

## ***Interactive comment on “Three-year observations of halocarbons at the Nepal Climate Observatory at Pyramid (NCO-P, 5079 m a.s.l.) on the Himalayan range” by M. Maione et al.***

**M. Maione et al.**

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Referee comment: Abstract: The abstract mainly summarizes the structure of the paper but does not highlight the main conclusions of the study sufficiently. Please try to be more specific both qualitatively and quantitatively for the final version.

Reply: We have modified the abstract adding more specific information —

Referee comment Section 1, Introduction, p. 22341, line 20-27: the description of emissions sources of methyl halides needs major revision. To my knowledge terrestrial plants are not considered as a significant source for CH<sub>3</sub>Br as mentioned here, maybe the authors refer to CH<sub>3</sub>Cl here; please check the reference “Yokouchi et al.,

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2002”, for CH<sub>3</sub>Br it should be “Yokouchi et al., 2000”.

Reply: The emission of CH<sub>3</sub>Br from terrestrial plants has been reported by “Yokouchi Y., D. Toom-Sauntry, K. Yazawa, T. Inagaki and T. Tamaru, Recent decline of methyl bromide in the troposphere, *Atmospheric Environment* 36 (2002), pp. 4985–4989” and also by “Rhew, R. C., B. R. Miller, M. K. Vollmer, and R. F. Weiss (2001), Shrubland fluxes of methyl bromide and methyl chloride, *J. Geophys. Res.*, 106, 20,875–20,882, doi:10.1029/2001JD000413” and many other references one the most recent being “Rhew, R.C., C. Chen, Y.A. Teh and D. Baldocchi, Gross fluxes of methyl chloride and methyl bromide in a California oak–savanna ecosystem, *Atmospheric Environment* 44 (16) (2010), pp. 2054–2061. We will incorporate these references. —

Referee comment Section 1, Introduction, p. 22341, line 20-27: Also for CH<sub>3</sub>Cl the oceans are not mentioned as major source.

Reply: we will include the citation of the paper “Moore, R. M., Groszko, W., and Niven, S. J.: Ocean-atmosphere exchange of methyl chloride: Results from NW Atlantic and Pacific Ocean studies, *J. Geophys. Res.*, 101(C12), 28529– 28538, 1996” and incorporate the reference. —

Referee comment Section 1, Introduction, p. 22341, line 20-27: In addition, when describing CH<sub>3</sub>Cl it is important to note that a major emission source from biomass burning in Asia is the use of biofuels (wood, agricultural waste and dung), especially in the Indian subcontinent. Here I like to refer to the INDOEX project....

Reply: we thank the reviewer for signalling these important references, and we have changed the text accordingly adding the following references : Andreae, M. O. and P. Merlet 2001, Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles*, 15/ 4, 955–966; Scheeren, H.A., J. Lelieveld, J.A. de Gouw, C. van der Veen and H. Fischer 2002, Methyl chloride and other chlorocarbons in polluted air during INDOEX, *Journal of Geophysical Research* 107 (D19), 8015, doi:10.1029/2001JD001121. The suggested reference to the paper of Streets et al,

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2003 has not been included because dos nor specifically refer to Methyl Halides. —

Referee comment Section 1. Introduction, line 27-29: I And the description of the objectives of the study unclear. They write: “The objective of the study is to quantify trends in the atmospheric record and identify more precisely source regions and origin of halocarbons in the High Himalaya, in particular resulting from biomass burning emissions and tropical vegetation.” First of all, the majority of the halocarbons in this study are merely of anthropogenic origin apart from the methyl halides, which also have strong anthropogenic sources such as biofuel burning. Then the most important potential source region for pollution to the NCO-P station, apart from local forest fires, appears to be the Indo Gangetic Plains, as already described by Bonasoni et al., ACP, 10, 2010. Please try to be more specific here.

Reply: we agree that this last sentence can be unclear. Nonetheless we have clearly specified in the previous paragraph that the study is mainly focused on the study of anthropogenic species. Therefore we will modify this last sentence for clarification: —

Referee comment Section 2, Methods, first paragraph: for clarity I feel the authors should include a few lines highlighting the meteorological conditions at the site.

Reply: we modify the text accordingly to the referees' comment and add the reference Bollasina, M., Bertolani, L., Tartari, G.: Meteorological observations at high altitude in Khumbu Valley, Nepal Himalayas, 1994–1999, Bull. Glaciol. Res., 19, 1–11, 2002. —

Referee comment Section 2.1 Analytical Methodology, line 14, “The optimization. . . is reported elsewhere (Maione et al., 2004).”: here I would like to see a summary of the instrument performance for the interest of the reader by writing “The optimization. . . is reported elsewhere in detail (Maione et al., 2004). Here we briefly summarize the main performance characteristics of the GS-MS system. . . . etc.”, then for the following sentence I would recommend a more specific description of what the “improved accuracy of the analytical data of the second part of the data set” actually means. Line 19-21: please include the accuracy of the used scales here.

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Reply: we modify the text accordingly to the referees' comments adding more details and a table reporting specific data on accuracy. —

Referee comment Section 2.2 Baseline determination, first paragraph: try to avoid vague descriptions like "... "old", well mixed. . . ", "... by long lifetimes", and "... fresh, not well mixed . . . Instead, try to be more specific using phrases like "...of the order of months to years. . . "for "old", "typically of the order of a few days" for "fresh", etc. Section 2.2 Baseline determination, second and third paragraph: I find the description of the statistical method to alter the data a bit unclear. Please try describing the method more clearly and in greater detail. Later the authors refer to Giostra, 2010 where the data altering method for the Mt. Cimone data are described. Please indicate in more detail where this reference can be found (in a journal, proceeding or via personal contact of the author?) or provide, if possible, another reference.

Reply: we have modified the text according to the referee's suggestions. —

Referee comment: Section 3 Results and discussion, first section: also here try to be more specific on characterizing atmospheric lifetimes instead of just writing "long atmospheric lifetimes" and "relatively short lifetimes".

Reply: we have modified the text —

Referee comment Section 3.1.1, p. 22347, last sentence of this section: what lacks here is a discussion on the meaning of Figure 4. Please elaborate.

Reply: the discussion has been added —

Referee comment: Section 3.1.3, line 7-16, table 3 discussion: include the coefficient of correlation  $r^2$  in table 3 to show the significance of the calculated trends. In addition, I think it would be useful to include global mean trends from Clerbaux and Cunnold (WMO report no. 50, 2007) to where the authors refer to in the text. The authors write about data in italics or bold to denote the level of significance; this notation is however absent in table 3.

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Reply: The comment of the referee is unclear. In the table we report a trend whose significance is represented by standard error (standard deviation of the mean) . The italics and bold notation disappeared formatting the PDF document. A column has been added reporting trends from Clerbaux and Cunnold 2007. —

Referee comment Section 3.1.4, p. 22349, third paragraph from line 15 and on: I ãAnd the paragraph on potential source regions based on the analysis of LAGRANTO 5-day back-trajectories unclear and in lack of detail. Please provide more detail about the methodology by Maione at al. (Sci. Total Environm., 391, 2008) and explain better the meaning of ãAgure 6. The graphical quality of ãAgure 6 should be improved (see Technical comments for further explanation). The authors should elaborate more on the general pattern of atmospheric transport and related source areas of pollution that are relevant for the site as described by Bonasoni et al. (ACP, 10, 2010).

Reply: we have modified the text adding a more detailed description of the method and we have modified Figure 6. Concerning the request to elaborate more on the general pattern of atmospheric transport and related source areas of pollution that are relevant for the site as described by Bonasoni et al. (ACP, 10, 2010), our reply is that the analysis reported in figure 6 is referred to a data set much smaller than that available for other parameters analysed by Bonasoni et al. 2010 based on continuous measurements. —

Referee comment Section 3.2 Methyl halides: as already mentioned in my earlier comment on section 1, the discussion on the methyl halides is incomplete and ignores the importance of biofuel emission in the Indo Gangetic Plains. Please elaborate.

Reply: we have modified the text also giving reference to the INDOEX campaign results. —

Referee comment Section 4. page 22351, Conclusions, line 7: the authors write that “Halocarbons measurements at the site provide a different picture with respect to time series from other global background stations. . . ”. Be clearer here on what these

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differences are and what the more general characteristics of the observed time series are.

Reply: we feel that this comment is not relevant because we have described in the following paragraph the peculiarity of the site of being close to sources of Montreal gases. —

Referee comment Line 12-14, “For those compounds characterized. . . tropical vegetation and biomass burning.”: this study shows no evidence for a significant contribution of tropical vegetation to methyl halide emissions. Hence this remark is not relevant in the conclusions and should therefore left out as such. Next the authors mention the observation of elevated CH<sub>3</sub>Cl from local forest fires. This is repeated further on in line 22-24. Please avoid repetition here. Moreover, the specific forest fires cases should be seen separately from the fact that the baseline CH<sub>3</sub>Cl appears to be relatively high which could also reflect the influence of biofuel emissions from the Indo Gangetic Plains, as already discussed earlier. Line 21 and next: add “, notably the dense populated Indo Gangetic Plains.” after “..of the Indian sub-continent”. In the next sentence the authors write “The atmospheric circulation. . . ..of the contribution from China”. Is this conclusion based on the general study of the meteorological conditions at the site by Bonasoni et al. (2010) or is it related to the specific conditions during the sampling days? Please clarify.

Reply: We have modified the text accordingly. —

Technical comments: have all been accepted and incorporated in the text —

The revised manuscript is attached as well as the revised figures (Figure 4, 6 and 7)

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/10/C13113/2011/acpd-10-C13113-2011-supplement.pdf>

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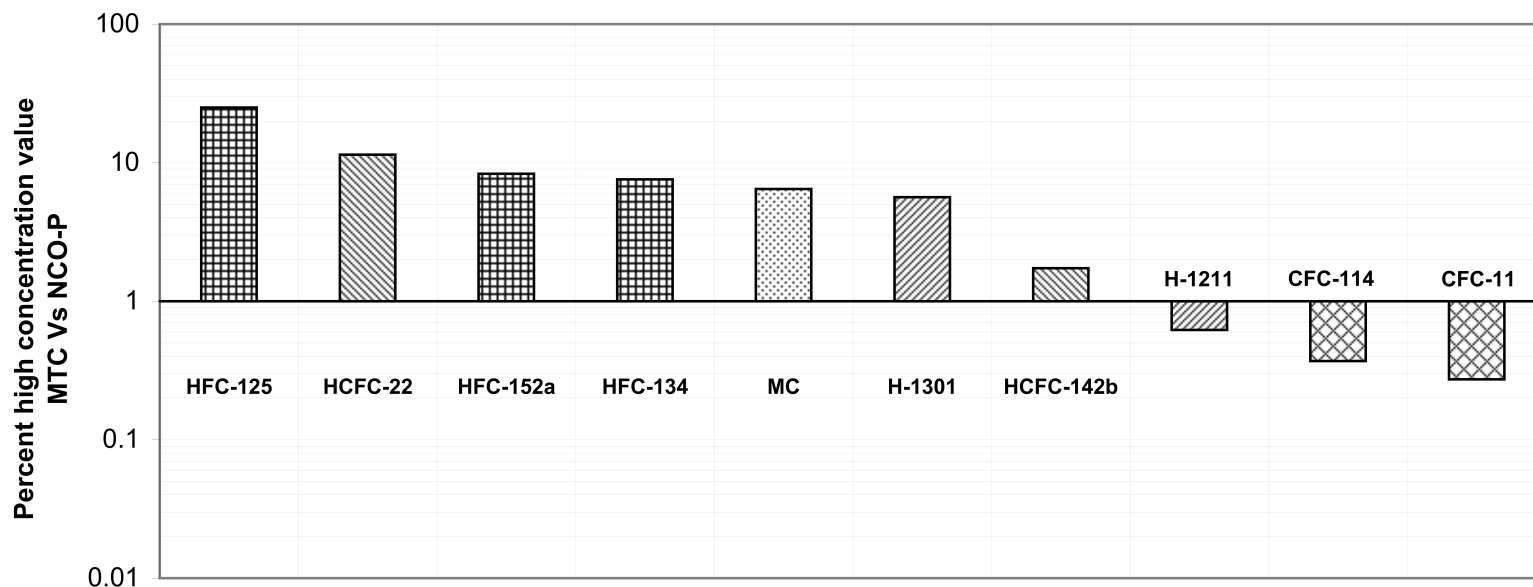
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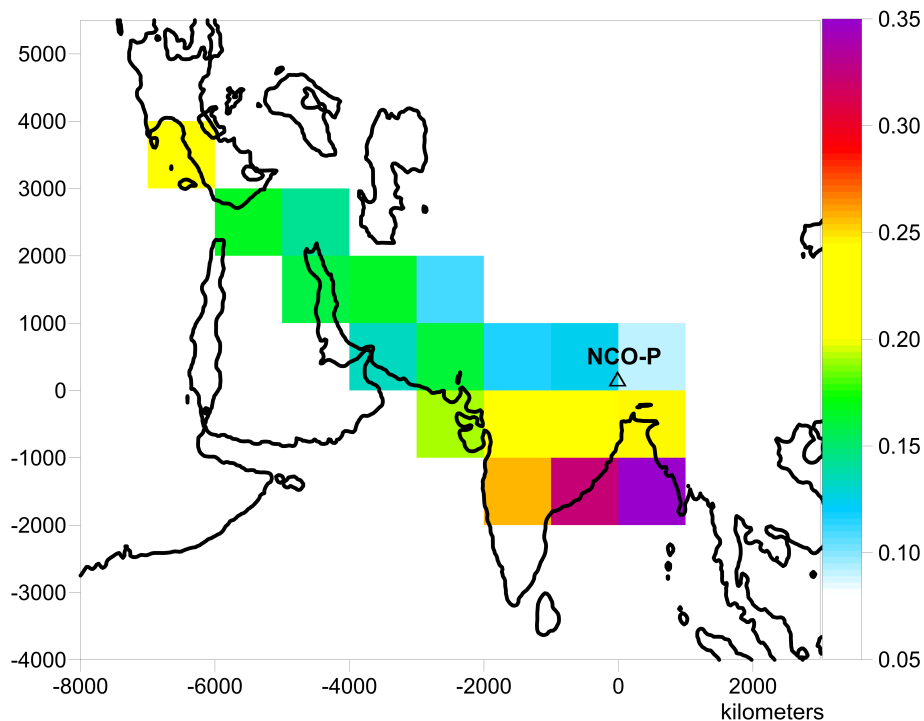
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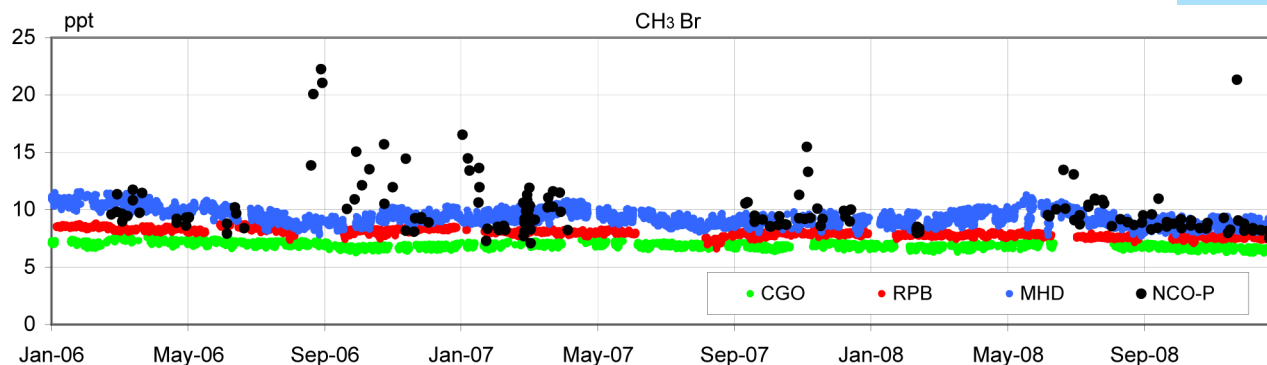
**Fig. 1.** Figure 4: Ratios of the occurrence of high concentration values at MTC with respect to NCO-P. Chemically homogeneous classes of compounds are denoted by identical shading.

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**Fig. 2.** Figure 6. Map of conditional probability of potential sources of anthropogenic halo-carbons, based on observations at NCO-P observatory. The scale represents the fraction of polluted trajectories over

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**Fig. 3.** Figure 7. CH<sub>3</sub>Br measured at NCO-P (all data) compared with baseline data at at MHD (blue), RPB (red) and CGO (green).

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