Long term measurements of optical properties and their hygroscopic enhancement" by M. Hervo et al.

Answers to Referee #2

The authors would like to thanks Referee #2 for his detailed review. The comments from the reviewer are underline and the answers from the authors are not.

GENERAL COMMENTS

1. The manuscript discusses long term measurements of optical properties and their hygroscopic enhancement. The title is thus well covering the content. The manuscript reads easily, is nice compact and contains few unnecessary details.
2. As said, the title mirrors the content of the paper. Therefore, I consider section 5 on aerosol radiative effect more a side issue that distracts the reader from the main content. Of course, the radiative effect is likely one of the motivations to this study but with the simple equations, the radiative effect reduces to not much more than the ratio of dry and wet extinction coefficients. Moreover the radiative effect is calculated for a hypothetical atmosphere with 90%RH always. In the abstract, the more informative result for ambient RH is mentioned, but as this value is not motivated and discussed in the main text and does not add much. I suggest to remove section 5 and references to it in the abstract.

Section 5 was moved to the supplementary material to focus on the study core.

3. The humidity size growth of particles was measured at 90% RH by an HTDMA. In section 4, these measurements are used to obtain parameterization for different dominant air masses and the full range of RH. This is both very informative and useful. Also the comparison of the f(90%) values for the different air masses is a fair comparison. The parameterizations can thus be used for discrete relative humidities when no humidity growth or scattering enhancement is measured. This is one of the key outcomes of the paper, to my opinion.
   For the period that the HTDMA was operational, the same kappa theory could be applied to extrapolate to ambient RH, so that the real relative humidity dependence of the optical properties, both extensive and intensive, could be calculated (assuming volume mixing rules for refractive indices etc). In the manuscript variations (year-to-year, season, diurnal) of dry optical properties are discussed in detail. The discussion including humidification enhancement is limited to f(90%). In line 6 of page 27744 it is mentioned that "marked seasonal variation with maximum enhancements during the warm season" is shown. This maximum is caused by the more frequent occurrence of oceanic air masses mentioned page 27743 line 7. However page 27734 line 21 it is mentioned that the station is frequently in clouds in winter. The summer aerosol is thus more hygroscopic whereas the winter aerosol encounters likely higher relative humidities. The net effect is not shown in the paper, but the radiative effect for ambient conditions is

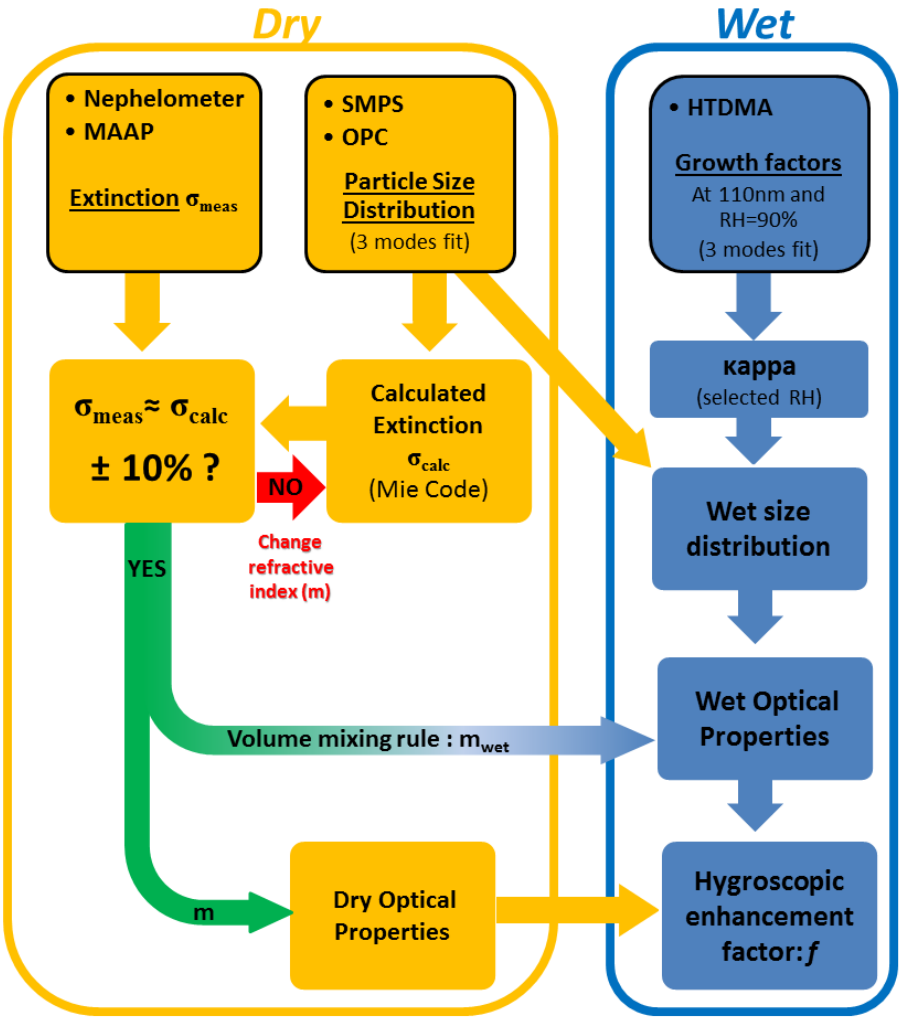
given in the abstract. I would like to see more of the enhancements, and variability thereof, for ambient conditions.
This question is now developed in the section 5 of the supplement.

4. In section 3.2 measurements at PdD are linked to spatial origin by using backtrajectories. The spatial information here is not alike the temporal information in section 3.1. The title of this section should not be spatial variation but something like "spatial origin". In the last para of page 27741 it is explained briefly how aerosol properties are attributed to spatial origin. I celebrated the compactness of this paper in the first lines of this review. However, here some important information is missing. A reader cannot reproduce figures 3, when PdD observations and the Hysplit model are available. At least the "weights" should be given and it should be described how figures 3 are made. I can only guess how all information is combined or averaged, for the intensive properties some weighing is included or not? Please explain a bit more.

The title of the section was replaced by spatial origin.

The following sentence was added to describe more precisely the weighting process:" *The weight is varying linearly from 1 for the first point of the back trajectory, to 0.5 for the last one, as already described in Asmi et al. 2012.*"

5. In section 2.2 modes for size (nucleation, accumulation, coarse) and growth (hydrophobic and (very) hydrophilic) are mentioned. I not sure how the various modes are later used. Moreover many steps are taken to finally come up with the parameterization. What measurement are used when and how are they treated could all be fit in a logical diagram. I suggest to include such a diagram for clarification. The diagram could contain e.g. HTDMA size selected 110 nm, growth at 90% RH, applied to size modes (SMPS,OPC), kappa theory, Mie theory, dry properties, volume mixing, etc. From such a diagram it should become clear what information is used from what instrument.
The following diagram was added to summarise the algorithm used.



6. Optical properties are measured in period 2006-2012 (abstract), absorption coefficient is even measured from 2001 onwards (section 3.1.2). However in none of the tables or figures the full period is shown. The longest seems to be shown in Fig 1. (2008- 2012). I can imagine that the availability of the MAAP is the reason for this selected period. However in the text it is not completely clear to me whether the full period or the selected period is used for the shown values. Please check and remove the seven year period from the abstract if not all data is used.

   Indeed the main focus of this study is the period 2008-2012 when the nephelometer and the MAAP were measuring simultaneously. Therefore, the abstract was corrected mentioning this period.

   Additionnally to this main period of study, long-term measurements were used to determine if pluri-annual trends were measured for variables that were measured on longer periods. The Mann Kendall test was applied to the measurement from 2006 to 2012 for the scattering coefficient and from 2001 to 2012 for the absorption coefficient. It is now mentioned in the text.

7. Throughout the document past tense is used consequently. Sometimes this is confusing when it does not become clear if it is a finding of this study or something general. "Balls are spherical" or "Balls were spherical". E.g. at page 27739 line 23-24. “the portion of

combustion aerosols, which were more absorbing, were expected. . ..”Where they more absorbing in this study or is the generally the case?

The document was proofread to avoid these grammatical mistakes.

SPECIFIC AND TECHNICAL COMMENTS

All specific and technical comments have been corrected according to the Referee suggestions.

Abstract is clear and to the point, accept for the last few lines. In the sentence starting in line 14, I prefer to see the (average or median) scattering enhancement for real ambient RH instead of the hypothetical 90%RH. In abstract aerosol radiative forcing for RH=90% is given (see also general comments). I do not understand why this is relevant information. It does not become clear to me in the main text. I suggest to remove this from the abstract. The enhancement for ambient RH is relevant, but pops up as a surprise in the abstract. I wonder where it comes from. Are measured surface relative humidities used in combination with the parameterizations or real PBL humidity profiles, or. If not discussed in maintext, please remove from abstract.

Page 27732 Line 9 “seasonality” do you mean seasonal (and diurnal) variation of the PBL?

P27736 line 1, SD is given is this the geometric standard deviation.

P27736 line 11, “Diffusion” should that be “scattering”

P27736 line 28 “this is not direct measurement” please rephrase.

P27737 line 2 “on average” is here meant generally or some mathematical average.

P27737 line 10 “respectively”refers to what?

P27737 line 22 “albedo . . ..” Typo?

P27738 line 5 SD 25.9 is this geometrical SD and what does it mean if it encompasses the nil value? I suggest to used percentile 25 and 75 like in fig. 1.

P27738 line 21 “At boundary layer sites” In Fig 1 results for PdD are shown but this is not a boundary layer site, is it?

P27738 line 27 “so that at altitude sites” is this a general conclusion for al altitude sites or just for this specific site?

P27739 line 7 “very similar” I would say they are in the same range

P27739 line 14 “Intrinsic” is this the correct word?

P27739 line 18 “other high altitude sites” this reference is used frequently. I suggest to list the sites that are referred to several times.

They are listed P27739 line 9: Jungfraujoch, Izana, Monte Cimone

P27739 line 25 “second, purely . . ..larger” In this study or in the Himalayas? This hypothesis can be tested by selecting cases with RH below 80%, right?

Indeed: If only cases with RH <80% are selected the 10th percentile is 15% higher. (0.81 instead of 0.77).

P27740 line 21 “This value”should be these values.

P27740 line 23 averages and medians are compared

P27741 line 2 typo diurnal

P27741 line 3 “), From” should be a period.

P27742 line 11 “could be” but it is not? Or do you suggest a paradox.

Indeed we suggest a paradox

P27742 line 19 I do not understand the arguments on ageing. The general picture I have is that when scattering aerosols condense on highly absorbing aerosols, the “shell” acts as a lens and absorption is enhanced. Here you suggest reduced absorption by scattering aerosol, right?

We suggest that scattering aerosols are enhanced in larger proportion compared to absorbing aerosols (presumably mostly combustion aerosols) in continental air masses compared to oceanic air masses. Chemical analysis of the aerosol of different origins indicated the same in earlier studies (Bourcier et al. 2012). This is now better explained in the text.

P27742 line 28 others remove “s”

P27743 line 22 “spatial scale” should be “spatial origin”

P27746 line 8 “The impact of hygroscopicity on the absorption coefficient was quantified from our measurements” This is rather “tricky” This is based on volume mixing of the refractive indices, but how do you treat the absorbing particles. The MIE theory you apply assumes spherical particles with a single refractive index and thus perfectly internally homogeneously mixed particles. This is assumption is not valid for most light absorbing particles. Please discuss a little.

As suggested by referee #2, the uncertainties introduced by the volume mixing rule on the retrieved refractive index is not discussed. We now moderate the wording, “quantified is now replaced by “estimated” and we now specify that our calculation assumes that no “coating” effect is taking place on absorbing particles.

P27747-P27748top extensive parameters show clear seasonal variation and variability according to text or is accidentally the same message included twice?