

The paper "Spectroscopic real-time monitoring of NO<sub>2</sub> for city scale modelling", by Hundt et al, reports on mobile NO<sub>2</sub> measurements performed in the city of Zurich with a QCL spectrometer placed on a tram roof. Data are analyzed and compared to AQM sites where CLDs measurements are available. A statistical data analysis is presented to provide NO<sub>2</sub> concentration maps.

This work is interesting however it is hard for a reader to judge because some clue information are missing as listed in the general comments. The manuscript will need a major revision to be considered for publication in AMT.

### **I) General comments :**

1) The need of mobile measurements is well introduced but an overview of available instruments is missing (commercial or laboratory prototypes) . Here CLDs and electrochemical sensors are mentioned and references are given for instruments previously developed by the authors in the MIR. However there is no mention to the many different spectroscopy techniques used in the MIR as well as in the blue spectral region with ECDL or LED. Please compare the performances of the spectrometer developed with these previous works that have to be cited in this paper.

2) P2,L28 " The portable instrument was specifically built for this campaign". Figure 1 is just a scheme that could fit to all multipass spectrometer. The reader expects a detailed description of the technique (ICW mode) and the set-up to understand how the good performances reported in the following are reached. A spectra recorded by the instrument should be shown. Please, detail the "Quantum cascade laser spectrometer" section in this way (see detailed comments in the following).

3) The statistical model is based on average NO<sub>2</sub> measurements during 3s (P6,L18- P9,L15). Note that the response time of the instrument is longer : 9.6s. Averaging at this time scale should be performed prior to analysis.

4) NO<sub>2</sub> concentration is expected to be sensitive to the solar activity, NO, and O<sub>3</sub> concentrations, and to ambient temperature and humidity. Variations of NO<sub>2</sub> measurements should be studied according to these different parameters. It is hard to believe that all these parameter effects are removed by the offset correction.

5) QCLAS measurements is compared to CLDs measurements taken as reference values. Specification of the CLDs are expected : sensitivity, absolute accuracy, intercomparison of the CLDs...

### **II) Detailed comments :**

**a) In the abstract and the introduction :** please mentioned the technique used and the wavenumber of the laser.

#### **b) Introduction :**

1) This work is dedicated to urban monitoring, please specify the NO<sub>2</sub> concentration range to clarify the instrument performances required in term of detection limit.

2) Insert references to previous NO<sub>2</sub> spectrometers (see General comments)

3) Please gives accurate numbers concerning instrument performances :

P1, L30 "accurate" ?

P2, L9 : "not fast enough"?

P2 L11 "not short enough"

P2 L13 "relatively high"

P2 L16 : "high sampling rates"

**c) 1.1 QCL spectrometer :**

1) The iCW operation was published in Fischer 2014, reporting on a different instrument that operates at  $7.7\mu\text{m}$ . In this paper, a short description of this technique is expected and a detailed description of the optical set-up at  $6.25\mu\text{m}$  as well. I could not find mention to an optical etalon : How is obtained the frequency linearization ?

2) Please show a recorded spectra around  $1600\text{ cm}^{-1}$ . How many  $\text{NO}_2$  lines do you monitor ? Is the spectra large enough to monitor water ?

3) In figure 1 : the detailed optical scheme would be more useful than the schematic content of the instrument

4) P3, L17 : please specify that the  $100\mu\text{m}$  orifice is placed at the inlet of the cellule and give the corresponding flow value.

5) P3 L17-19 : The response time is a main concern. It is measured to be equal to 9.6s by switching from  $\text{NO}_2$  free air to  $\text{NO}_2$  sample. Is this really consistent with the complete exchange time of the cell according to the flow and pressure values ?

6) P3L22 : what is the acquisition rate ?

7) P3, L23-25: Concerning the fitting procedure : Please indicates which parameters are fixed (such as P probably) or left variable. The instrument not being temperature stabilized, what about the temperature parameter ? Is the path length deduced fixed?

8) Water absorption has strong absorption lines around  $1600\text{cm}^{-1}$ , and its concentration is changing during tram measurements. How is that taken into account ? Do you include a fit of water line to monitor it ? is it included in the base line?

9) P3,L29: please make explicit what you call "zero-point offset".

10 ) The dynamic range of the instrument should be given somewhere in the paper

**d)2.3.  $\text{NO}_2$  concentration measurements at fixed sites**

Indicates that it is performed by CLD. Please give the specifications of these instruments (see general comments).

e) P5 L17 : "  $\text{NO}_2$  concentrations encountered in Zurich are moderate compared to other cities." Please specify here the typical range.

**f) 2.5 Statistical modelling**

1) P6, L18 :  $\text{NO}_2$  concentrations should be averaged to 9.6s, the response time (see general comments)

2) Please provide the reference (Breinam1984) concerning the "regression tree approach" at first mention: P6, L32

**g) 3.1. Instrument performance and stability**

1) The Allan deviation is expected to increase at long integration time due to technical drifts or ambient parameters change. Can the author provide a longer set of measurements to show when it appears?

2) In figure 3, in the upper panel can you confirm that Dark-Noise measurements are the upper one or is there a color inversion? I would understand an offset of about 1.5 ppb is the measured in "NO<sub>2</sub> free air" rather than Dark-Noise, from a non perfect filtering or other effects.

3) P8, L1 : Concerning the "measurement strategy" I understand how the drift is corrected but this process relies on the zero NO<sub>2</sub> gas production in-situ. Did you test that there is no NO<sub>2</sub> trace in this gas? I would expect some NO<sub>2</sub> trace at the ppb level (according to fig 1). And the residual concentration is expected to depend on NO<sub>2</sub> concentration in ambient air. Furthermore, the sensitivity to water concentration should be investigated. Can you provide some details?

Another aspect should be detailed : how is calibrated the correction, on which reference measurement? Did you performed a referenced measurement prior in-situ measurements? Did you repeat it during the measurement campaign?

4) Allan deviation only demonstrate the stability of the instrument but not the accuracy. Caption of figure 3 is to be modified and 2 P7, L24-25 : "precision" should be replaced by "stability"

5) P8, L8-14 : I would expect the correction factor to change in time due to passivation of the surfaces or water interferences : the calibration performed in June is probably different during winter. Did you test this point?

6) The correction factor of 1.2 is applied. But how was the instrument firstly calibrated?

7) P9L2: Please specify the absolute accuracy of the CLD to justify the 1 ppb accuracy of the instrument.

8) P9L15-19 : as mentioned previously, measurements should be averaged to 9.6s resulting in a longer route segment.

**h) 3.2 Intra-urban and temporal variation in NO<sub>2</sub> concentration**

1) Fig 6 : in the caption please clarify which AQM station (STA or SCH) is close to which terminal stations (Seebach or Triemli)

2) P11L7 : again measurements should be averaged to 9.6s.

3) P11,L15-17: I don't understand why "this analysis clearly shows the advantage of a mobile measurement device that does not rely on fixed sites for the determination of its calibration"

parameters". I would assume that the main reason for the discrepancies observed with SCH measurements is due to the difference of averaging time : 9.6s response time for the QCLAS and 1min for the CLD. SCH being impacted by heavier traffic than STA , fast events of strong pollution will have more impact.

4) Figure 8 : What is the meaning of points after the 7th stop ?

5) In this section, temporal variation of NO<sub>2</sub> should be studied as a function of other parameters : NO, O<sub>3</sub>, H<sub>2</sub>O,T,solar activity (see general comments).

**i) Model validation by means of three seconds NO<sub>2</sub> measurements from the tram**

Should be performed with 9.6s averaging measurements

**j) 3.3.2 Model validation by means of 30 minutes mean NO<sub>2</sub> concentrations at fixed AQM stations**

1) Fig 10: change the color code of NO<sub>2</sub> concentration to allow to distinguish between very low concentrations (presently in white) from the zone where the model cannot compute concentration. Presently, the spatial coverage is not clear.

2) P17, L29-30 : " The obtained RMSE values range between 1.5 ppb at HEU (elevated background site) and 7.5 ppb at RGS (roadside)." Given values are not consistent with Fig 11.

3) What is the error/shift value that can be attributed to the absolute calibration of the different CLDs ? Were they regularly calibrated on the same gas sample ?

**k) Conclusion**

1)P20L2 : " QCLAS spectrometer that operated autonomously.... over a period of four months" should be added a mention to the intervention for optical alignment

2) Please indicate clearly that the 2ppb agreement is obtained after a correction factor of 1.2 derived from these measurements

3) 3s to be replaced by 9.6s.

**Typesetting:**

- Section 2.1 is missing
- please enlarge the text in the graphs or improve the resolution to allow easy reading in figure 5,6 ,7,9,10 and 11
- P1 L10,L22 and everywhere: "measures" should be replaced by "measurements" ?
- P4L13 : a word is missing "measurements are available THEY are depicted..."
- P16L9-10 : " The frequency a particular variable is effectively used"