employ the averaged background spectra over the ambient interval or do you use the updated background spectra for the subsequent ambient spectra? How much do these subsequent background spectra change (i.e., the difference of background spectra).
9. Line 90, what scrubber breakthrough are you referring to, breakthrough in EtO or H₂O? The text implies EtO breakthrough, but this should be spelled out.
10. Line 95 where you indicate the autobackground cycles, I am confused by the cycle values. Shouldn't the mobile measurements employ more frequent background measurements to capture the greater potential due to spatial changes in H₂O and the reverse for stationary samples? Please further explain.
11. In Table S1 please indicate what * refers to in the Table next to the value 0.999
12. In Fig. 2c, you should add to the Y-axis label the units ppb²
13. Line 103: I would change the wording “Measurements average down well” to something like “The variance improves with averaging time....”, which better describes the plot Fig. 2c.
14. Line 107: You should reword “Optical alignment minimizes ...” to something like “Adjustments to optical alignment” Could small changes in the multipass highly dense spot pattern or resulting changes in optical cell noise also be a partially responsible?
15. In Fig. 2c, you should more clearly highlight in the plot the results for the EtO 1 hour smooth. As plotted, I have a hard time recognizing this 1 hour smooth. Are you referring to the portion of the variance between 10³ to 10⁵ sec? If so, you should darken this more in the plot.
16. On Line 108: I would think about rewording the statement “Continuous vibrations are less impactful..”, as the red in-motion variance clearly shows reduced performance relative to the blue stationary performance. I think you are referring to the very large negative 0.5 ppb instantaneous deviations and not the more sustained red variance. Maybe adding a caveat to your statement?
17. It would be useful to provide an additional Fig. 3b plot showing only the hourly measurements with an expanded scale from say -0.05 to +0.05 ppb. This would highlight

better the two regimes. I just now saw this information is contained in your Fig. S4 and would leave it up to the co-authors to include a new Fig. 3b.

18. Line 128: It would be important to point out the importance of your observations that indoor laboratory air echoes outside air offset by 3 hours to highlight that a typical building ventilation system only minimally removes EtO by a factor of 2.
19. The back trajectory in Fig. S6 provides very useful information but the Google Street View inset really doesn't add anything. I would recommend providing a more convincing view of this facility (if you can legally show a picture of this sterilization facility) or remove the inset.
20. In the Figures S4 showing facility A and the wind barbs, the facility A site indicator should be made larger in each case. Also the conventional definition of a wind barb indicates the direction from which the wind is blowing. The explanation in the caption of Fig. S7 and Fig. 5 indicating the wind barbs pointing into the wind is a little confusing given the conventional definition. This needs to be clarified.