Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2020-291-RC2, 2020

Atmospheric Measurement Techniques Discussions

Interactive comment on "Combined use of Mie-Raman and fluorescence lidar observations for improving aerosol characterization: feasibility experiment" *by* Igor Veselovskii et al.

Anonymous Referee #2

Received and published: 14 September 2020

General: The paper contains new and very interesting observations obtained with a new approach of a fluorescence lidar for aerosol characterization. This lidar feasibility study is clearly worthwhile to be published in AMT. Nevertheless, the interpretation of the observations needs to be improved. A clear and more systematic separation of the different fluorescence contributions would be helpful to better follow the discussion. The argumentation is partly week and a bit speculative.

Minor revisions are requested.

The abstract has to be updated and adjusted .... after all the suggested improvements.

P2, L39: Burton et al. 2012... Only one reference here? What about own papers:

Veselovskii et al., 2015, 2020, what about Tesche 2011, Tellus, SAMUM 2, what about all the efforts within the ACTRIS EARLINET group on aerosol typing... during the last five years.

P2, L44-47: I have my doubts that aerosol particles can be clearly identified and quantified in cloud layers. Ok, you can detect them, but it is already well known that interstitial aerosol particles are always present in clouds. It is impossible to have aerosol particle free clouds.

P3, L76-84: These are confusing statement. I am puzzled by the wording ..'external' vs 'internal' mixing of aerosol particles within liquid water droplets. There is only ONE scenario: It is impossible to have droplets without a CCN, and it is also impossible to have clouds without interstitial aerosol particles (non-activated particles). So, there is only this ONE scenario: a mixture of interstitial aerosol particles (not acting as CCN) and droplets, each of the droplets nucleated on a CCN. The CCNs may be completely dissolved in the droplet, or survived as a solid particle within the droplet, as is the case for dust or soot CCN. The interstitial aerosol particles (in the cloud) may be much larger than the particles outside the cloud (because of strong water uptake at 100% rel. humidity), so the aerosol backscatter efficiency of particles within the cloud may be larger by a factor of 5 and even more, compared to the aerosol backscatter outside the cloud layer.

I recommend to avoid to introduce ... internal and external mixtures! There is only this ONE scenario: interstitial aerosol particles and cloud droplets. Now we need a clear differentiation: What is the contribution of dry particles to fluorescence? What is the contribution of fully deliquescent (dissolved, solution) aerosol particles? Sulfate particles are fully dissolved at high humidities? Can we be sure that the fluorescence signal in clouds is exclusively from interstitial aerosol particles? No contribution by cloud droplets? That needs to be carefully discussed.

Dust particles with liquid shell .... produce an enhanced fluorescence signal (lens

effect)! Is that checked? Is there a reference for that?

P5, L157: Please keep in mind that RH increases from dry conditions (e.g., RH of 40%) to moist conditions (e.g., 80%, 90%, 95%) already a few hundred meters below cloud base, and then to 100% above cloud base. The aerosol particles grow by water uptake, change their backscatter efficiency and the fluorescence capability, some get liquid, some remain dry. Then in the cloud, cloud droplets come into play, backscatter efficiency of interstitial aerosol particles (at 100% humidity) may be much larger than for the aerosol particles below the cloud, or before the cloud formed. All this needs to be considered. . . in the cloud observations of backscatter and fluorescence signals.

Is all this known well enough to quantify the aerosol-related (non-droplet) fluorescence signal in clouds? To my opinion this is not the case. We do not know much about this. So, I have my doubts about Eq.(7).

P6, L176, the particle fluorescence capacity is introduced. I would recommend to do that in form of an equation. Now, the fluorescence signal will change with increasing humidity and water uptake and this in a different way as the total backscatter signal. Again, I think the knowledge about water uptake and the link to fluorescence signal changes is just qualitative. . .. So there is no clear knowledge about the increase of the capacity G with water uptake. . ..

P6, L176, beta_L ˆa is introduced in Eq. (1) but not beta_L or betaˆa.

P6, L176: The capacity G_F expresses what? The fluorescence signal changes probably when aerosol particle get a wet coating, the backscatter signal changes by water uptake, so there is no clear reference (denominator), and thus, what does G_F indicate?

P6, L185 and L187: Again, because of the not well-known impact of water uptake, I do not belive that you can quantify N and V at conditions with rising humidity just below cloud base or even within the cloud? . . . so that you can not estimate fluorescence

cross sections accurately enough. If you want to present it please clearly state that there are many questions how trustworthy this estimation is.

3. Observations. . ..

A general comment: Trajectory analysis would be helpful for all cases discussed. There is no need to show them all, but it would improve the discussion .... to know more about the origin of air masses, and the kind of aerosol mixtures. . .

And it would also be helpful to have something like a bullet point list or an overview table, what aerosol produces fluorescence, what does not cause fluorescence, the same for droplets or water in aerosol particles. . .., what is producing fluorescence, and what does not. . .. And please provide references that support these statements

I ask for such a table because I learned more and more about fluorescence in detail . . .. from page to page of the manuscript, without having a complete picture right in the beginning of the discussion. Such an overview would facilitate all discussions and complex interpretation of the shown observations.

P7, L205-206: Do you mean external mixing of dust and biological /organic particles, or do you mean internal mixing, this would mean coating or partly coating of dust particles with organic material.

P7, L215: Pure water is not fluorescing, but what about the aqueous solution of dissolved aerosol particles (before becoming a droplet when acting as CCN). Again, what about the change in fluorescence efficiency with increasing water uptake and finally even change of phase (from dry and solid to totally liquid-acid aerosol particle)?

P8, in general and to mention again: would be nice to have HYSPLIT backward trajectories. . . to learn more about aerosol mixtures observed and what kind of aerosol are candidates for causing significant fluorescence.

P9, L266-280: Again the discussion part with N and V, I am not convinced that this is a fruitful part. Yes, there are numbers, but can we trust them?

3.2. Fluorescence of aerosol particles within clouds

This section is very interesting but, at the same time, a bit confusing. A more systematic way of presentation would be useful: What causes fluorescence, what not, what is the impact of water uptake, what happens with fluorescene when droplets are formed, with 'liquid' CCN and with solid CCN, etc. ... So, a bullet point list or a Table... would be nice.

P10, L290-299: Again, this separation of externally and internally mixed cloud. ... As mentioned above, there is only ONE scenario:

In all clouds, there is just a mixture of interstitial aerosol particles (not acting as CCN) and cloud droplets nucleated on the available CCNs.

Furthermore: In the cloud we have 100% relative humidity, so the interstitial aerosol particles are not dry, and most of them are just solution droplets (before activation ... to become cloud droplets). And the water droplets, on the other hand side, are formed on part of the solution droplets (CCN) but now consist almost entirely of water.

So please rephrase, and avoid external and internal mixing...

P10, L311: What do you mean here. ...? Fluorescent aerosol particles are inside the water particles. Ok, but must they be solid? If they are dissolved in the aqueous solutions, will there still be a fluorescene signal? May be droplets and CCN in the droplet do not produce any fluorescence signal, and fluorescence is only caused by the interstitial aerosol particles, and the increase in the fluorescence signal arises from water uptake effects?

So, what shows Fig4? ...in contrast to Fig5? If there is a difference, what is the reason?

P11, L322: Please, do not switch from one wavelength to another. That makes comparisons confusing. If beta1064 is 0.07 Mm-1 sr-1, then the 532 nm backscatter coefficients below the cloud is probably about 0.15 to 0.2 Mm-1 sr-1. That should be men-

tioned. And then we have an increase by a factor of roughly 3000 when you measure cloud a cloud beta532 value of 500 Mm-1 sr-1, and the fluorescence signal increases just by a factor of 5. ... that is a good proof that water does not produce a fluorescence contribution. Please state that, if my comment is true, and if there is definitely no cross talk...

P11, L327-328: Again almost the same increase in beta 532 when we start from about 0.04 Mm-1 sr-1 for 532nm (estimated from 1064 nm information) and end up at 130 Mm-1 sr-1. All this should be given in more clearly way ... by using ONE wavelength.

But can we trust an increase by factor 5 of the aerosol-related fluorescence backscatter when the elastic backscatter increase by a factor of 3000? Can we be sure that there is no cross talk, not stray light somewhere, nothing? What causes the increase. ...? The lens effect? Is there another explanation?

P11, L342 For insoluble particles. ... increase of fluorescence ... by lens effects. ... Yes that can be, but it remains speculations, most of urban aerosol is sulfate aerosol (and not BC-containing haze) and sulfate particles dissolve completely ... and then there is no lens effect...

P12, L358-362: This is again a non-acceptable speculation. The two cloud layers may have formed in two different air masses with different aerosol types, and the different aerosol types caused different levels of fluorescence.

Figure 2, would be nice to have backward trajectories... and thus origin of air masses for the two cases.

Figure 3, again: what is the origin of the aerosol (according to HYSPLIT trajectories)?

Figure 4, strong increase of cloud beta532 but not of fluorescence beta, what can we conclude? Strong increase of beta532 by droplet backscatter, and at least significant increase of the fluorescence signal because of water uptake of interstitial particles ...

And again, HYSPLIT trajectories would be nice to all the cases discussed. The trajec-

tories must not be shown in detail, but information about origin and mixture of aerosol particles would be helpful.

Final remark:

This is a good paper and needs only some minor clarifying information and a clear definition of the cloud environment (with interstitial non-CCN aerosol particles and CCN-based cloud droplets).

Afterwards (in the comparison. . .before vs within the cloud) a clear definition and systematic separation of the contributions by dry aerosol particles, wet aerosol particles, dissolved aerosol particles, cloud droplets nucleated on sulfate aerosol, and cloud droplets formed on soot and dust particles to fluorescence and to backscatter would be good and would make the discussion easier.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2020-291, 2020.