Authors' Response to Interactive Comment by Referee #1 (Atmos. Meas. Tech. Discuss., 3, C2061-C2063, 2010) on Reinap, Wiman, Gunnarsson and Svenningsson: "Dry deposition of NaCl aerosols: theory and method for a modified leaf-washing technique" (Atmos- Meas. Tech. Discuss., 3, 3851-3876, 2010)

We appreciate the efforts made by Referee #1 in evaluating our contribution. In particular, we value the Referee's assessment of our experiments as having "been performed robustly" and that our "description of experiments and calculations are sufficiently complete and precise to allow their reproduction". Since the Referee's comments are organised as responses to an AMTD Editorial Office questionnaire, and not as a set of questions and recommendations addressed to the Authors, we found it necessary to design our response in a manner that deviates somewhat from otherwise common structures. Nevertheless, out responses cover all issues raised by the Referee.

It appears that the Referee has overlooked the fact that *our study addresses the usability of chloride and sodium as tracers in experimental work* wherein leaves (oak leaves, in our case) have been exposed one way or another to an aerosol. In our case, the exposure procedure happens to involve a wind tunnel and an aerosol generator of bubble-bursting type, but our tracer-usability investigations could have used other designs (such as direct spraying of the leaves with NaCl aerosols with appropriate particle-size distributions). Hence, *the focus of our contribution is not on throughfall research* but on assessing whether some simple and cost-efficient compound and method – rather than those based on for instance radioactive tracers – can be used – *in laboratory experiments, not in the field* – to represent an aerosol with certain desirable characteristics; whether that compound can then be washed-off from oak leaves exposed to that aerosol; and what the wash-off conditions should then be.

We were thus searching for easy-to-handle and inexpensive tracers and, having tested a number of options (among them, "fingerprinting" our aerosol with various substances, including yttrium), focused on the potentials of chloride, with NaCl as a possible candidate. Like other substances, chloride and sodium pose problems *in field-based throughfall studies*, inasmuch throughfall represents a highly complex chain processes and mechanisms. As is well known since long, these involve, *inter alia*, precipitation chemistry, intrinsic leaf chemistry, and leaf-surface interactions with gaseous and aerosol deposition as well as contamination resulting from for instance insects, pollen, fungi, and vegetative fragments. While, naturally, we appreciate (and have consulted) the Referee's suggestions as to treatises on some of *those* aspects, we are well aware that the realm has been addressed by a huge body of literature over the years, with works by Parker being particularly often referred to. We found it appropriate in our paper to invoke the throughfall-research realm to *some* extent, because "the principles that underpin the wash-off technique have some relationships with those applied in forest biogeochemistry" (p. 3853); lines 10 – 11). At the same time, we find it quite sufficient with a mere few references (including to research – of interest to the choice of tracers – from 1948) of particular importance to interpreting results from our wash-off experiments. As is clearly expounded on in our paper, our wash-off technique is *not intended as a contribution to the throughfall-research area.* Instead, as we observe on p. 3853, our approach is that "many of these difficulties [that pertain to throughfall studies] can be avoided or minimized in controlled wash-off techniques applied on leaf material exposed to an aerosol in a wind-tunnel environment" (p. 3853; lines 16 –18).

However, it seems that we have not been successful in conveying that focus of our contribution because the Referee, after stating that "the problem is in their [the experiments'] utility", chooses a route that we find interesting in itself – *but* of quite limited relevance to evaluating *this* paper. Instead of concentrating on whether our experiments help provide a method with which a simple substance – NaCl – can be used to track an aerosol before and after its being captured by leaves, and on the requirements that we found necessary if so used (pre-exposure wash-off step, followed by three post-exposure wash-off steps at precisely given time intervals), the Referee discusses problems with throughfall studies and wind-tunnel-based studies of deposition.

As is obvious from the above, we certainly agree with the Referee's standpoint that throughfall studies (albeit not in our focus in this contribution, then) are compounded by many problems. In regard to wind-tunnel based investigations of aerosol-deposition processes, *this* contribution of ours does not present, *and does not intend to present*, either details of the wind-tunnel system that we used for exposing leaves to the NaCl aerosol or constraints and opportunities for interpreting data in terms of for instance deposition velocities. What our paper *does* state is that approaches to studying aerosol deposition to vegetation systems include a vast array of methods, all of which hold various strengths but also suffer from a variety of problems and development needs. (For instance, the many attempts with using throughfall methods in the field for quantifying particle deposition face well-known problems as addressed by the Referee *and* by ourselves; uses of various types of surrogate particle-collecting surfaces for emulating leaves may fail to simulate potentially highly important characteristics of living leaves; flux measurements such as based on eddy correlation may not adequately capture effects of relative-humidity on particle size and may therefore misrepresent particle-concentration gradients and fluxes in forests; mathematical-physical models based on aerosol-mechanics alone may not work well even for seemingly simple collector geometries; wind-tunnels need to be very large to enable simulation of wind- and turbulence patterns occurring under real field conditions; theories for turbulence itself are in several respects still unsatisfactory; the solving of realistic versions of the Navier-Stokes equations requires very large amounts of computer capacity.) Again, the Referee thus raises issues that are interesting in themselves but that go far beyond the focus of *this* paper. This paper does not intend to broaden issues to what general scientific stance to take to inevitable imperfection in scientific experimentation.

With respect to Referee's comments that remain to be addressed in this Authors' response, we have difficulties with their internal consistency. The Referee states:

"the concepts have all been well-rehearsed before", and,

"the development of the equations (which are well known) is rather complicated, and the assumption of first-order kinetics is neither explained nor justified".

Either, first-order kinetics and the associated equations belong to concepts that "have all been well-rehearsed before" and, if so, would not need to be explained or justified – or, if explanation and justification are needed, the Referee's comments imply that the concepts in question have not been well-rehearsed before.

Our approach is intended to emphasize the dynamics as $t \rightarrow \infty$ (and not for the first few minutes), because we wanted to find an appropriate number of wash-off steps and appropriate time-durations between each step, *for our particular system* (chloride, sodium, oak leaves,

and MQ-water). Such information is fundamental to assessing uncertainty in wash-off techniques. For our system, we found that our wash-off data are well explained by first-order reaction dynamics. Since this is a simple explanation (and thus, in line with the Occam razor principle, to be favoured over more complex alternatives) we wanted to be particularly conscientious with respect to explaining conditions that would, ideally, have to be met for building our data interpretation on first-order reactions. This led us to presenting physically and mathematically very precise definitions of the differential equations involved and the details of the sub-functions that they include. Because (as explicitly discussed in our paper and based on pertinent references) sodium might pose particular problems if used *as a tracer in experimental work* we wanted to obtain estimates of the magnitude of this potential problem with *our wash-off technique*. (Again, our aim was *not* to address the magnitude of the problem in field-throughfall studies.)

Our paper gives answers to the questions regarding appropriate number of wash-off steps and appropriate time-durations between each step, for this particular system. It appears that the Referee finds this experimental approach "robust". But, it seems that in remarking that "no new information was obtained" the Referee has access to such data since long. However, the Referee does not provide a precise reference – to laboratory-based wash-off techniques that ensure the usability of chloride, sodium, or any other simple-to-handle and inexpensive substance useful as a tracer – to enable us to agree with the statement that "no new information was obtained". Nor do we find ourselves able to agree with the Referee's statement that our system "does not progress our understanding of the capture of particles by vegetation". This disagreement is because *this* paper does not set out to detail the entire system in use here, and does not purport to introduce and discuss opportunities and constraints pertaining to either wind-tunnel based studies of aerosol deposition or to other methods.

In conclusion, we find the Referee's observations interesting but in major respects of limited relevance to our results in this contribution. In part, the misunderstandings that seem to be at hand might emanate from an insufficiently clear focus in our contribution. For instance, the title would seem to better be "Theory and method for a laboratory-based leaf-washing technique for oak leaves exposed to artificially generated NaCl aerosols".

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