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Letter to the Editor

 Shotgun lipidomic analysis of chemically sulfated sterols compromises analytical sensitivity: Recommendation for large-scale global lipidome analysis

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Shotgun lipidomics affords comprehensive and quantitative analysis of lipid species in cells and tissues at high-throughput [1–5]. The methodology is based on direct infusion of lipid extracts by electrospray ionization (ESI) combined with tandem mass spectrometry (MS/MS) and/or high resolution Fourier transform mass spectrometry (FTMS) for identification and quantification of lipid species [6]. Shotgun lipidomics affords extensive lipidome coverage by combining the analysis of lipid extracts in positive and negative ion mode [1, 3]. Notably, sterols such as cholesterol and ergosterol exhibit low ionization efficiency in ESI [7]. For this reason, chemical derivatization procedures including acetylation [8] or sulfation [9] are commonly implemented to facilitate ionization, detection and quantification of sterols for global lipidome analysis [1–3, 10].

In the course of large-scale lipidomic analyses using an LTQ Orbitrap XL mass spectrometer (Thermo Scientific) equipped with a robotic nanoESI source TriVersa NanoMate (Advion Biosciences), we observed a pronounced decrease in the sensitivity of negative ion mode analysis following repeated injections of samples subjected to chemical sulfation. This decrease in sensitivity was observed only for negative ion mode analysis of lipid species in underivatized lipid extracts. No decrease in sensitivity was observed for negative ion mode analysis of chemically sulfated sterols or for positive ion mode analysis of lipid species in underivatized lipid extracts.

To investigate the decrease in analytical sensitivity in further detail we (i) spiked a yeast lysate with internal standards and executed two-step lipid extraction as previously described [3].

Abbreviations: ESI, electrospray ionization; FTMS, Fourier transform mass spectrometry; MS/MS, tandem mass spectrometry; PI, phosphatidylinositol

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(ii) show that changing polarity is a way to overcome this analytical problem; and (iii) recommend that analysis of chemically sulfated lipid extracts is scheduled at the end of all other analyses in order not to compromise sensitivity and data quality of large-scale global lipidome analyses.

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