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Interactive Comment

Interactive comment on "A multimethodological approach to study the spatial distribution of air pollution in an Alpine valley during wintertime" *by* R. Schnitzhofer et al.

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The authors want to thank the reviewers for their good comments and suggestions that helped to improve the quality of the manuscript.

Referee #1 (A. Stohl)

Section 3.2: When describing the synoptic situation, adding a weather map would be a good idea.

REPLY: We feel that the paper would not significantly benefit from an additional figure showing a weather map.

P3990, L15: You mention 10 vertical VOC profiles but Fig. 4 suggests there were only 4.

REPLY: 10 vertical VOC profiles have been measured, but only the 4, plotted in Fig. 5 are marked by a grey bar in Fig. 4. Missing data in Fig. 5 indicate the times of the other 6 soundings.

P3990, L26: You say that pollutants are trapped in cold pools. However, is there a cold pool at the observation site? Is it located in a topographic depression?

REPLY: Maybe cold pool is the wrong term in this case. The sentence has been rewritten in the revised version of the manuscript. "Near ground emitted pollutants are trapped in the stable surface boundary layer."

P3994, L16: You mention titration of O3 by NO. Have you plotted Ox=O3+NO2? This quantity should be conserved in the presence of titration.

REPLY: Ox was measured and the concentration varies by about 20% throughout the whole valley in the afternoon of 1 February 2006. Compared to the observed changes of a factor of 50 for O3 and NO2 throughout the valley atmosphere the Ox signal is quite constant.

P3996, L14 and conclusions: you say that pollutants did not significantly overshoot the main crest and, thus, pollution was not exported from the valley atmosphere. Is this really true? The lidar backscatter signal shown in Fig. 10 does show that enhanced backscatter can be found also above the crest. The signal appears weaker than at lower levels, however, this is to be expected even when substantial export does occur. According to Henne et al. (2004), there can be an injection layer just above crest height (see their Fig. 13), which is influenced by the synoptic flow. Even though their picture is more valid for summer-time conditions, if slope winds are active (as you show), it

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may exist also in winter. Wind speeds in this layer should be much higher than in the valley atmosphere. Thus, as soon as pollution is injected into this layer by the slope winds, there would be rapid dilution by the stronger winds aloft and this would reduce the lidar backscatter signal above crest height compared to the signal seen lower down. However, the weaker signal does not necessarily mean that the mass flux out of the valley was low! You could probably estimate mass fluxes by looking at pollutant concentrations near crest height and the wind speeds at these altitudes. The conclusion that there was no pollution export at all from the valley is probably wrong in any case because the aircraft in-situ data and, especially, the lidar data does show some enhancements near and above crest height on the sun-exposed side of the valley.

REPLY: Figures 9 and 10 only show a small section of the valley (+- 4 km from the center of the valley), a wider section is plotted in Figure 2 (+- 10 km). The lowest crest elevation within the 20x6 km box plotted in Figure 1 is about 1600 m asl (above sea level) and the mean crest level is close to 2000 m asl. The terrain is still increasing further northwest ending at 1900 m asl at the lowest mountain pass withinin the investigation area. Elevated concentration of air pollutants could be found up to 1900 m asl (compare Fig. 10) and although there were no strong winds aloft on 1. February 2006 ($<$ 2 m/s at 2500 m asl), which would have led to a rapid dilution, some pollution export might have occurred. About 20 km in the valley downward direction there is the Achenseepass, with 941 m asl the lowest pass in the region. Significant air pollution transport is expected to occur there. However, we agree that pollution export most likely happens even during wintertime, when slope winds are well developed and stronger winds exist above crest height. Therefore we rewrote the respective parts in the abstract, in section 3.2.5, and in the conclusion. "This vertical mixing due to thermally or dynamically driven slope winds reduces the concentration of air pollutants at the bottom of the valley and causes the formation of elevated pollution layers."

"The export of air pollutants from the valley into the free troposphere could not be

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quantified with this experimental setup but is expected to be small on 1 February 2006, since the vertical transport did not significantly overshoot the main crest and a reverse of the slope wind circulation after sunset partly transported the polluted air back to the valley bottom (Harnisch et al. 2008). However, a significant export of air pollutants might occur during situations with even more active slope winds and stronger winds above crest height (compare 24 January 2006; Gohm et al. 2009), via a process described by (Henne et al. 2004)"

"Despite the stable stratification of the valley atmosphere, a vertical transport of air pollutants via slope winds reduces their concentration at the bottom of the valley, causes the formation of elevated pollution layers and even exports pollutants from the valley during specific situations in wintertime."

Fig. 3: Explain why the benzene and acetone data are missing during the intensive campaign periods. I assume the PTRMS was used for the balloon but it is somewhat irritating that data are missing exactly in the periods you are most interested in.

REPLY: Indeed ground-based benzene and acetone data are missing during the periods when the PTRMS was used for vertical VOC profile measurements. However, the remaining benzene data of the ground-based measurements on 1 February 2006 are plotted in Fig. 4.

Language: P3985, L10: in Tyrol IN THE YEAR 2005. P3986, L15: in France IN THE YEAR 2003 P3988, L23: downward-LOOKING P3989, L13: cold fronts passage -> cold front passage P3989, L18: condition -> situation P3991, L14: reverse -> reversal P3993, 18: in 150 m -> at 150 m (similar at other places) P3993, L19: lost in strength -> weakened. Caption Fig. 9: this times -> these times, or the same times.

REPLY: The typos have been corrected in the revised version of the manuscript.

Reference:

Henne, S., et al. (2004): Quantification of topographic venting of boundary layer air to

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the free troposphere. Atmos. Chem. Phys. 4, 497-509.

REPLY: The reference has been included in the revised version of the manuscript.

Anonymous Referee #3

Last sentence of the abstract not clear. The amount of pollution export and air renewal can probably not be quantified in this study

REPLY: The amount of pollution export could not be quantified in this study. The last sentence has been rewritten in the revised version of the manuscript. "This vertical mixing due to thermally or dynamically driven slope winds reduces the concentration of air pollutants at the bottom of the valley and causes the formation of elevated pollution layers."

p3986, lines 7-12 : Henne et al. (2004) should be cited here as well.

REPLY: This paper has been added to the reference list of the revised version of the manuscript.

p3987, lines 9-10 : Important are not only the height of the mountains but also the crest heights (actually the lowest crest heights). These are especially important for the pollution export. In Figures 9, 10, these crest heights seem much lower than 2500 masl making it more likely that pollution export is possible. Deeper valleys with higher lowest crest heights (like some valleys in Switzerland) might not have any export in winter.

REPLY: Figures 9 and 10 only show a small section of the valley (+- 4 km from the center of the valley), a wider section is plotted in Figure 2 (+- 10 km). The lowest crest elevation within the 20x6 km box plotted in Figure 1 is about 1600 m asl (above sea level) and the mean crest level is close to 2000 m asl. The terrain is still increasing further northwest ending at 1900 m asl at the lowest mountain pass in the investigation area. Elevated concentration of air pollutants could be found up to 1900 m asl (compare Fig. 10) and therefore some pollution export might occur. However, about 20 km in the valley downward direction there is the Achenseepass, with 941 m asl the lowest pass in

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the region. Significant air pollution transport is expected to occur there. In the revised version of the manuscript the sentence has been extended to: "The mountains on either side of the valley reach over 2500 m m.s.l., with a mean crest height of 2000 m. m.s.l. and a lowest mountain pass of 1900 m m.s.l., within the investigation area."

p3989 Why were acetone and benzene chosen to be shown here. Some measurements and concentration levels could be compared to Gaeggeler et al. (2008) who also showed that wood burning is important for many VOCs in trafficated Alpine valleys during winter.

REPLY: Benzene was chosen, because it's the only VOC, for which a legislation threshold will be introduced in 2010. Acetone was chosen because it has many different sources and a long atmospheric lifetime and therefore somehow tracks the accumulation of air pollutants over longer periods with restricted dilution conditions. In the revised version of the manuscript the reference Gaeggeler et al. (2008) has been included and Section 3.1 has been extended by: "The mean values for benzene and acetone averaged over the whole measurements period were 1.25 ppbv and 3.1 ppbv, respectively. This exceeds values measured in Zürich in winter 2005/06 (Gaeggeler et al. 2008) by a factor of 1.8 and 2.5."

Chapter 3.2.1 : One could mention that the features of the diurnal cycle are similar to previous observations in summer in Alpine valleys (Prevot et al. (2000a)

REPLY: This is mentioned in the revised version of the manuscript. "The observed diurnal variation is similar to previous observations at this location and in other Alpine valleys throughout the year (Schnitzhofer et al. 2008, Prevot et al. 2000a)."

p3991, lines 27-28: I cannot see that the relative benzene contribution increases with altitude. The change with altitude might not only be due directly to difference in the lifetime of the VOCs but in general due to different features of the free tropospheric VOC composition compared to the composition at the valley floor (which surely is also connected to VOC lifetimes).

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REPLY: The relative share of benzene does not increase with height when compared with all VOCs plotted in Fig. 6, but compared to toluene it does. In the revised version of the manuscript the sentence was changed to: "The relative portion of acetone increased with height, due to its compared to the other compounds, longer atmospheric lifetime and therefore higher tropospheric background concentration."

chapter 3.2.2. The feature of the vertical VOC gradients might be compared to the gradients in Prevot et al. (2000a).

REPLY: In the revised version of the manuscript we added: "Compared to the morning values the VMR of some compounds have increased by a factor of two. This is similar to the diurnal variation of VOCs in higher air layers observed in the Swiss Alps during summertime (Prevot et al. 2000b), where increased VOC values could be found up to 4000 m m.s.l. during undisturbed clear weather conditions."

p3993, line 5: more relevant for air quality would be the PM1 volume and not the aerosol number which will be dominated by the two lowest channels of the OPC.

REPLY: PM1 values have been calculated in order to redraw Fig. 8 in the revised version of the manuscript. The respective sentence in section 3.2.3 has been changed to: "In Fig. 8 particle concentrations from 0.3-1 μm (i.e. the sum from the 5 smallest available GRIMM size bins; PM1) are plotted together with the potential temperature."

p3995, line 1: here the aerosol surface concentration rather than the number of particles would be more relevant to compare to the scattering. One might discuss somewhere that in the updrafts probably also the relative humidity is enhanced. An increase in humidity and growth of hygroscopic particles yields higher aerosol number (because smaller particles grow into the range of the OPC) but also higher aerosol surface and volume. Relative humidity data should be available from the aircraft data.

REPLY: We agree that the aerosol surface concentration would be more relevant to compare with the backscatter signal of the lidar measurements, but unfortunately size

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resolved particle measurements were only available in one minute time resolution and therefore not suitable for the comparison with the lidar data in Fig. 10. For a detailed comparison of lidar- and the in-situ particle measurements we want to refer to Harnisch et al. 2008.

p3995, line 13: as mentioned above I do not believe that there is no pollution export. I find it rather interesting that there is likely pollution export even in winter but this surely depends finally strongly on the depth of the valley. Henne et al. (2005) did not find any increase in water vapor above 2000 masl using climatological data of radio soundings north and south of the Swiss Alps. In valleys with lower minimum crest heights, pollution export might be possible. Increased humidity layers might be found downwind of the Alps at lower altitudes in winter between 800 and 2000 masl.

REPLY: We agree that pollution export in the investigation area is even possible in winter during situations with active slope winds and stronger winds above crest height. The export is expected to be small on 1 February 2006, since elevated pollution levels could be found up to 1900 m asl, which is also the height of the lowest pass within the investigation area, and wind aloft are week. However, significant pollution export might occur during days with even more active slope winds and stronger winds aloft (for example 24 January 2006, compare Gohm et al. 2009). Therefore we rewrote the respective parts in the revised version of the manuscript.

Abstract: "This vertical mixing due to thermally or dynamically driven slope winds reduces the concentration of air pollutants at the bottom of the valley and causes the formation of elevated pollution layers."

Section 3.2.5: "The export of air pollutants from the valley into the free troposphere could not be quantified with this experimental setup but is expected to be small on 1 February 2006, since the vertical transport did not significantly overshoot the main crest and a reverse of the slope wind circulation after sunset partly transported the polluted air back to the valley bottom (Harnisch et al. 2008). However, a significant

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export of air pollutants might occur during situations with even more active slope winds and stronger winds above crest height (compare 24 January 2006; Gohm et al. 2009), via a process described by (Henne et al. 2004)"

Conclusion: "Despite the stable stratification of the valley atmosphere, a vertical transport of air pollutants via slope winds reduces their concentration at the bottom of the valley, causes the formation of elevated pollution layers and even exports pollutants from the valley during specific situations in wintertime."

p3996, line14: it is not clear what the authors mean by very shallow slope wind layer. I would expect an even shallower slope wind layer during the night. During the day it should depend on the inversion strengths. The stronger the inversion strength in the middle of the valley, the shallower should the slope wind layer probably be. Such slope winds might be in general difficult to be detected by the aircraft. So I guess it is difficult to say how shallow the slope wind layer really is.

REPLY: We were not able to quantify the thickness of the slope wind layer because it could not be directly detected by aircraft wind measurements. The distribution of air pollutants indicates the vertical transport by slope winds. The thickness of the slope wind layer indeed correlates with the inversion strength, otherwise the transport of pollutants towards the middle of the valley at the height of the most stable stratification could not be explained. We believe the slope wind layer to be shallow due to the stable stratification throughout the entire valley atmosphere. However "very shallow" has been changed to "shallow" in the revised version of the manuscript.

References:

Henne et al. (2004) Quantification of topographic venting of boundary layer air to the free troposphere, Atmos. Chem. Phys., 4, 497-509. Henne et al. (2005) Climatology of mountain venting-induced elevated moisture layers in the lee of the Alps, J. Appl. Meteorol., 44, 620-633. Gaeggeler et al. (2009) Residential wood burning in an Alpine valley as a source for oxygenated volatile organic compounds, hydrocarbons and or-

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ganic acids, Atmos.Environ., 42, 8278-8287. Prevot et al. (2000a) Diurnal variations of volatile organic compounds and local circulation systems in an Alpine valley, Atmos. Environ., 34, 1413-1423. Prevot et al. (2000b) Influence of road traffic on volatile organic compound concentrations in and above a deep Alpine valley, Atmos. Environ., 34, 4719-4726

REPLY: The references have been included in the revised version of the manuscript.

Reference:

Harnisch, F., Gohm, A., Fix, A., Schnitzhofer, R., Hansel, A., and Neininger, B.: Spatial distribution of aerosols in the Inn Valley atmosphere during wintertime, Meteorol. Atmos. Phys., doi:10.1029/2006GL028325, 2008.

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