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Comment on comment by Markus Rapp and Franz-Josef Lübken on "Ice iron/sodium film as cause for high noctilucent cloud radar reflectivity"

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[1] Rapp and Lübken (RL09) state that there is excellent, precise agreement between observations and polar mesosphere summer echo (PMSE) models based on radar reflection by gas-phase electrons and conclude that because of this excellent, precise agreement other models should not be considered. In particular, they provide figures demonstrating quantitative agreement between observations and a heating/overshoot model. Careful consideration of the models presented by RL09 shows that there are inconsistencies in the assumptions regarding Lambda (i.e., the ratio of the number density of electrons attached to aerosol particles to the number density of free electrons) and there are quantitative errors in the calculation of the radar reflection. In addition, RL09's claim that the correlation between PMSE maximum and bite-outs is either spurious or misrepresentative is refuted using radar measurements. Finally, it is shown that there is a shortcoming (not discussed by RL09) in the Bellan 2008 model, and a modification addressing this shortcoming is briefly described.

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1. Introduction

[2] In their comment on work by *Bellan* [2008] (B08), *Rapp and Lübken* [2009] (RL09) state that there is a wealth of independent observations that the scattering of radar waves comes from gas-phase electrons and conclude that alternate theories need not be considered. In particular they state "The excellent agreement between theory and observations reinforces the crucial role of free electrons for PMSE and *excludes* the metal electron hypothesis. In summary, the PMSE heating experiment *can only be explained* by free electrons and not by metallic electrons" (italics added). However, agreement between a particular theory and observations does not "prove" a theory is correct since in principle alternate theories could exist that are also consistent with observations. All one can do is disprove a theory by showing that it is inconsistent with observations.

[3] Contrary to RL09's claim that the electron gas-phase scattering model is compelling, the literature contains many instances where agreement between existing PMSE theory and observations is not good. For example, *Blix et al.* [2003] stated "However, it is still an open question which role the aerosols play in the creation process of PMSE."

[4] We show in this comment that the gas-phase electron scattering model is not as compelling as RL09 suggest by listing several questionable aspects. We also make some comments about the metal scattering model including a recently discovered shortcoming and how this shortcoming might be addressed.

2. Issues Regarding Fraction of Electrons Residing on Aerosol

[5] The extent to which electrons are free or attached to aerosols is quantified by the parameter [*Cho et al.*, 1992; *Rapp et al.*, 2002]

$$\Lambda = \frac{Z_a \overline{N}_a}{\overline{N}_e}, \tag{1}$$

where Z_a is the number of electrons attached to an aerosol particle, $\overline{N}_a$ is the spatially averaged aerosol density, and $\overline{N}_e$ is the spatially averaged gas-phase electron density. $\Lambda = 0$ thus corresponds to no electrons being attached to the aerosol particles, $\Lambda = 1$ corresponds to half the electrons being free and half attached, and $\Lambda = \infty$ would be the limit of all electrons attached and none free (i.e., bite-out). *Cho et al.* [1992] postulated that strong PMSE only occurs when $\Lambda >$ 1.2. Until 2003 it was generally believed that this criterion held true (e.g., see the paper by *Rapp and Lübken* [2000], one of the papers cited by RL09 as being a theoretical model in excellent agreement with observations). However, by

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making direct measurements of Λ in the presence of PMSE, *Blix et al.* [2003] found strong PMSE when $\Lambda \ll 1$ and stated "We find that PMSE occur for rather small amounts of charged aerosols. This is in contradiction with previous, mainly theoretical studies predicting that PMSE only occur when the ratio between aerosol charge number density and the number density of electrons is larger than about 1." Thus, there is poor agreement between pre-2003 theory and observations regarding the relation between Λ and PMSE strength. In 2003, Rapp and colleagues presented two postulates for why the predictions of *Cho et al.* [1992] regarding PMSE dependence on Λ were not in accord with observations.

[6] In the first of these 2003 papers, *Rapp et al.* [2003] proposed the empirical ad hoc proxy quantity $Z_a \overline{N}_a r_d^2$ as being crucial for the existence of PMSEs (here r_d is the aerosol radius). They found heuristic agreement between this ad hoc proxy and PMSE for certain examples, and concluded that "The agreement between this proxy and the main characteristics of PMSEs implies that simple microphysical models do not satisfactorily explain the PMSE physics and need to be improved." We note that the proxy model is not consistent with the presumption that the gas-phase electron density is spatially modulated by the aerosol density. This inconsistency is because ambipolar electron diffusion, the means for the required spatial modulation of gas-phase electrons, is predicted by *Rapp et al.* [2003] to depend on r_d. However, for a given aerosol charge, r_d affects only the aerosol diffusion and hence only the lifetime of PMSE, not the force exerted on electrons. If the aerosols do not diffuse as is presumed to be the case during PMSE, then electrons are physically influenced only by the charge residing on aerosol particles, not by the physical size of the aerosol particles. Specifically, two different sets of aerosol particles having the same Z_a and the same spatial Bragg-scale density distribution but different values of r_d would produce identical modulations of the electron density and so produce identical PMSE.

[7] In Appendix B of the second of the 2003 papers, *Rapp and Lübken* [2003] argued that *Cho et al.*'s [1992] $\Lambda \sim 1$ criterion resulted from *Cho et al.* [1992] making a physically unrealistic choice for the initial condition of electron charge density spatial inhomogeneity. Specifically *Rapp and Lübken* [2003] noted that *Cho et al.* [1992] assumed that the electrons in a quasi-neutral configuration were initially positively correlated with the aerosol distribution whereas in fact the electrons should be anticorrelated with the aerosol distribution because the negatively charged aerosol particles repel electrons. However, P. M. Bellan (Meta-equilibrium state of multi-species ambipolar diffusion and its relevance to Polar Summer Mesospheric Echoes, submitted to *Journal of Atmospheric and Solar-Terrestrial Physics*, 2010) recently showed that if the electrons have an initially positively correlated, quasi-neutral density perturbation n_0 such as assumed by *Cho et al.* [1992] and *Rapp and Lübken* [2000], the electrons and the ions will quickly relax to a different quasi-neutral configuration where the electrons have the anticorrelated density $n_e = -\Lambda n_0/(\Lambda + 2)$. This relaxation takes place in less than 5 milliseconds, which is the time scale for Bragg-relevant ambipolar diffusion of electrons and ions with no aerosol motion; Bragg-relevant refers to the Fourier component of the electron spatial inhomogeneity at the Bragg wavelength. This rapid relaxation results in $|n_e/n_0|$ being order unity when Λ is order unity or larger but in $|n_e/n_0|$ being small and proportional to Λ when Λ is small. Thus, the rapid relaxation of a positively correlated initial electron perturbation produces a much larger relaxed state for Λ order unity or larger than for Λ small compared to unity. This dependence on Λ indeed provides a spurious prediction that PMSE should be large if Λ is large compared to unity, because the assumed initial positively correlated electron density perturbation is non-physical. On this point, we are in agreement with *Rapp and Lübken* [2003]. The relaxed anticorrelated electron perturbation then proceeds to decay on the Bragg-relevant aerosol diffusion time scale $(k_B^2 D_a)^{-1}$ where $k_B = 4\pi/\lambda$ is the Bragg wave number and λ is the radar wavelength. However, this issue of rapid relaxation from a spurious initial condition is different from the issues of response to HF heating and to the existence of PMSE in the presence of bite-outs, since these latter issues involve time scales much longer than 5 milliseconds. Also, as shown in section 4, the choice of a Gaussian bump is inappropriate for characterizing Bragg scattering and the details of how such a bump evolves does not relate to what is critical to the Bragg scattering process.

[8] The claimed precise, excellent theoretical prediction RL09 provide in their Figure 1 is extracted from Figure 1 of *Havnes et al.* [2004]. Examination of the original Figure 1, from *Havnes et al.*, 2004], shows that the solid curve (10 nm) in RL09's Figure 1 is for the case where $\overline{N}_a = 2 \times 10^7$ m^{-3} and the dashed curve in RL09's Figure 1 is for the case where $\overline{N}_a = 4 \times 10^6$ m^{-3} while for both situations $\overline{N}_e = 2 \times 10^9$ m^{-3}. Thus, the dashed curve has $\overline{N}_a/\overline{N}_e = 10^{-2}$ and the solid curve has $\overline{N}_a/\overline{N}_e = 2 \times 10^{-3}$. Since a 10 nm aerosol is presumed to be charged to $Z_a \simeq 1$ and a 50 nm aerosol particle is presumed charged to $Z_a \simeq 5$ both the solid and dashed lines in RL09's Figure 1 correspond to situations where $\Lambda = 0.01 \ll 1$, i.e., to situations where *Rapp et al.* [2002] have argued that PMSE should be negligible. Thus RL09 are mixing up large and small Λ situations since they refer without distinction to *Havnes et al.* [2004] (where $\Lambda = 0.01$) and to *Rapp and Lübken* [2000] (where $\Lambda = 2$).

[9] RL09 state "A direct proof of the importance of electrons in the gas phase comes from the active modulation of PMSE using HF heating. *Chilson et al.* [2000] were the first to use a powerful HF heating radar in order to enhance the electron temperature to ~3000 K at altitudes where PMSE was simultaneously observed by the EISCAT VHF radar." However, careful reading of the work by *Chilson et al.* [2000] shows that *Chilson et al.* [2000] never claim that electrons were heated to 3000 K. In fact no direct, independent measurement of electron temperature has ever been reported, so RL09 are simply asserting without proof that electrons are heated to 3000 K. A proof would require a direct, independent measurement of electron temperature verifying that this temperature indeed rises from 150 K to 3000 K in the relevant region.

[10] According to the gas-phase PMSE model, a fraction α of existing electrons deposit on aerosol particles so that the remaining gas-phase electrons have a density propor-

tional to $1 - \alpha$; thus $\Lambda = \alpha/(1 - \alpha)$ and $\alpha = \Lambda/(1 + \Lambda)$. Furthermore, spatial inhomogeneities of the aerosol particles are supposed to spatially modulate the electron density. The PMSE is supposed to result from radar waves Bragg reflecting from the resulting electron spatial inhomogeneity. Thus, the strength of the electron inhomogeneity is approximately proportional to the product of the strength of the modulating substance (i.e., the charge on the aerosol which is proportional to α) and the number of electrons that are being modulated (i.e., to $1 - \alpha$). Hence, if PMSE is due to spatial inhomogeneities of charged aerosols spatially modulating the gas-phase electrons, PMSE should scale approximately as some power of $\alpha \times (1 - \alpha) = \Lambda/(1 + \Lambda)^2$ and so should vanish both when $\alpha = 0$ (no electrons reside on the aerosol particles) and when $\alpha = 1$ (no free electrons available to be modulated, i.e., bite-out). The maximum of the function $\alpha - \alpha^2$ occurs when $\alpha = 1/2$, which corresponds to $\Lambda = 1$. This is not to say that PMSE could not occur at small Λ and small α, but at these values, PMSE would not be maximized for the given total electron density. Assuming $\Lambda = 2$ (i.e., α=2/3), as done by RL09 when they cite *Rapp and Lübken* [2000] as part of their argument, corresponds to a situation where increasing Λ should cause a reduction in $\Lambda/(1 + \Lambda)^2$ and so should cause a reduction in PMSE (undershoot), not an overshoot. The possibility of undershoot due to high α is discussed by Bellan (submitted manuscript, 2010) and also has been discussed by *Biebricher et al.* [2006], who presented an observation of undershoot but no measured values of Λ. The inconsistent correlation between PMSE strength and measured Λ values from *Blix et al.* [2003] casts doubt on PMSE scaling with $\Lambda/(1 + \Lambda)^2$. The claim that PMSE can only be explained by gas-phase electrons is thus subject to question and it is reasonable to consider that other mechanisms could be important when Λ is large so that the density of gas-phase electrons is very small.

3. Bite-Outs, HF Heating, and PMSE

[11] RL09 do not deny that bite-out at maximum PMSE causes a dilemma nor do they question the existence of bite-outs in general. In fact, the last sentence of the paper by *Rapp and Lübken* [2003] states that electron bite-outs are regularly found at PMSE altitudes. Bite-outs (i.e., $\Lambda \to \infty$) are predicted by models of HF heating, have been measured in rocket flights, and also have been observed using incoherent radar backscatter measurements. If the rocket observation of bite-out is representative of the electron density over the volume sampled by the radar and if PMSE is caused by spatial modulation of gas-phase electrons by negatively charged aerosol particles so PMSE scales approximately as the square of $\Lambda/(1 + \Lambda)^2$, then PMSE should be strongly attenuated at a bite-out and not, as observed, be at its maximum value. Simply put, the radar cannot reflect from the gas-phase electrons if the gas-phase electron density is nearly zero. RL09's solution to this dilemma is to argue that the regular observation of bite-outs at PMSE altitudes should be dismissed as being an instrumental problem.

[12] RL09 do not explain why the rocket density bite-out measurement should be discarded whereas other rocket measurements should be retained. *Cho et al.* [1992] were concerned about this issue also and noted that two different types of rocket measurements were in agreement with each other, indicating that the rocket measurements are not spurious. In effect, RL09 want to discard what has been measured (radar reflection observed when bite-outs are observed to be present) while invoking what has not been measured (twentyfold increase in T_e due to HF).

[13] The other solution to the bite-out dilemma RL09 propose is that the ~kilometer high bite-out sampled by the rocket is due to horizontal spatial inhomogeneities. RL09 characterize this spatial inhomogeneity as being "minor." However, the difference is on the order of a fiftyfold reduction in electron density that is horizontally localized but has approximately kilometer vertical uniformity, i.e., a vertical tunnel and not a minor inhomogeneity. Such a vertical tunnel in the turbulence is at odds with the fundamental assumption underlying coherent scattering, namely that the radar is reflecting from Bragg-wavelength fluctuations in homogeneous turbulence. Since there are sheared horizontal winds with velocities ~10 m s^{-1}, a fiftyfold horizontally localized density depletion would be sheared by a distance of 10 km in about an hour, and so it is unrealistic to expect such large horizontal density inhomogeneities to persist.

[14] The rocket bite-out measurements are supported by radar measurements. *Rottger et al.* [1990] presented incoherent scattering radar measurements of bite-outs and found an association between bite-outs and large PMSE. *Rottger et al.* [1990] state in their abstract "A narrow electron density depletion seen in the EISCAT data appears to occur at the same altitude as the CUPRI PMSE and is consistent with earlier rocket measurements" and in their text "We notice that the maximum of the CUPRI PMSE occurs just at the altitude of the electron density depletion observed with the EISCAT UHF radar." These radar measurements are not subject to RL09's arguments that bite-out is horizontally localized or that rocket measurements are spurious. Thus the correlation between bite-outs and maximum PMSE cannot be discarded. The dilemma remains and provides motivation for models where PMSE is not the consequence of gas-phase electrons.

4. Inappropriate Modeling of Bragg Scattering by *Rapp and Lübken* [2000]

[15] *Rapp and Lübken* [2000] assume, as is generally accepted, that PMSE is a result of Bragg backscattering. As representative of the fundamental structure relevant to Bragg scattering, they use an electron profile that is initially a single Gaussian bump with width corresponding to half the radar wavelength. This use of a Gaussian bump fails to take into account the fundamental nature of Bragg scattering. Bragg scattering results from the nonlinear interaction between a radar wave $\sim\cos(kx - \omega t)$ and a *periodic* density inhomogeneity having spatial dependence $\sim\cos(2kx)$. The product $\cos(kx - \omega t)\cos(2kx)$ resulting from modulation of the radar signal by the density inhomogeneity contains a component $\sim\cos(kx + \omega t)$, i.e., a backscattered wave. It is thus only the Fourier component of electron spatial inhomogeneity at $k_B = 2k$ that contributes to the backscattering.

All other spatial Fourier components do not contribute to Bragg scattering. Thus examination of the evolution of the spatial profile of an initial Gaussian bump is not relevant to the Bragg scattering that causes PMSE because only the Bragg component of the electron profile matters. To see this, consider two temporally evolving identical Gaussian bumps separated by a distance $\pi/2k$ (i.e., separated by a quarter wavelength of the radar wave). This pair of evolving Gaussian bumps will produce no PMSE no matter how steep the density gradients of the individual bumps become, because the combination of the two evolving bumps has no spatial inhomogeneity at the Bragg wavelength. Equivalently, one could say that the reflection produced by the bump farther from the radar transmitter is 180° out of phase from the reflection produced by the bump closer to the radar transmitter. The 180° phase difference results because for the bump farther from the radar transmitter the incident radar signal travels $\lambda/4$ farther up and then its reflection must travel farther $\lambda/4$ down.

5. Incorrect Power Scaling by *Havnes et al.* [2004]

[16] RL09 claim that their Figure 1 demonstrates excellent quantitative agreement between observations and theory. The claimed excellent quantitative agreement cannot in fact be true because of an error in the theoretical calculation. According to *Havnes et al.* [2004] the PMSE scattering power is proportional to the gradient of the electron density in one periodic structure. Specifically, in their equation (12), *Havnes et al.* [2004] presume that scattered power scales as $R \sim n_e(E) - n_e(C) \sim \tilde{n}_e$ where E and C refer to the edge and center of a periodic structure. This assumed linear proportionality of Bragg scattering power on $\tilde{n}_e$ is inconsistent with Bragg scattering and so the predicted scattering power in RL09 Figure 1 is quantitatively incorrect even for the $\Lambda \ll 1$ regime. Bragg scattered power scales as $\tilde{n}_e^2$ as discussed by *Landau and Lifshitz* [1960, p. 401, equation (97.13)] and also as shown in the derivation of scattering of B08. As justification for their equation (12), *Havnes et al.* [2004] cited *Ginzburg* [1970, pp. 236–237, equation (16.31a)]. The scattering coefficient R defined by *Ginzburg* [1970, pp. 236–237, equation (16.31a)] was for electric field *amplitude* $\tilde{E}$, and not for wave power, the scattering coefficient of which scales as $|\tilde{E}|^2$.

6. Metallic Coating

[17] RL09 argue that metallic coating is improbable. However, the proposal that aerosols can become metal-coated is not new and was in fact proposed by *Cho et al.* [1992], one of the papers on which RL09's analysis is based. RL09 argue that the aerosol particles survive for only a few minutes and so could not be around long enough to become metal-coated. However, *von Zahn and Berger* [2003] have shown that after nucleation, ice aerosol particles can live many days and can migrate large distances during this time. Also, *Rapp and Lübken* [2003] assumed that 50 nm aerosol particles would not significantly diffuse for several hours; such a consideration implies that the aerosol particles last more than a few minutes. The details of how ice aerosol particles become metal-coated are not well known at this time, and without such knowledge it is premature to assert that metal-coating is impossible on a time scale of a few hours.

7. Issue Regarding the Geometry of Metal-Coated Dust Grains

[18] We now mention a new and important issue that has come to light which is separate from those raised by RL09. B08 argued that because the ice grains are smaller than the metallic skin depth, the radar electric field should penetrate the ice grains and drive oscillating electric currents that would radiate the observed PMSE signal. Reconsideration of this argument shows that while it is true that electric fields can penetrate metal having dimensions smaller than the skin effect without attenuation, this turns out to be an insufficient condition for the radar electric field to drive the required oscillating currents needed to produce scattering. The problem is that a spherical metal-coated ice grain will become polarized by the radar wave electric field [*Jackson*, 1999, pp. 157–158], and the electric field due to the polarization will oppose and nearly cancel the radar electric field at the surface. The situation is similar to Rayleigh scattering from a solid metal sphere. A reconsideration of radar scattering by metal-coated ice grains is underway, and preliminary results indicate that suitably large scattering could be obtained if the ice grains were highly elongated, i.e., needle shaped. In this case the ice grains would behave like radar chaff and their sedimentation properties would be quite different from spherical grains so that much heavier grains could stay aloft much longer than spherical grains having the same mass. Details on this analysis will be provided elsewhere.

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