Atmos. Chem. Phys. Discuss., 3, S1573–S1579, 2003
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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on “Intercomparison of aircraft instruments on board the C-130 and Falcon 20 over southern Germany during EXPORT 2000” *by* N. Brough et al.

Anonymous Referee #2

Received and published: 21 September 2003

Review of "Intercomparison of aircraft instruments on board the C-130 and Falcon 20 over southern Germany during EXPORT 2000", Brough et al., Atmos. Chem. Phys. Discuss., 3, 3589-3623, 2003.

General comments.

Overall a good report of a useful comparison exercise between measurements of NO, NOy, O3, and CO between two major European research aircraft.

The authors could improve the utility of this report substantially if they would include a brief section on the implications of their comparison results on any subsequent scientific use of the data. For example, the NOy data are judged to be in agreement within the combined estimated uncertainties; all well and good. However, there remains a substantial offset between the two NOy data sets, on the order of 90 pptv at 500 pptv ambient NOy, and a slope difference of ca. 15%. While this is within the combined uncertainties, two additional points could and should be made in this report. 1). The comparison does not fully test the stated accuracy of the DLR instrument, as the UEA uncertainties are a factor of ca. 3 larger. 2). This level of discrepancy in NOy can have a substantial impact on interpretation of "missing NOy", especially in light of the recent report of Day et al. ("On alkyl nitrates, O3, and the 'missing NOy'", J. Geophys. Res., 108(D16) 4501, doi:10.1029/2003JD003685, 2003). Were an NOy budget to be constructed from both aircraft data sets, despite the noted agreement in NOy, two completely different conclusions on the magnitude of missing NOy would be drawn.

What would be the recommendations of Brough et al. in this regard? Are the DLR data sufficiently more accurate and precise (see Tables 1 and 2) to support that kind of analysis? Are the UEA data insufficient in this regard, at the ambient levels encountered in the EXPORT 2000 mission? Is neither data set judged to be accurate at the relevant levels to permit an NOy balance calculation, if the individual NOy species measurements were available? The authors should be encouraged to include their assessment of the remaining discrepancies, which could add a very useful dimension of information to the present report.

After these issues have been addressed, I would recommend its publication in Atmospheric Chemistry and Physics with the additional corrections as outlined below.

Specific comments.

> Introduction, p.3591, line 22: "To obtain a good instrument comparison it is advisable that the mixing ratios of the compounds to be measured are not consistently near to the detection limits of the instruments, as this can result in erroneous statistical analysis [sic]."

True, insofar as the comparison of data is intended to provide information only on instru-

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mental calibrations. But wouldn't an ideal comparison include ambient data throughout the full range of atmospheric relevance, including (at least for NO and NOy instruments) levels at and below detection limits? Certainly for CO and CO2 the ambient background is typically well above detection limits throughout the atmosphere, but accurately quantifying a difference between ambient levels of, e.g., 1 and 10 pptv of NO can be critically important for determining the photochemical ozone tendency of an air mass. The above sentence, limiting a comparison exercise to relatively high mixing ratios of a compound, seems to dismiss the importance of accurate knowledge of instrumental zero, or background, levels. For the present report, most of the background tropospheric concentrations of NO were close to both instrumental quantitation limits, so this would seem especially relevant for the data sets in question. Could another sentence be added here discussing the importance of testing instrument behaviour through intercomparison exercises at both high and low ambient mixing ratios?

$>$ Experimental, p. 3593, line 20: "... and there was no verbal in-flight discussion of concentration profiles."

Was the rest of this comparison performed blind, as well? For the data presented here, was there any communication during the data reduction process, and were any of the data resubmitted after the initial data evaluation took place?

$>$ Experimental, p. 3594, line 7: "The inlet was specifically designed to minimize sample-surface interactions and consequently had a low-volume permitting fast sampling of aerosols up to 4 [micrometers] thus allowing discrimination of nitrate aerosols (Ryerson et al., 1999)."

On the contrary, the inlet described by Ryerson et al. (1999) was reported to experience extensive sample-surface interactions: "... all the sampled air contacts an inlet surface, and NOy instrument response time to HNO3 is governed by sample-surface interactions." No quantitative description of aerosol transmission was provided, other than the assumption that the majority of aerosol by mass was excluded due to inlet design and orientation. The extent to which the NOy inlet in the present study was similar to that described by Ryerson et al. is not clear; however, the cited reference does not support the conclusion in the sentence quoted above.

> Experimental, p. 3594, line 23: "The sensitivity of the NO channel was 6.1 +/- 0.9 cps pptv-1 ...".

The imprecision given here of (0.9/6.1) = +/-15% in the derived sensitivity seems rather high for a well-operated instrument. When combined with an uncertainty in the calibration tank gas mixing ratio of (0.1/1.01) = +/-10%, plus an estimated uncertainty in sample and calibration mass flow controller calibrations of ca. 4% absolute, and even assuming perfectly stable background count rates in the NO instrument, these values seem to be inconsistent with the accuracies for the UEA NO data stated in Table 1 of +/- 12% at 50 pptv. The scatter in the derived NO sensitivities alone can account for more than the total imprecision given in Table 1.

> Experimental, p. 3595, line 22: "The major contributor to the inaccuracy for each detector was the instrument artifact signal."

At low levels of ambient NO, yes; however, an imprecision of +/-15% in the NO channel sensitivity derived from the in-flight calibrations suggests that at levels well above the detection limit, the calibration uncertainty dominates the measurement inaccuracy.

> Experimental, p. 3598, line 21: "The O3 standard source was calibrated against O3 measurements at the global watch station at Hohen Peienberg [sic], Germany."

Although unstated, I assume this O3 standard source was based on UV absorption at 254 nm. If so, this provides a direct measurement of O3 number density and is subject only to uncertainty in the measured cross section, gas temperature, pressure, and cell path length - it is most emphatically not capable of being calibrated, unless Beer's Law is itself being "calibrated". Please be careful in the wording here; comparison or referencing of two nominally absolute measurements cannot be a calibration.

> Results and discussion, p. 3601, line 4: "At an indicated aircraft speed of 180 knots this would give plumes [sic] widths of between 0.7 km and 4.3 km." This calculation assumes the aircraft crossed the plumes at 90 degrees to the long axis of the plume. Depending on the intercept angle, an aircraft could take e.g., 48 s to cross a 1-km-wide plume if the flight track vector had a substantial component along the plume, rather than across it. Variations of plume mixing ratio with altitude would also add uncertainty to this calculation.

> Results and discussion, pp. 3601 and following: In the discussions of uncertainty, the observed differences in fitted slopes between the UEA and DLR instruments are described, e.g., as "... well within the overall instrument uncertainty of the UEA NOy (21% at 450 pptv) and just outside those for the DLR instrument (8% at 450 pptv)."

The distinction is meaningless; the applicable metric is whether or not the observed discrepancy in the fitted slope can be encompassed within the combined uncertainty of the two instruments, presumably obtained by addition in quadrature. The only statistically meaningful conclusion is that the observations are not different from a slope of 1.0 within the combined uncertainty of the two instruments. It is further true that the relatively larger UEA uncertainty does not provide as robust or rigorous a test of the DLR instrument as could be hoped, but that is the nature of the comparison exercise. It follows that conclusions drawn from data obtained using the instrument of lower uncertainty will be defensible to a greater degree, but the comparison suggests that no substantial, unknown sources of error exist in either measurement.

> Results and discussion, p. 3603, line 10: "[A significant increase in CO] was clearly not observed by the DLR instrument and would appear to indicate real variations in ambient CO since good agreement was obtained for the rest of the comparison period."

Another possibility that could account equally well for the observed but transient instrumental difference would be short-term instability in one or the other of the CO instruments. Without additional evidence for "real variations in ambient CO", there is

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no a priori reason to exclude these data just because they differ. If that were the case, every comparison data point that disagreed for whatever reason could be written off, making the comparison exercise meaningless.

Are there ancillary data available that further point to this period being different? The O3 data might help in this regard, but more support for this assertion must be presented here or it weakens the entire comparison exercise. "Air mass sampling differences due to the spatial arrangement of the aircraft" are mentioned - well, were these spatial arrangements any different for this 3-min period than for the rest of the exercise? Please present some aircraft positional data to support this assertion, and at least mention the possibility of short-term instrument instability as another, if less palatable, explanation for the observed discrepancy.

> Summary, p. 3608, line 23: "The degree of agreement lends confidence to the accuracy of all observed measurements and indicates the accuracy to be within 12% and 15% for the NO and NOy respectively."

These uncertainties are the percentage differences of the fitted slopes from unity from the comparison exercises, but they are not the accuracy of the instruments - those remain as given in Tables 1 and 2, of 12% and 21% for the UEA measurements and 8% and 7% for DLR for NO and NOy, respectively. The comparison exercise can not result in improvements (for UEA, at least) in accuracy of the instrumental data, as the above statement implies; the exercise can only provide an assurance that the uncertainties are appropriately stated. Surely the DLR data quality has not been degraded by a factor of 2 as a result of this exercise? The uncertainties remain as given in Table 2, and this statement is somewhat misleading; better to simply restate the two instruments uncertainties here in the summary.

Technical corrections.

> Acknowledgement, p. 3609, line 26: It is David Parrish, not Martin Parrish, at the Aeronomy Laboratory.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3589, 2003.

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