

Continuous Measurements of H₂ for the Estimation of Anthropogenic Sources



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Introduction

Molecular hydrogen (H₂) is one of the most abundant trace gases in the atmosphere. However, only little attention has been paid until now to investigate the sources and sinks as well as the role of H₂ that it plays in atmospheric chemistry. At present, more and more ideas evoke that praise the hydrogen economy (the use of hydrogen to store and carry energy) for replacing the fossil fuel energy system. Before the hydrogen economy becomes reality, the impact of the different emissions on the whole atmosphere has to be investigated in detail and understanding the current budget is a necessity. Since November 2002, continuous measurements of H₂ and carbon monoxide are performed at a road site near Zurich. The aim of this study is to obtain a reliable data set for the modelling of actual emissions related to fossil fuel burning and especially road traffic.

Study Site/ Experimental

A commercial gas chromatograph (Reduction Gas Analyzer RGA3 from Trace Analytical, Inc.) is used to measure molecular hydrogen (H₂) and carbon monoxide (CO) by hot mercuric oxide reduction and UV absorption detection. The instrument is located in Duebendorf, in an industrial and densely populated area near Zurich. A main road passes the station 150 m south, a highway around 750 m north of the sampling site.

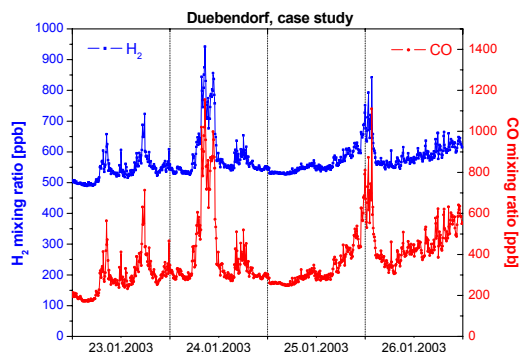


Fig. 1: Case study illustrating the identical main sources for H₂ and CO (predominantly traffic) at the sampling site.

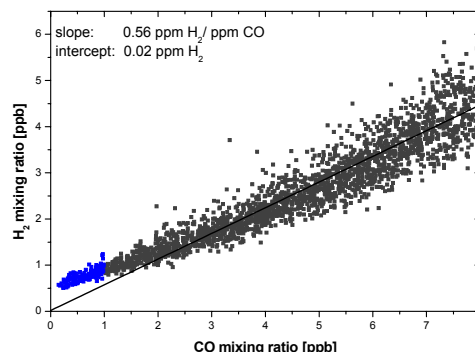


Fig. 2: H₂ versus CO in the Gubrist tunnel, in October and November 2002. Only data with considerable traffic influence (CO > 1 ppm; data in grey) were considered for the regression. Above 8 ppm of CO, the detector reached its saturation.

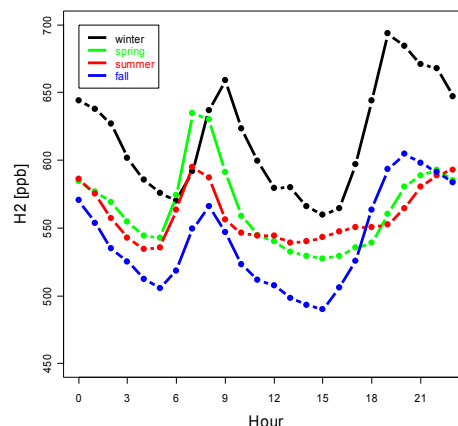


Fig. 3: Mean daily cycle of H₂ for each season; data from November 2002 to November 2004.

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Results

Fig. 1 shows a 4-days case study of H₂ and CO measurements in Duebendorf. The time series of both compounds are strongly correlated reflecting similar sources (predominantly traffic) at this sampling site. The data compare well with results obtained in a highway tunnel near Zurich in October and November 2002 (see Fig. 2) taken with the same instrument where a slope of 0.56 ppm H₂ per ppm CO was obtained. These data corroborate the considerable importance of traffic as a major source of H₂.

Fig. 3 presents the mean daily H₂ cycle for each season. Again, the bimodal structure reflects the enhanced traffic densities during the rush hours. In general, the lowest H₂ mixing ratios were measured in fall in agreement with results from flask samples collected at 50 locations all over the world between 1991 and 1996 [1]. This H₂ depletion can be explained by the stronger H₂ uptake by soils under warm and dry conditions [2] and the enhanced sink strength due to OH oxidation in summer.

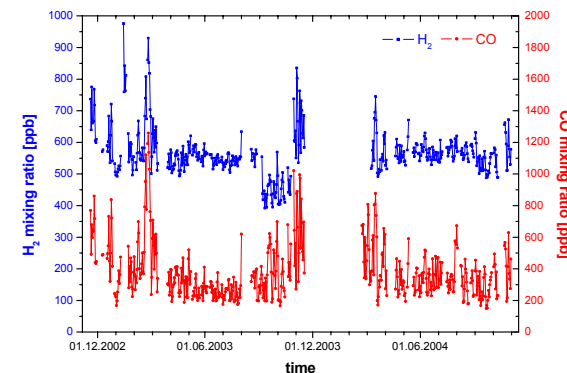


Fig. 4: Time series of daily averaged H₂ and CO mixing ratios for the whole investigated period.

Fig. 4 illustrates daily averages of the H₂ and CO mixing ratios from November 2002 to November 2004. Remarkably low H₂ concentrations were measured in fall 2003 after the exceptionally dry and warm summer. As the CO concentrations are not depleted at the same time and the CO data agree well with the continuous CO measurements in Zurich (ca. 7 km south-west of Duebendorf), an instrumental artefact can be excluded and the low H₂ mixing ratios can be most likely attributed to a more pronounced H₂ sink strength (soil uptake).

Outlook

Further evaluations of the anthropogenic H₂ emissions were conducted during a second tunnel study in November/December 2004 and will be compared with chassis dynamometer tests measuring the direct H₂ emissions of cars.

References

- [1] Novelli et al., JGR 104 (D23), 30'427-30'444 (1999).
- [2] Schuler & Conrad, Biol. Fert. Soils 12, 127-130 (1991).