

## ***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.***

### **Anonymous Referee #1**

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### **General Comments**

The authors present important results from the first three-year of observations of a comprehensive set of halogenated hydrocarbons from air samples collected at the NCO-P high altitude station in the Himalayas. The presented data provide a valuable insight in how background conditions in central Asia evolve in light of the rapid socio-economic development of Asia. The authors make a useful attempt to compare filtered time series from NCO-P to a couple of relevant background stations and they discuss atmospheric trends.

Overall, the quality of the English is sufficient and the explanations are concise. How-

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ever, in a number of cases the writing is rather vague or confusing, notably explanations are sometimes too brief or incomplete relying only on a reference to other work. The abstract is too descriptive and lacks some (quantitative) conclusions. The objective of the study needs to be better specified. In addition, the quality of a number of graphs is insufficient.

On the whole I feel the paper is interesting enough for publication in ACP but after considerable improvements on a number of points presented below to make the paper clearer, better structured and more attractive to the reader.

### Specific comments

**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.

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  $\text{CH}_3\text{Br}$  as mentioned here, maybe the authors refer to  $\text{CH}_3\text{Cl}$  here; please check the reference “Yokouchi et al., 2002”, for  $\text{CH}_3\text{Br}$  it should be “Yokouchi et al., 2000”. Also for  $\text{CH}_3\text{Cl}$  the oceans are not mentioned as major source. In addition, when describing  $\text{CH}_3\text{Cl}$  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. The INDOEX project conducted in 1999 over the Indian Ocean was the first large scale field experiment where the so-called Atmospheric Brown Cloud from the Indian sub-continent was thoroughly investigated over the Indian Ocean (e.g., see Lelieveld et al., Science, 291, 2001, and references therein) From the INDOEX campaign it became apparent that the major contribution to biomass burning as a source of trace gases and aerosols was not coming from natural fires but rather from the use of biofuels (agricultural waste, wood fuels, and dung) notably in air masses originating over the Indo Gangetic Plains

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(e.g., see Scheeren et al., JGR, 107, 2002; and Streets et al., Global Biogeochem. Cycles, 17(4), 1099, 2003; and references therein). Hence, the authors should do a better job in referring to this important source category. Overall comment, please check the estimates and references for the CH<sub>3</sub>Cl and CH<sub>3</sub>Br sources and sinks, e.g. with a recent paper by Gebhardt et al., *Atmos. Chem. Phys.*, 8, 3185–3197, 2008, and references therein.

Section 1. Introduction, line 27-29: I find 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.

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

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.

Section 2.2 Baseline determination, first paragraph: try to avoid vague descriptions

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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 filter 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 filtering 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.

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”.

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.

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.

Section 3.1.4, p. 22349, third paragraph from line 15 and on: I find 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 et al. (Sci. Total Environ., 391, 2008) and explain better the meaning of figure 6. The graphical quality of figure 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).

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.

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

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  $\text{CH}_3\text{Cl}$  from local forest fires. This is repeated further on in line 22-24. Please avoid repetition here. Moreover, the specific forest fire cases should be seen separately from the fact that the baseline  $\text{CH}_3\text{Cl}$  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.

### Technical comments

p. 22341, line 25: write  $\text{CH}_3\text{Cl}$  for  $\text{CH}_3\text{Cl}$ .

p. 22346, line 7: change “. . ., following the procedure described above.” to “. . ., following the statistical procedure described in section 2.2.”

p. 22347, line 21: add an estimate and a reference to the lifetime of inter-hemispheric

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exchange.

p. 22347, line 23: change “Data clearly show...” to “The HCF-152a data clearly show...”.

p. 22348, section 3.1.3 Atmospheric trends, line 1-7: The sentence is too long and not well structured. Remove “to attempt”; start a new sentence after “NCO-P:”

p. 22349, line 19: rewrite the phrase to “A preliminary attempt has consisted in calculating...”, to “The frequency of occurrence of polluted trajectories....period of the measurement has been calculated.”

p. 22349, line 22: rewrite “..the localization of potential sources...” to “the location of potential source regions...”

p. 22361: figure 3: please use a color coding instead of shadings to identify chemically homogeneous classes.

p. 22362, figure 4: where the bar legend overlaps with the bar graphic it is difficult to read. Please use a color coding to discriminate between chemically homogeneous classes.

p. 22365, figure 6: the graphical quality of the land boundaries is rather poor. Please use a higher resolution map here and zoom in on the region of interest. In the figure caption change “Halocarbons” to “halocarbons”. Please explain the meaning of the color legend in the figure caption.

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 22339, 2010.

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