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Interactive comment on "The use of tunnel concentration profile data to determine the ratio of NO_2/NO_x directly emitted from vehicles" *by* X. Yao et al.

X. Yao et al.

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1.This paper reports measurements of longitudinal NO, NO2 and O3 profiles in two tunnels of c.4 km in length in Hong Kong from which the authors conclude that the primary on-road vehicular NO2/NOx ratio was less than 2%, substantially lower than is usually quoted for this ratio. Reliable information on this primary ratio is important for accurate modelling of ambient NO2, which is subject to air quality standards in most countries. The general aim of the work undertaken is thus highly relevant and within the scope of ACP.

However, the amount of data and detail provided in the paper are both relatively

slight and consequently I have reservations that there is sufficient substantiation of (or caveats to) the main conclusion to justify publication at this stage. The authors attribute observed concentrations differences between the north and south-bound tubes of the Tai Lam Tunnel to different fuel characteristics which shows that fuel/fleet characteristics are important in interpreting and reporting data.

Response: Details on on-road vehicles in Hong Kong and MAP measurements are added in the revision. We believe that the amount of data provided in this study is sufficient to support our conclusion. We presented measurements in two tunnels (four tubes). In each tube, five runs were carried out and the trends of concentration profiles of air pollutants are repeatable not only in the same tube at different times, but also in the different tubes. We also used year-long on-road measurements and a 24-h continuous on-road measurement to substantiate our discussion. We are not that interested in reporting the definitive NO2/NOX ratio, we are more interested in demonstrating that measuring direct vehicular NO2/NOX ratio in the presence of ozone has a lot of problems.

2.I have the following additional points: (1) The authors cite literature from 1979 (Hilliard and Wheeler) and 1983 (Lenner and Lindqvist) for previous and comparator presentations of vehicle primary NO2/NOx ratio. Are such comparatively old measurements relevant to the modern vehicle fleet given the huge changes in engine and exhaust technology in the lost 25 years?

Response: Hilliard and Wheeler in 1979 reported that the primary NO2/NOx ratio to be 5%. In the past decades, due to the continuous introduction of new and improved control technology, NOx emitted from vehicles decreased, especially for gasoline vehicles. However, recent papers reported vehicle primary NO2/NOx ratios to be substantially higher than 5% and sometimes up to 30% (Introduction Section in ACPD-2005-0344). Theoretically, NO is the main product of combustion processes, to measure directly emitted NO2, it is important to do so in an environment that is absent of secondary reactions triggered by oxidants such as ozone to transform the NO. It is, therefore

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paramount to find a site where ozone does not exist and there are plenty of vehicles moving around. The vehicular NO2/NOx ratio does not depend on the absolute amount of NO that is emitted but the ratio of the engine generated NO and NO2. From our experience, tunnels fit this requirement. We initiated this study because we have a mobile air monitoring platform (MAP) which allows us to take air pollutant measurements in just about anywhere we can drive the vehicle. Our results show that the measurements in open air and at the end section of tunnels lead to substantial overestimation of the ratio.

(2) To what extent are measurements from the mobile platform genuinely representative of the "ambient" NO, NO2 and O3 concentrations at the point in the tunnel at which the measurement is taken, as opposed to being influenced by emissions from the vehicle in front?

Response: See our response to Comment 4, Reviewer 1. For more details also refer to Yao et al., (AST, 2005).

(3) The stated ventilation rates for both tunnels are large which I crudely estimate might give rise to several air exchanges per hour. Thus the effect of external air introduced into the tunnel may be greater than the authors acknowledge. The authors also provide an estimate for an in-tunnel wind speed of Ÿ5 m s-1 (p12729) but make no comment on the direction of this wind flow: is the wind flow in the tunnel moving with or against traffic flow? is it moving outwards in both directions from the centre of the tunnel?

Response: The inflow of fresh ambient air can influence the concentration profiles of NO, NO2 and O3 in the tunnel. The influence is discussed in the Results Section and no sources of these pollutants are missing in our discussion. The mechanical ventilation in tunnel is always in the direction traffic flow. Furthermore, the piston-effect caused by the moving vehicles is also in the direction of the traffic. More information about the tunnel has been added in the revision.

(4) The authors should investigate in more detail a total OX (NO2 + O3) approach of

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evaluating their data as described by Clapp and Jenkin, Atmos. Environ. (2001).

Response: See our response to Comment 6, Reviewer 1.

(5) The authors discount the termolecular reaction of NO+NO+O2, yet the NO concentrations presented are likely high enough for this reaction to contribute to generation of NO2.

Response: Admittedly, we are not able to experimentally differentiate between NO2 directly emitted or transformed, and admittedly, we considered only the most important and fastest secondary reaction between NO and O3. Our measurements show that in the mid-section of the tunnels where O3 is minimum, secondary NO2 is also the smallest, and the measured NO2/NOx ratio in this work is substantially lower than that in the literature. Had we been able to exclude the NO+NO+O2 reaction, the ratio would be even smaller. This is why we reported the 2% as the upper limit of vehicular emitted NO2/NOx. The comment provided by the reviewer, in essence, fortifies our argument.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 12723, 2005.